

FRAUNHOFER INSTITUTE FOR SOLAR ENERGY SYSTEMS ISE

SOLAR ENERGY AND SYSTEMS RESEARCH

Markus Mundus

ULTRASHORT LASER PULSES FOR ELECTRICAL CHARACTERIZATION OF SOLAR CELLS



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SOLARE ENERGIE- UND SYSTEMFORSCHUNG / SOLAR ENERGY AND SYSTEMS RESEARCH

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Ultrashort Laser Pulses for Electrical Characterization of Solar Cells

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> Presented by Markus Mundus

> > at the



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To my father.

Contents

1	Inti	roduct	ion	1
2	Phy	sics a	nd Characterization of Solar Cells	7
	2.1	Semic	onductor and Solar Cell Basics	7
		2.1.1	Intrinsic and Extrinsic Semiconductors	8
		2.1.2	Charge Carrier Transport	11
			2.1.2.1 The Continuity Equation	11
			2.1.2.2 The Drift-Diffusion Approximation	11
		2.1.3	Charge Carrier Generation and Recombination	13
	2.2	Electr	ical Characterization and Calibration of Solar Cells	14
		2.2.1	Solar Cell Calibration	15
		2.2.2	Highly Accurate $I_{\rm SC}$ Measurement $\ldots \ldots \ldots$	16
			2.2.2.1 Broadband Approach and Spectral	
			$Mismatch \ . \ . \ . \ . \ . \ . \ . \ . \ . \ $	16
			2.2.2.2 Differential Spectral Responsivity Method	17
3	Ult	rashor	t Laser Pulses & Nonlinear Optics	23
	3.1	Basic	Principles	23
	3.2	Nonlii	near Optics	25
		3.2.1	From Linear to Nonlinear Optics	26
		3.2.2	Survey on Nonlinear Optical Effects	29
			3.2.2.1 Second Order Processes	29
			3.2.2.2 Nonlinear Refractive Index	32
			3.2.2.3 Multi-Photon Absorption	33
		3.2.3	Nonlinear Wave Equation	33
		3.2.4	Coupled Amplitude Equations and Phase-Matching	34
		3.2.5	Supercontinuum Generation	36
4	Inte	eractio	n of Ultrashort Laser Pulses and Solar Cells	41
-	4 1	Introd	luction	42

	4.2	Model	ling of Solar Cell Response Under Pulsed Irradiation	44
		4.2.1	$J_{\rm SC}$ in Terms of Excess Carrier Density $\ldots \ldots$	44
		4.2.2	Coupled Differential Equations for	
			Radiation-Semiconductor Interaction	45
		4.2.3	Reduction of the Set of Differential Equations	47
		4.2.4	Quasi-Steady-State Approximation	51
		4.2.5	Final Modeling Equations	55
	4.3	Exper	imental Study of TPA Impact on J_{SC}	56
		4.3.1	Linear Versus Nonlinear Carrier Generation	56
		4.3.2	Experimental Setup	57
			4.3.2.1 Laser Source	57
			4.3.2.2 Double-Ring-Resonator Setup	58
		4.3.3	Experimental Results	62
	4.4	Simula	ation of TPA Impact on $J_{\rm SC}$	63
		4.4.1	Validation of Simulation Approach	63
		4.4.2	Pulsed Versus Continuous Illumination	66
	4.5	Conclu	usions	67
5	Ten	nporal	Shaping of Ultrashort Laser Pulses	71
5	Ten 5.1	n poral Strate	Shaping of Ultrashort Laser Pulses gies for Temporal Shaping	71 71
5	Ten 5.1	n poral Strate 5.1.1	Shaping of Ultrashort Laser Pulses gies for Temporal Shaping	71 71 72
5	Ten 5.1	n poral Strate 5.1.1 5.1.2	Shaping of Ultrashort Laser Pulses gies for Temporal Shaping	7 1 71 72 73
5	Ten 5.1 5.2	nporal Strate 5.1.1 5.1.2 Impuls	Shaping of Ultrashort Laser Pulses gies for Temporal Shaping	71 71 72 73 75
5	Ten 5.1 5.2	nporal Strate 5.1.1 5.1.2 Impula 5.2.1	Shaping of Ultrashort Laser Pulses gies for Temporal Shaping	71 71 72 73 75 75
5	Ten 5.1 5.2	poral Strate 5.1.1 5.1.2 Impula 5.2.1 5.2.2	Shaping of Ultrashort Laser Pulses gies for Temporal Shaping	71 71 72 73 75 75 75
5	Ten 5.1 5.2	nporal Strate 5.1.1 5.1.2 Impula 5.2.1 5.2.2 5.2.3	Shaping of Ultrashort Laser Pulses gies for Temporal Shaping	71 71 72 73 75 75 78 79
5	Ten 5.1 5.2	poral Strate 5.1.1 5.1.2 Impula 5.2.1 5.2.2 5.2.3 5.2.4	Shaping of Ultrashort Laser Pulses gies for Temporal Shaping	71 71 72 73 75 75 75 78 79 81
5	Ten 5.1 5.2	poral Strate 5.1.1 5.1.2 Impula 5.2.1 5.2.2 5.2.3 5.2.4 Chara	Shaping of Ultrashort Laser Pulses gies for Temporal Shaping	71 71 72 73 75 75 75 78 79 81 85
5	Ten 5.1 5.2 5.3	poral Strate 5.1.1 5.1.2 Impula 5.2.1 5.2.2 5.2.3 5.2.4 Chara 5.3.1	Shaping of Ultrashort Laser Pulses gies for Temporal Shaping A Brief Overview on Temporal Shaping Approaches Temporal Shaping Strategy in This Work	71 71 72 73 75 75 75 78 79 81 85 85
5	Ten 5.1 5.2 5.3	poral Strate 5.1.1 5.1.2 Impuls 5.2.1 5.2.2 5.2.3 5.2.4 Chara 5.3.1	Shaping of Ultrashort Laser Pulsesgies for Temporal ShapingA Brief Overview on Temporal Shaping ApproachesTemporal Shaping Strategy in This WorkSe Response of Step-Index Multimode Optical FibersIntroduction to Step-Index Fiber OpticsMode Coupling in Optical FibersGloge's Time-Dependent Power Flow EquationNumerical Solution to the Power Flow Equationcterization of Multimode FibersEFPIR-Method5.3.1.1General Program Flow	71 71 72 73 75 75 75 78 79 81 85 85
5	Ten 5.1 5.2	poral Strate 5.1.1 5.1.2 Impula 5.2.1 5.2.2 5.2.3 5.2.4 Chara 5.3.1	Shaping of Ultrashort Laser Pulsesgies for Temporal ShapingA Brief Overview on Temporal Shaping ApproachesTemporal Shaping Strategy in This WorkSe Response of Step-Index Multimode Optical FibersIntroduction to Step-Index Fiber OpticsMode Coupling in Optical FibersGloge's Time-Dependent Power Flow EquationNumerical Solution to the Power Flow Equationcterization of Multimode Fibers5.3.1.1General Program Flow5.3.1.2Downhill Simplex Method	71 71 72 73 75 75 78 79 81 85 85 85
5	Ten 5.1 5.2	poral Strate 5.1.1 5.1.2 Impula 5.2.1 5.2.2 5.2.3 5.2.4 Chara 5.3.1	Shaping of Ultrashort Laser Pulsesgies for Temporal ShapingA Brief Overview on Temporal Shaping ApproachesTemporal Shaping Strategy in This WorkSe Response of Step-Index Multimode Optical FibersIntroduction to Step-Index Fiber OpticsMode Coupling in Optical FibersGloge's Time-Dependent Power Flow EquationNumerical Solution to the Power Flow Equationcterization of Multimode Fibers5.3.1.1General Program Flow5.3.1.2Downhill Simplex Method5.3.1.3Parametrization of Attenuation and	71 71 72 73 75 75 78 79 81 85 85 85
5	Ten 5.1 5.2	poral Strate 5.1.1 5.1.2 Impuls 5.2.1 5.2.2 5.2.3 5.2.4 Chara 5.3.1	Shaping of Ultrashort Laser Pulsesgies for Temporal ShapingA Brief Overview on Temporal Shaping ApproachesTemporal Shaping Strategy in This WorkSe Response of Step-Index Multimode Optical FibersIntroduction to Step-Index Fiber OpticsMode Coupling in Optical FibersGloge's Time-Dependent Power Flow EquationNumerical Solution to the Power Flow Equationcterization of Multimode Fibers5.3.1.1General Program Flow5.3.1.2Downhill Simplex Method5.3.1.3Parametrization of Attenuation and Diffusion	71 71 72 73 75 75 78 79 81 85 85 85 86 89
5	Ten 5.1 5.2 5.3	poral Strate 5.1.1 5.1.2 Impula 5.2.1 5.2.2 5.2.3 5.2.4 Chara 5.3.1	Shaping of Ultrashort Laser Pulses gies for Temporal Shaping A Brief Overview on Temporal Shaping Approaches Temporal Shaping Strategy in This Work se Response of Step-Index Multimode Optical Fibers Introduction to Step-Index Fiber Optics Mode Coupling in Optical Fibers Mode Coupling in Optical Fibers Gloge's Time-Dependent Power Flow Equation Numerical Solution to the Power Flow Equation cterization of Multimode Fibers 5.3.1.1 General Program Flow 5.3.1.2 Downhill Simplex Method 5.3.1.3 Parametrization of Attenuation and Diffusion Validation of EFPIR-Method	71 71 72 73 75 75 75 78 79 81 85 85 85 85 86 89

Contents

		5.4.1	Effects on Spatial Intensity Distribution	94
		5.4.2	Effects on Polarization and Coherence	96
	5.5	Fiber	Concept for Conditioning of Ultrashort Pulses	100
		5.5.1	General Concept	102
		5.5.2	Design Principles	103
6	DSI	R-Setu	p Based on Ultrashort Laser Pulses	107
	6.1	Introd	luction to DSR-Measurements	108
	6.2	Setup	Description and Characterization	109
		6.2.1	Radiation Sources	111
		6.2.2	Modes of Operation $\ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots$	113
		6.2.3	Spectral Shaping of Ultrashort Laser Pulses	114
		6.2.4	Spatio-Temporal Shaping of Laser Pulses	120
		6.2.5	Peripheral Equipment	125
	6.3	Optica	al Efficiency and Loss Analysis	126
	6.4	Meası	rement Uncertainty Analysis	128
		6.4.1	Optical Measurement Uncertainties	129
			6.4.1.1 Spectral Uncertainties	129
			6.4.1.2 Spatial Non-Uniformity	133
			6.4.1.3 Longitudinal Test Object Position	136
		6.4.2	Further Measurement Uncertainties	137
		6.4.3	Combined Measurement Uncertainty	139
		6.4.4	Uncertainty in Short Circuit Current	146
	6.5	Concl	usions & Comparison	148
		6.5.1	General Presentation of the New Setup	148
		6.5.2	Measurement Uncertainty of the New Setup	149
		6.5.3	Comparison to State-Of-The-Art Facilities	150
7	Sup	ercont	tinuum Radiation for Solar Cell	
	Cha	racter	rization	155
	7.1	Introd	luction	156
	7.2	Differ	ential Supercontinuum Measurement Approach	157
		7.2.1	Generation of Supercontinuum Radiation	158
		7.2.2	Spectral Shaping of Supercontinuum Radiation	159
		7.2.3	Prospects for Tackling the Lack of UV Radiation .	161

	7.3	 Spectral Performance of Shaped Supercontinuum 7.3.1 Spectral Mismatch and Uncertainty 7.3.2 Capabilities of Shaped Supercontinuum 7.3.2 Full Spectrum Applacie 	$162 \\ 162 \\ 164 \\ 167$
	7.4	Short Circuit Current Measurements	107 170
	7.5	Conclusions	171
8	$\mathbf{E}\mathbf{Q}$	E-Measurement of CPV Modules	173
	8.1	Challenges and State-Of-The-Art	174
	8.2	Experimental Approach	176
		8.2.1 Spectral, Spatial and Angular Requirements	176
		8.2.2 Setup Description	178
	8.3	Validation of Measurement Approach	183
		8.3.1 Device Under Test: CPV Mono Module	183
		8.3.2 Measurement Results	184
9	Sun	nmary & Outlook	187
De	eutso	che Zusammenfassung und Ausblick	197
\mathbf{A}	Anł	narmonic Oscillator Solution	209
в	Nor	nlinear Schrödinger Equation	213
\mathbf{C}	On	the Negligence of Light Trapping	217
D	Gre	en's Function Approach for Solving the	
_	Cor	ntinuity Equation	221
\mathbf{E}	Mo	nte Carlo Method for Measurement Uncertainty	225
Bi	bliog	graphy	231
Li	st of	Publications	247
Acknowledgments			251

Abbreviations & Symbols

Abbreviations

Air Mass
\mathbf{A} cousto- \mathbf{O} ptical \mathbf{M} odulator
Cumulative Distribution Function
Concentrator Photovoltaics
Circumsolar Ratio
Continuous \mathbf{W} ave
Difference-Frequency Generation
Device Under Test
D ifferential S pectral R esponsivity
\mathbf{E} xternal \mathbf{Q} uantum \mathbf{E} fficiency
Estimation of Fiber Properties from Impulse Responses \Box
$\mathbf{F} \text{inite } \mathbf{E} \text{lement } \mathbf{M} \text{ethod}$
Free Carrier Absorption
\mathbf{F} ull- \mathbf{W} idth-at- \mathbf{H} alf- \mathbf{M} aximum
Group Velocity Dispersion
${\bf G} uide$ to the Expression of Uncertainty in Measurement
Light Emitting Diode
Linearly Polarized
Monte Carlo
Numerical Aperture
Near-Infrared Region
\mathbf{O} ff-axis \mathbf{P} arabolic \mathbf{M} irror
\mathbf{O} ptical \mathbf{P} arametric \mathbf{A} mplification
Optical Parametric Oscillator
$\mathbf{P} \text{hotonic } \mathbf{C} \text{rystal } \mathbf{F} \text{iber}$

\mathbf{PDF}	P robability D ensity F unction
\mathbf{PV}	\mathbf{P} hoto \mathbf{v} oltaics
\mathbf{QSS}	\mathbf{Q} uasi- \mathbf{S} teady- \mathbf{S} tate
RC	Reference Cell
\mathbf{RMS}	\mathbf{R} oot- \mathbf{M} ean- \mathbf{S} quare Deviation
SFG	$\mathbf{S} um\text{-}\mathbf{F} requency \ \mathbf{G} eneration$
SHG	Second Harmonic Generation \mathbf{S}
\mathbf{SPM}	${\bf S} elf {\bf - P} hase \ {\bf M} odulation$
\mathbf{SR}	\mathbf{S} pectral R esponsivity
\mathbf{SRH}	\mathbf{S} hockley- \mathbf{R} ead- \mathbf{H} all
STC	Standard Test Conditions
TBP	$\mathbf{T}ime\textbf{-B}andwidth\textbf{-P}roduct$
\mathbf{TC}	Test Cell
THG	Third Harmonic Generation
\mathbf{TIR}	$\mathbf{T} otal \ \mathbf{I} n ternal \ \mathbf{R} effection$
Ti:Sa	\mathbf{Ti} tanium- \mathbf{Sa} pphire
TPA	$\mathbf{T} \text{wo-} \mathbf{P} \text{hoton } \mathbf{A} \text{bsorption}$
\mathbf{UV}	\mathbf{U} ltra- \mathbf{V} iolet
VIS	Vis ible Spectral Region

Physical Constants

Vacuum speed of light	c_0	=	$2.997 \ 925 \cdot 10^8 \ {\rm ms}^{-1}$
Planck constant	h	=	$6.626~070\cdot 10^{-34}~{\rm Js}$
Reduced Planck constant	\hbar	=	$h/2\pi = 1.054 572 \cdot 10^{-34} \text{ Js}$
Boltzmann constant	$k_{\rm B}$	=	$8.617 \ 330 \cdot 10^{-5} \ \mathrm{eVK}^{-1}$
Electron mass	m	=	$9.109 \ 383 \cdot 10^{-31} \ \mathrm{kg}$
Elementary charge	q	=	$1.602 \ 177 \cdot 10^{-19} \ \mathrm{C}$
Vacuum permittivity	ε_0	=	$8.854 \ 188 \cdot 10^{-12} \ \mathrm{Fm}^{-1}$
Vacuum permeability	μ_0	=	$1.256 \ 637 \cdot 10^{-6} \ \mathrm{Vs(Am)}^{-1}$

Roman Symbols

Ã	Modal fiber attenuation in matrix form	1
A	Solar cell area	cm^{-2}
$ ilde{A}_{\mathbf{k},\mathbf{k}}$	Attenuation coefficients in $\tilde{\mathbf{A}}$	1
a	Core radius of an optical fiber	m
a_{n}	Strength of n-th order nonlinearity	$m^{-(n-1)}s^{-2}$
	in anharmonic oscillator	
В	Magnetic flux density	$\rm V sm^{-2}$
b_{n}	Electron mobility	$\mathrm{cm}^{2}\mathrm{V}^{-1}\mathrm{s}^{-1}$
$b_{ m p}$	Hole mobility	$\mathrm{cm}^{2}\mathrm{V}^{-1}\mathrm{s}^{-1}$
C	Speckle contrast	1
c_{TBP}	Time-bandwidth-product constant	1
D	Electric flux density	Cm^{-2}
D	Diffusion coefficient	$\mathrm{m}^2\mathrm{s}^{-1}$
$ ilde{ ext{D}}$	Modal dispersion in matrix form	1
$D_{\mathrm{C,V}}$	Conduction and valence band	$eV^{-1}cm^{-3}$
	density of states	
D_{a}	Ambipolar diffusion coefficient	$\mathrm{m}^{2}\mathrm{s}^{-1}$
$D_{\rm cl}$	Correlation length of waveguide	m
	disturbances	
$ ilde{D}_{\mathrm{k,k}}$	Modal dispersion coefficients in $\tilde{\mathbf{D}}$	1
D_{n}	Electron diffusion coefficient	$\mathrm{m}^{2}\mathrm{s}^{-1}$
D_{p}	Hole diffusion coefficient	$\mathrm{m}^{2}\mathrm{s}^{-1}$
D_{λ}	Group velocity dispersion parameter	$\rm pskm^{-1}nm^{-1}$
d	Modal diffusion coefficient	rad^2m^{-1}
d'	First derivative of modal diffusion	$radm^{-1}$
	coefficient after θ	
$d_{\rm eff}$	Effective nonlinearity	pmV^{-1}
\mathbf{E}	Electric field	Vm^{-1}
$E_{\rm C}$	Conduction band edge energy	eV
$E_{\rm F}$	Fermi level	eV

$E_{\rm F,n}$	Electron quasi-Fermi level	eV
$E_{\rm F,p}$	Hole quasi-Fermi level	eV
$E_{\rm g}$	Band gap energy	eV
$E_{\rm ph}$	Photon energy	eV or J
$E_{\rm Sim}$	Spectral irradiance of solar simulator	$\rm Wnm^{-1}cm^{-2}$
$E_{\rm STC}$	Spectral irradiance of STC	$\rm Wnm^{-1}cm^{-2}$
$E_{\rm V}$	Valence band edge energy	eV
E_{λ}	Spectral irradiance	$\rm Wnm^{-1}cm^{-2}$
F_{restore}	Restoring force in Lorentz model	Ν
f	Fermi-Dirac distribution	1
$f_{\rm rep}$	Pulse repetition rate	s^{-1}
$f_{\rm x}$	Measurement correction term	1
G	Generation rate	$\mathrm{s}^{-1}\mathrm{m}^{-3}$
Η	Magnetic field	Am^{-1}
Н	Fiber impulse response	W
Ι	Optical intensity	Wm^{-2}
$I_{\rm av}$	Average optical intensity	Wm^{-2}
$I_{\rm peak}$	Peak optical intensity	Wm^{-2}
$I_{\rm SC}$	Short circuit current	mA
$I_{ m SC}^{ m STC}$	Short circuit current at STC	mA
$J_{\rm SC}$	Short circuit current density	$\rm mAcm^{-2}$
j	Current density	Am^{-2}
j	Measured current value	А
k	Wave number	m^{-1}
	(also natural number in EFPIR)	(1)
$k_{\rm cov}$	Coverage factor	1
k_2	Group velocity dispersion	s^2m^{-1}
L	Fiber length	m
l	Azimuthal mode number	1
M	Total number of fiber modes	1
	(and Monte Carlo draws)	(1)
MM	Spectral mismatch factor	1

Abbreviations & Symbols

m	Radial mode number	1
$m_{\rm n,p}^*$	Electron/hole effective mass	kg
N	Electron density in Lorentz model	${\rm cm}^{-3}$
	(also natural number)	(1)
N'	Total number of discretized angles	1
	in EFPIR	
$N_{\rm A}$	Acceptor doping density	cm^{-3}
$N_{\rm C}$	Effective conduction band density of states	cm^{-3}
$N_{\rm D}$	Donor doping density	cm^{-3}
$N_{\rm sol}$	Soliton order number	1
$N_{\rm V}$	Effective valence band density of states	cm^{-3}
N_0	Maximum excess carrier density	cm^{-3}
n	Electron density	cm^{-3}
	(also refractive index)	(1)
$n_{\rm co}$	Core refractive index of optical fiber	1
$n_{\rm cl}$	Cladding refractive index of optical fiber	1
$n_{\rm i}$	Intrinsic carrier density	cm^{-3}
n_0	Linear refractive index	1
n_2	Nonlinear refractive index	$\mathrm{m}^{2}\mathrm{W}^{-1}$
Δn	Excess carrier density	m^{-3}
Р	Polarization density	Cm^{-2}
Р	Modal power in time domain	W
p	Hole density	cm^{-3}
	(also modal power in frequency domain)	(J)
р	Modal power in matrix form	W
Q	Ratio of two current measurements	1
q	Mode number	1
	(also elementary charge)	(C)
R	Recombination rate	$\rm s^{-1} cm^{-3}$
r	Electron displacement in Lorentz model	m
$r_{\rm corr}$	Correlation coefficient	1
$S_{ m i}$	Stokes parameters $(i = 1, 2, 3)$	1

S_{0}	Front surface recombination velocity	ms^{-1}
s_0	Spectral responsivity	$AW^{-1}m^2$
\widetilde{S}	Differential spectral responsivity	$AW^{-1}m^2$
T	Pulse period	S
$T_{\rm trans}$	Transition rate	s^{-1}
\mathcal{T}	Temperature	Κ
$t_{\rm fac}$	Duty cycle	1
U	Measurement uncertainty	%
	(also potential energy)	(J)
u	Standard measurement uncertainty	%
$V_{\rm Stokes}$	Degree of polarization	1
v_g	Group velocity	ms^{-1}
W	Fiber properties in matrix form	1
W	Device or wafer thickness	m
w	Total number of spatial steps in EFPIR	1

Greek Symbols

α	Material absorption coefficient	cm^{-1}
$lpha_{ m bb}$	Linear band-to-band absorption coefficient	cm^{-1}
$\alpha_{\rm FCA}$	Free-carrier absorption coefficient	cm^{-1}
β	Two-photon band-to-band absorption	cmW^{-1}
	coefficient	
$\beta_{ m lm}$	Propagation constant of lm -th fiber mode	m^{-1}
γ	Damping constant	s^{-1}
	(also fiber attenuation)	(m^{-1})
δ	Dirac-delta	1
$\delta lpha_{ m FCA}$	Change in FCA by single ultrashort pulse	m^{-1}
ε	Permittivity	Fm^{-1}
ε_r	Relative permittivity	Fm^{-1}
$\eta_{ m opt}$	Optical efficiency of concentrator optics	1
η_n	Electrochemical potential of electrons	eV

θ	Propagation angle with respect to	mrad
	fiber axis	
$ heta_c$	Critical angle	mrad
κ	Perturbation strength	m^{-1}
λ	Wavelength of radiation	nm
λ_0	Center wavelength of radiation	nm
μ	Permeability	$VsA^{-1}m^{-1}$
μ_0	Chemical potential	eV
ν	Frequency of radiation	s^{-1}
$ u_{ m f}$	Degree of freedom	1
$\nu_{ m f,eff}$	Effective degree of freedom	1
$\sigma_{ m n}$	Electron conductivity	$\mathrm{AV}^{-1}\mathrm{cm}^{-1}$
σ_{lpha}	Linear band-to-band absorption	cm^2
	cross section	
σ_{eta}	Two-photon band-to-band absorption	$\mathrm{cm}^{4}\mathrm{W}^{-1}$
	cross section	
$\sigma_{ m FCA}$	Free carrier absorption cross section	cm^2
τ	Minority carrier lifetime	S
$ au_{1/\mathrm{e}^2}$	$1/e^2$ pulse duration	S
$ au_{ m FWHM}$	FWHM pulse duration	S
$ au_{ m p}$	Pulse transit time	S
$ au_{\mathrm{trans}}$	Excess carrier transit time	S
Φ	Photon flux	$\mathrm{s}^{-1}\mathrm{cm}^{-2}$
φ	Phase	1
	(also electrical potential)	(V)
$\chi^{(n)}$	n-th order susceptibility	$(mV^{-1})^{n-1}$
ω	Angular frequency	$rads^{-1}$
ω_0	Natural frequency	$rads^{-1}$

Chapter 1

Introduction

Photovoltaics and the Role of Accurate Measurements

Photovoltaics (PV) has evolved to a cost competitive power production tool in the last decade. Initially being largely driven by the idea of clean and sustainable electricity production, surcharges and feed-in tariffs have fostered a tremendous deployment of PV systems. Owing to the economies of scale and technological advances this resulted in a PV system price decline of about 75% within less than a decade [1]. In fact, for the last 34 years the PV learning curve is closely following a trend of 20% price decrease with each doubling of cumulative production capacity [2]. This development has been and is still facilitating grid parity of PV in an increasing number of markets and sites [3–5], thereby stoking the expectation of further growth in PV. Promising indicators thereof are recent numbers of 40 GWp global PV deployment in 2014 (equivalent to an annual increase of 28%) and forecasts for 2015 in the range of 51 GWp [1]. Long-term forecasts predict a global PV capacity of even more than 4,500 GWp in 2050, being equivalent to 16% of global electricity production from PV [6]. Thus, the success story of PV has most likely just begun.

As stated above, not only economic but also technological advances are supporting the further growth of PV. E.g. researchers are successfully striving for technological solar cell improvements, thereby beating solar



Figure 1.1: Evolution of solar cell efficiency for various technologies from the 1970s to the present. Adapted from [7] as of August 2015.

cell efficiency records on a quite regular basis (see Fig. 1.1, adapted from [7]). Moreover, new materials (e.g. perovskite, see also Fig. 1.1) as well as solar cell concepts (e.g. heterojunction and/or back-contact cells [8]) emerge and demonstrate the enormous potential for further technological advances in PV.

The described progresses in PV are steadily accompanied by a tremendous effort in the development of highly sophisticated measurement methods and facilities. In particular, there is a major interest in highly accurate measurements that satisfy the rising economic importance of PV and enable assessment of its technological advances.

From a technological point of view, measurements at lowest uncertainties are compulsory for a reliable evaluation of new developments and their potential. Especially in the light of generally decreasing relative improvements as certain technologies approach their limit, highly accurate measurements are desperately required.

1 Introduction

From an economic point of view, lowest possible measurement uncertainties are motivated by the direct relation of investment and measurement uncertainty. A typical uncertainty of 2% in solar cell maximum power accumulates to an annual investment uncertainty of 6 Million \in for a mono-type silicon solar cell manufacturer producing 1 GWp per year.¹ On a global scale, this accumulates to an investment uncertainty of approximately 900 Million \in in 2014.² Consequently, improving measurement uncertainties by a factor of two would yield a reduction of investment uncertainty by 3 Million \in for the assumed cell manufacturer and more than 1,100 Million \in on a global scale.

Subject of This Work

The most relevant contribution to the uncertainty in production lines is given by the short circuit current $(I_{\rm SC})$ uncertainty of the applied reference solar cells [10–12]. These reference cells are typically calibrated by certified labs like the Fraunhofer ISE CalLab that are permanently aiming at lower uncertainties using several approaches for measuring the $I_{\rm SC}$. Besides integral outdoor and indoor calibration methods using the solar irradiance (outdoor) or an artificial reconstruction of the solar irradiance (indoor) [13], the differential spectral responsivity (DSR) method [14] has been proven to provide versatility, reliability and lowest uncertainties in determining the $I_{\rm SC}$.

In the DSR-method, the spectral responsivity $(s(\lambda))$ of the test cell is measured by chopped monochromatic light and the $I_{\rm SC}$ is determined from integrating the product of $s(\lambda)$ and the considered standard solar spectrum. A further reduction of measurement uncertainties in the DSRmethod is often impeded by the limited spectral power of conventionally

¹Assuming a mono-type silicon solar cell price of $0.3 \in Wp$ [9].

²Assuming an uncertainty of 2.6% in the maximum power of PV modules [10], 40 GWp PV power installation and $2.13 \in /Wp$ PV system price. The price per Wp in this example is obtained from a weighted average of PV system prices in China, Japan, the United Kingdom, Germany, France and Australia for residential, commercial and utility-scale systems in 2013 and is based on data from [1, 6]. In order to account for balance of system costs, 40% of the resulting total PV system price is considered for calculating the economic impact of measurement uncertainty.

applied spectrally filtered white light sources. Therefore, there is a rising interest in application of monochromatic light sources. As one promising monochromatic radiation source, spectrally tunable ultrashort pulse laser systems were proposed [15, 16] and have been recently applied for a further $I_{\rm SC}$ measurement uncertainty reduction [17].

This work aims at the development of a highly accurate DSR-facilty based on ultrashort laser pulses at the Fraunhofer ISE CalLab. The new setup seeks to outperform existing facilities in terms of $I_{\rm SC}$ measurement uncertainty, thereby significantly contributing to an improved calibration chain for research and industry. Furthermore, the work aims for a detailed investigation of the interaction of ultrashort laser pulses and solar cells under short circuit conditions. This analysis is supposed to give insight into potentially occurring additional uncertainties and allows for a general assessment of applicability of ultrashort laser pulses in highly accurate solar cell measurements. Moreover, new measurement methods beyond standard applications are to be investigated that become possible owing to the special features of the ultrashort laser pulses and the new setup.

Outline of This Work

This thesis is organized in nine chapters.

In this **Chapter 1** the necessity of the developments presented in this thesis is motivated, followed by a distinct description of the work's subject and this outline of the thesis.

In Chapter 2 and Chapter 3 some general theoretical background on semiconductors and solar cells as well as ultrashort laser pulses and nonlinear optics is reviewed. Although the subsequent chapters of this thesis are written as comprehensible and self-contained as possible, some of the described and evaluated effects as well as some of the terminology used in the remainder of this thesis will be clarified initially in Chapters 2 and 3.

In Chapter 2 basic semiconductor physics is reviewed including intrinsic and extrinsic semiconductors as well as charge carrier transport, generation and recombination. Regarding solar cell physics a special focus lies on short circuit conditions, the cell condition of major importance for this thesis.

In Chapter 3 the basic principle of ultrashort laser pulses is briefly described. Furthermore, the topic of nonlinear optics is introduced and a review of those major nonlinear optical effects is given that are applied in the remainder of this thesis (second order processes and phase matching, multi-photon absorption and supercontinuum generation).

In Chapter 4 the interaction of ultrashort laser pulses and solar cells under short circuit conditions is addressed. Starting from the development of a general theoretical model describing the pulse-cell interaction, experimental studies are presented that make the impact of nonlinear contributions to this interaction evident. Based on these findings the theoretical model is validated and applied for finding major conclusions regarding feasibility of ultrashort laser pulses in highly accurate solar cell measurements.

In Chapter 5 temporal shaping of ultrashort laser pulses by fiberoptical components is regarded. After a short discussion of prospects for temporal shaping of such pulses, the theoretical background of multimode fiber impulse responses is given. Based on this knowledge a new characterization method for step-index multimode fiber properties is introduced. Furthermore, some additional effects induced by multimode fibers on propagating radiation are experimentally demonstrated. Finally, a monolithic fiber-based device is presented that allows for efficient, robust and reproducible pulse-to-cw-conversion of ultrashort laser pulses.

In Chapter 6 the new ultrashort pulse laser-based facility for DSRmeasurements is described. Starting from a general description of the applied laser system and some technical details, the spectral shaping components are introduced including a purpose-made prism monochromator. Afterwards, the designed optical setup is presented that serves for highly uniform irradiation of the measurement plane. An analysis of the setup's optical efficiency demonstrates its capabilities as compared to conventionally applied DSR-systems. Subsequently, the measurement uncertainty of the new setup is discussed in detail including a consideration of each particular contribution and an uncertainty propagation to obtain the combined measurement uncertainty.

In Chapter 7 a new method for fast and accurate short circuit current measurements based on chopped and spectrally shaped supercontinuum radiation is presented. Prior to a description of this novel measurement approach, the general advantages of supercontinuum radiation as compared to incoherent white light sources are discussed. Afterwards, the spectral performance of supercontinuum radiation is investigated based on simulations assessing the spectral mismatch and its uncertainty in comparison to state-of-the-art solar simulators. Finally, first measurement results are presented demonstrating applicability, speed and accuracy of the new method.

In Chapter 8 first external quantum efficiency measurements of a concentrator photovoltaic module are presented. After reviewing the challenges associated with such measurements, the developed experimental approach and the experimental setup for conducting those is introduced. First measurement results given thereafter represent a proof of principle and, thus, applicability of the new method for electro-optical characterization of concentrator modules.

In **Chapter 9** the thesis is concisely summarized and an outlook regarding pending work and future developments is given.

Chapter 2

Physics and Characterization of Solar Cells

In the first part of this chapter the theoretical background on semiconductors and solar cells is given that is essential for the remainder of this thesis. The discussions in this part of the chapter are based on textbooks by Sze [18] and Würfel [19]. In the second part of this chapter some general aspects on characterization and calibration of solar cells are given. The literature references being used are particularly noted in the respective passages.

2.1 Semiconductor and Solar Cell Basics

Electrons in isolated atoms exhibit distinct energy levels. If a closely spaced group of periodically arranged atoms is considered, their mutual interaction causes splitting of the distinct energy levels and essentially continuous energy bands are formed. The fully occupied band of highest energy is called *valence band* (with energy $E_{\rm V}$ as highest energy in this band), the next higher one is termed *conduction band* (with energy $E_{\rm C}$ as lowest energy in this band). Depending on the properties and distances of the atoms, the valence and conduction band might be overlapping or are

separated by a forbidden energy band gap $E_{\rm g}$. If this band gap is such high that it cannot be transcended by thermally or electrically excited electrons, the material is considered an insulator and is non-conducting. On the contrary, materials (like metals) are conducting if the bands overlap or if the conduction band is partly filled. In semiconductors the conduction band is empty at absolute zero temperature, but $E_{\rm g}$ is small enough so that an electron can be thermally excited to the conduction band leaving a hole behind in the valence band (e.g. $E_{\rm g} = 1.12$ eV in silicon at room temperature).

2.1.1 Intrinsic and Extrinsic Semiconductors

The integrated charge carrier densities in intrinsic semiconductors are given by

$$n = \int_{E_{\rm C}}^{\infty} f(E) D_{\rm C}(E) dE \qquad (2.1a)$$

$$p = \int_{-\infty}^{E_{\rm V}} [1 - f(E)] D_{\rm V}(E) dE \qquad (2.1b)$$

for electrons n and holes p with conduction and valence band density of states $D_{C,V}(E)$ and Fermi-Dirac distribution f(E). The conduction and valence band density of states, which is the density of allowed energy states per unit volume, is given by

$$D_{\rm C}(E) = 4\pi \left(\frac{2m_{\rm n}^*}{h^2}\right)^{3/2} \left(E - E_{\rm C}\right)^{1/2}$$
(2.2a)

$$D_{\rm V}(E) = 4\pi \left(\frac{2m_{\rm p}^*}{h^2}\right)^{3/2} (E_{\rm V} - E)^{1/2}$$
 (2.2b)

with Planck constant h and $m_{n,p}^*$ as effective mass of the electrons and holes, respectively.

2.1 Semiconductor and Solar Cell Basics

The Fermi-Dirac distribution reads

$$f(E) = \frac{1}{\exp\left(\frac{E - E_{\rm F}}{k_{\rm B}\mathcal{T}}\right) + 1}$$
(2.3)

with Boltzmann constant $k_{\rm B}$, temperature \mathcal{T} and Fermi level $E_{\rm F}$ characterizing that energy level $E = E_{\rm F}$ at which half of the states are occupied.

For $E - E_{\rm F} > 3k_{\rm B}\mathcal{T}$ Eq. (2.3) can be approximated by the Boltzmann distribution reading $f(E) \approx \exp\left[-\left(E - E_{\rm F}\right)/k_{\rm B}\mathcal{T}\right]$ and the electron density in the conduction band is solved to be

$$n = N_{\rm C} \exp\left(\frac{E_{\rm F} - E_{\rm C}}{k_{\rm B}\mathcal{T}}\right) \tag{2.4}$$

with conduction band effective density of states

$$N_{\rm C} = 2 \left(\frac{2\pi m_{\rm n}^* k_{\rm B} \mathcal{T}}{h^2}\right)^{3/2}.$$
 (2.5)

Similarly, the hole density in the valence band is given by

$$p = N_{\rm V} \exp\left(\frac{E_{\rm V} - E_{\rm F}}{k_{\rm B}\mathcal{T}}\right)$$
(2.6)

with valence band effective density of states

$$N_{\rm V} = 2 \left(\frac{2\pi m_{\rm p}^* k_{\rm B} \mathcal{T}}{h^2}\right)^{3/2}.$$
 (2.7)

The number of electrons and holes is identical in intrinsic semiconductors at thermal equilibrium and an *intrinsic carrier density*

$$n_{\rm i}^2 = np = N_{\rm C} N_{\rm V} \exp\left(-\frac{E_{\rm g}}{k_{\rm B} \mathcal{T}}\right) \tag{2.8}$$

with $E_{\rm g} = E_{\rm C} - E_{\rm V}$ is defined.

The identity n = p becomes invalid in case of extrinsic semiconductors that are created by adding specific doping atoms. These doping atoms establish shallow electronic states in the band gap that are either located close to the conduction band (by adding *donor* atoms) or close to the valence band (by adding *acceptor* atoms). In both cases these electronic states are (virtually) completely ionized at room temperature, thereby enhancing conductivity of the semiconductor.

If donor atoms of density $N_{\rm D}$ are added, a so-called *n*-type semiconductor with $n \approx N_{\rm D}$ and $p \approx n_{\rm i}^2/N_{\rm D}$ is obtained. As $n \gg p$ in *n*-type material, electrons are called majority carriers, whereas holes are called minority carriers. The opposite holds in a *p*-type semiconductor with $p \approx N_{\rm A}$ and $n \approx n_{\rm i}^2/N_{\rm A}$ resulting from added acceptor atoms of density $N_{\rm A}$. Adjusting the Fermi level according to

$$E_{\rm F} = \begin{cases} E_{\rm C} - k_{\rm B} \mathcal{T} \ln \left(N_{\rm C} / N_{\rm D} \right) \\ E_{\rm V} + k_{\rm B} \mathcal{T} \ln \left(N_{\rm V} / N_{\rm A} \right) \end{cases}$$
(2.9)

for n- or p-type material the Fermi-Dirac distribution given in Eq. (2.3) can still be applied for calculation of carrier distributions.

However, in case of additional generation of electrons and holes (so called excess charge carriers Δn and Δp) by e.g. incident photons, the Fermi statistic with a single Fermi energy does not suffice for describing both, electron and hole distributions. Therefore, so-called quasi-Fermi levels $E_{\rm F,n}$ and $E_{\rm F,p}$ are introduced that allow computation of both electron and hole concentrations with

$$n = N_{\rm C} \exp\left(\frac{E_{\rm F,n} - E_{\rm C}}{k_{\rm B}T}\right),\tag{2.10a}$$

$$p = N_{\rm V} \exp\left(\frac{E_{\rm V} - E_{\rm F,p}}{k_{\rm B}T}\right).$$
(2.10b)

Multiplication of Eqs. (2.10a) and (2.10b) gives

$$np = N_{\rm C} N_{\rm V} \exp\left(\frac{E_{\rm C} - E_{\rm V}}{k_{\rm B} \mathcal{T}}\right) = n_{\rm i}^2 \exp\left(\frac{E_{\rm F,n} - E_{\rm F,p}}{k_{\rm B} \mathcal{T}}\right)$$
(2.11)

with quasi-Fermi level splitting $E_{\rm F,n} - E_{\rm F,p}$ that represents the maximum usable energy of photons absorbed by a semiconductor.

2.1.2 Charge Carrier Transport

2.1.2.1 The Continuity Equation

Charge carrier or particle motion due to concentration gradients is governed by the continuity equation. In presence of carrier generation G(z,t)and recombination rates R(z,t) the one-dimensional representation of the continuity equation¹ for excess charge carriers is given by²

$$\frac{\partial \Delta n\left(z,t\right)}{\partial t} = D_{\rm a} \frac{\partial^2 \Delta n\left(z,t\right)}{\partial z^2} + G\left(z,t\right) - R\left(z,t\right). \tag{2.12}$$

The equation describes motion of excess charge carriers over time $(\partial \Delta n/\partial t)$ in presence of generation and recombination rates that are detailed in Section 2.1.3. Spatial gradients of excess carrier concentrations $(\partial^2 \Delta n/\partial z^2)$ result in a preferred direction of the diffusion-driven statistical spatial motion of excess charge carriers. The strength of this motion scales with the ambipolar diffusion coefficient D_a that takes into account electrical field effects caused by different mobilities of electrons and holes.

In Chapter 4 of this thesis the continuity equation is applied for describing the spatio-temporal interaction of ultrashort laser pulses and excess charge carriers in semiconductors.

2.1.2.2 The Drift-Diffusion Approximation

The charge carrier flow, thus, the current flow in a semiconductor might be described by the drift-diffusion model according to^3

$$j_{\rm n} = \frac{\sigma_{\rm n}}{q} \nabla \eta_{\rm n} = \frac{\sigma_{\rm n}}{q} \nabla \left(\mu_{\rm n} - q\varphi\right) \tag{2.13}$$

¹For validity of this one-dimensional representation lateral homogeneity of the semiconductor needs to be assumed.

²Hereafter, charge neutrality ($\Delta n = \Delta p$) is assumed, which is a reasonable assumption outside of the space charge region of a *pn*-junction [20, p. 18].

³This discussion is restricted to electrons. In a similar manner, the equations can be applied to holes as well.



Figure 2.1: Schematic illustration of an illuminated pn-junction under short circuit conditions. Adapted from [21].

with conductivity σ_n , elementary charge q and electrochemical potential η_n . The electrochemical potential is the sum of chemical potential μ_n and electrical energy $q\varphi$ and is equivalent to the conduction band quasi-Fermilevel $E_{\rm F,n}$. In Fig. 2.1 an illuminated *pn*-junction under short circuit conditions is shown (adapted from [21]). The gradient of $E_{\rm F,n}$ causes a strong electron flow towards the *n*-side contact, whereas the gradient in $E_{\rm F,p}$ causes the holes to flow towards the *p*-side contact. In spite of this strong current flow, no energy is extracted as there is no potential difference between the *n*- and *p*-side contact for electrons or holes.

The electrical potential in Eq. (2.13) is rather the consequence of diffusion-driven balancing of charge carrier concentrations (driven by chemical potential gradients) than the cause of a measurable current [19]. Thus, the electrical potential is neglected in the subsequent derivation of a short circuit current that is being expressed in terms of carrier concentration gradients. With this, Eq. (2.13) becomes

$$j_{\rm n} = \frac{\sigma_{\rm n}}{q} \nabla \mu_{\rm n}. \tag{2.14}$$

Expressing the conductivity in terms of mobility $(\sigma_n = qnb_n)$ and applying the Einstein relation

$$\frac{b_{\rm n}}{D_{\rm n}} = \frac{q}{k_{\rm B}\mathcal{T}} \tag{2.15}$$

results in $j_n = qnD_n \nabla \mu_n / k_B \mathcal{T}$. With the gradient of the chemical potential of electrons

$$\nabla \mu_{\rm n} = k_{\rm B} \mathcal{T} \nabla \ln \left(n / N_{\rm C} \right) \tag{2.16}$$

and

$$\nabla \ln\left(n/N_{\rm C}\right) = \frac{\nabla n}{n} \tag{2.17}$$

the diffusion current finally reads

$$j_{\rm n} = q D_{\rm n} \nabla n, \qquad (2.18)$$

which is the usual representation of Fick's law of diffusion. In Chapter 4 of this thesis, Eq. (2.18) is applied for calculating relative short circuit current densities being extracted at the *pn*-junction of a solar cell.

2.1.3 Charge Carrier Generation and Recombination

As mentioned above, excess charge carriers Δn can be excited by illuminating a semiconductor with photons. This process is governed by the generation rate that has already been appearing in Eq. (2.12) above. Incident photons of flux Φ generate excess charge carriers in the semiconductor material according to its wavelength-dependent band-to-band absorption coefficient α (λ).⁴ With initial photon flux Φ_0 the exponential excess carrier generation profile reads

$$G(z,\lambda) = \alpha(\lambda) \Phi_0 \exp(-\alpha(\lambda) z). \qquad (2.19)$$

Once excess charge carriers are excited, they thermalize to the band edges of conduction and valence band by inelastic scattering processes with the semiconductor lattice, thereby dissipating heat into the material on a picosecond timescale. This relaxation is followed by various recombination processes that are combined in the recombination rate

$$R = \frac{\Delta n}{\tau} \tag{2.20}$$

⁴This absorption model is extended in Chapter 4.

with minority carrier lifetime τ . The recombination processes governed by Eq. (2.20) are radiative, Auger and Shockley-Read-Hall (SRH) recombination in the material's bulk. The minority carrier lifetime is a characteristic value of the material properties and might depend on the excess carrier density, also termed injection level. E.g. an increasing carrier injection level might reduce recombination via defects (SRH-recombination), whereas it gives rise to radiative or Auger recombination. Thus, τ is typically expressed as τ (Δn).

In the remainder of this work, the individual bulk recombination processes are not further addressed. Apart from a brief discussion of injection dependence of minority carrier lifetimes in Chapter 4 and application of a parametrization of intrinsic recombination given in [22], bulk recombination is not a subject of this work. Therefore, a more detailed discussion of the individual recombination processes is omitted in this chapter.

In addition to recombination in the material's volume, recombination at the semiconductor surfaces can occur, which is rather described by a surface recombination velocity (SRV) according to

$$R_{\rm surf} = S\Delta n. \tag{2.21}$$

In the course of this thesis, surface recombination is applied to the theoretical model developed in Chapter 4 in order to imitate short circuit conditions by strong carrier extraction at a solar cell's pn-junction.

2.2 Electrical Characterization and Calibration of Solar Cells

The electrical performance of solar cells is determined by several characteristic parameters that are short circuit current $(I_{\rm SC})$, open-circuit voltage $(V_{\rm OC})$, fill factor (FF), power at maximum power point $(P_{\rm MPP})$ and efficiency (η) . In general, the factor of major interest in solar cell research and industry is $P_{\rm MPP}$ the maximum power output of a solar cell or, equivalently, the solar cell efficiency η . However, regarding lowest possible uncertainties in measuring $P_{\rm MPP}$ and η in production lines the $I_{\rm SC}$ becomes most relevant, as it is the main contributor to those uncertainties (see Chapter 1 and [10–12]). Thus, from a calibration point of view, the $I_{\rm SC}$ is that photovoltaic parameter of major interest and calibration labs are striving for further reductions of $I_{\rm SC}$ uncertainties.

As this thesis aims for a significant improvement of the calibration chain by reducing the uncertainty of the $I_{\rm SC}$ in the Fraunhofer ISE CalLab, discussions in this thesis are restricted to the $I_{\rm SC}$. Consequently, the subsequent discussion of electrical characterization and calibration of solar cells is as well restricted to the $I_{\rm SC}$.

2.2.1 Solar Cell Calibration

Traceable and comparable measurements of the $I_{\rm SC}$ of solar cells initially require a distinct determination of measurement conditions. For solar cells in non-concentrating terrestrial applications the so-called *Standard Test Conditions* (STC) were introduced that define⁵

- a spectral distribution according to AM1.5g,
- an irradiance of 1000 W/m^2 and
- a cell temperature of 25° C.

The AM1.5g spectral distribution given in [23] is a simulated spectrum that represents the sun's spectral distribution after propagation through 1.5 times the atmosphere's *air mass* (AM) under certain atmospheric conditions (CO₂ level, ozone content etc.). The "g" stands for global and means that direct and diffuse spectral contributions are considered. An air mass of 1.5 is equivalent to a solar zenith angle of approximately 48.2° and represents conditions in temperate latitudes.

Calibrated measurements of solar cells in non-concentrating terrestrial applications seek to fulfill STC in order to provide traceability, comparability and reproducibility. For lowest possible uncertainties in solar cell production lines, industrial-type reference cells are applied featuring a calibrated $I_{\rm SC}$ and an assigned uncertainty. The traceable transfer from

 $^{^5\}mathrm{Please}$ note that these STC also account for measurements of the other previously introduced solar cell parameters.
primary standards to these industrial-type reference cells is typically provided by several intermediate steps. Based on different initial standards, metrology institutes perform primary calibrations of particular reference solar cells of approximately $2x2 \text{ cm}^2$ area with expanded uncertainties < 0.7%.⁶ These cells are then applied as references in certified labs like the Fraunhofer ISE CalLab for the calibration of industrial solar cells at expanded uncertainties of typically < 2% [16].

2.2.2 Highly Accurate $I_{\rm SC}$ Measurement

2.2.2.1 Broadband Approach and Spectral Mismatch

For highly accurate measurements of the $I_{\rm SC}$ the previously introduced STC have to be fulfilled as good as possible. The most direct approach to fulfill STC is represented by measuring the current response of a temperature-controlled solar cell under short circuit conditions to the solar irradiation. Choosing the correct time and atmospheric conditions the $I_{\rm SC}$ under STC is obtained ($I_{\rm SC}^{\rm STC}$). However, as perfect spectral conditions for such outside measurements are rare, only a few days of a year allow for highly accurate measurements. On all other days, the spectral distribution differs from the AM1.5g, thereby enhancing measurement uncertainties.

These spectral variations are (virtually) overcome by indoor measurements with solar simulators that replicate the solar spectral distribution by spectrally filtered white light sources or a combination of various lightemitting diodes (LEDs). Their major advantage is represented by the continuous availability of a sun-like spectrum independent of day-time, season and weather. However, as yet no solar simulator exhibits a perfect spectrum, the measured $I_{\rm SC}$ of the test solar cell is not $I_{\rm SC}^{\rm STC}$, but rather the short circuit current obtained with the simulator spectrum ($I_{\rm SC}^{\rm Sim}$). In order to retrieve the $I_{\rm SC}^{\rm STC}$ from solar simulator measurements, a spectral mismatch factor (MM) has to be applied. This mismatch factor corrects

⁶Expanded uncertainty refers to the standard measurement uncertainty expanded with coverage factor $k_{\text{cov}} = 2$, thereby achieving 95.45% coverage probability.

2.2 Electrical Characterization and Calibration of Solar Cells

for the differences of how a test (TC) and a reference cell (RC), with spectral responsivities $s_{\rm TC}(\lambda)$ and $s_{\rm RC}(\lambda)$, evaluate the simulator and solar spectrum, with spectral irradiances $E_{\rm Sim}(\lambda)$ and $E_{\rm STC}(\lambda)$. The mismatch factor reads

$$MM = \frac{\int s_{TC}(\lambda) E_{Sim}(\lambda) d\lambda \int s_{RC}(\lambda) E_{STC}(\lambda) d\lambda}{\int s_{TC}(\lambda) E_{STC}(\lambda) d\lambda \int s_{RC}(\lambda) E_{Sim}(\lambda) d\lambda}$$
(2.22)

and is applied according to

$$I_{\rm SC}^{\rm STC,TC} = \frac{I_{\rm SC}^{\rm Sim,TC}}{\rm MM} \frac{I_{\rm SC}^{\rm STC,RC}}{I_{\rm SC}^{\rm Sim,RC}}$$
(2.23)

for retrieving $I_{\rm SC}^{\rm STC,TC}$ from the calibrated value $I_{\rm SC}^{\rm STC,RC}$ of the reference cell and measured values $I_{\rm SC}^{\rm Sim,TC}$ and $I_{\rm SC}^{\rm Sim,RC}$.

Being solely a correction factor on first sight, MM induces an additional uncertainty $u_{\rm MM}$ that reduces accuracy of solar simulator methods. In Chapter 7 of this thesis the impact of $u_{\rm MM}$ for various solar simulator spectral distributions and combinations of test and reference cells is investigated in detail.

2.2.2.2 Differential Spectral Responsivity Method

An alternative approach that overcomes both, daily spectral variations and imperfect replication of the solar spectrum, is represented by a spectrally resolved method. As the short circuit current generated by a solar cell with spectral responsivity $s(\lambda)$ under spectral irradiance $E_{\lambda}(\lambda)$ is given by

$$I_{\rm SC} = \int s(\lambda) E_{\lambda}(\lambda) d\lambda, \qquad (2.24)$$

the $I_{\rm SC}^{\rm STC}$ is obtained from

$$I_{\rm SC}^{\rm STC} = \int s\left(\lambda\right) E_{\rm STC}\left(\lambda\right) d\lambda.$$
(2.25)

Since $E_{\text{STC}}(\lambda)$ is a standardized solar spectrum, only $s(\lambda)$ contributes to the uncertainty of $I_{\text{SC}}^{\text{STC}}$, stressing the importance of providing accurate $s(\lambda)$ measurements for successful application of this spectrally resolved method.

The most widely spread and accepted approach for measuring $s(\lambda)$ at high accuracy is the differential spectral responsivity (DSR) method [14].⁷ In the DSR-method the solar cell under test is simultaneously illuminated by chopped monochromatic radiation ($\Delta E(\lambda)$) and steady broadband bias irradiation (E_{bias}).⁸ Measuring both the steady (dc) current response to the bias irradiation ($j_{\text{b}} = j_{\text{SC}}(E_{\text{bias}})$) and the differential (ac) current response to the chopped monochromatic radiation ($\Delta j_{\text{SC}}(E_{\text{bias}})$) reveals the test device's differential spectral responsivity

$$\tilde{s}\left(\lambda, j_{\rm b} = j_{\rm SC}\left(E_{\rm bias}\right)\right) = \left.\frac{\Delta j_{\rm SC}\left(E_{\rm bias}\right)}{\Delta E\left(\lambda\right)}\right|_{j_{\rm b} = j_{\rm SC}\left(E_{\rm bias}\right)}$$
(2.26)

at wavelength λ and dc current level $j_{\rm b}$. Assessing the bias irradiation level via $j_{\rm b}$ is important, as the differential spectral responsivity evaluated by $\Delta j_{\rm SC} (E_{\rm bias})$ might vary with the current cell condition. In other words, the currently present carrier injection density induced by $E_{\rm bias}$ and evaluated by $j_{\rm b}$ might affect the differential cell response. This injection dependence of differential measurands can be assessed by varying $E_{\rm bias}$ in a DSR-setup and taking readings of $\tilde{s} (\lambda, j_{\rm b} = j_{\rm SC} (E_{\rm bias}))$ at each bias level.

In Fig. 2.2(a) an example for bias dependency is shown by plotting $i_{\rm b}$ over $E_{\rm bias}$. For ideally linear solar cells, the slope of the curve is invariant of $E_{\rm bias}$. Thus, the differential response of the cell, $\tilde{s} (\lambda, j_{\rm b} = j_{\rm SC} (E_{\rm bias}))$, is invariant of $E_{\rm bias}$ (and, therefore, independent of changes of injection

 $^{^{7}}$ The subsequent discussion of the DSR-method follows most closely that one presented by *Winter* in [24].

⁸The broadband bias irradiation injects an excess carrier density to the solar cell, thereby setting the test cell to a specific operating condition. For obtaining carrier generation profiles similar to natural conditions a spectral distribution similar to the AM1.5g is preferable. However, this spectral constraint is much less demanding than for solar simulators (see Section 2.2.2.1) and e.g. tungsten halogen lamps (equivalent to a Class C solar simulator according to IEC 60904-9, Ed. 2) are sufficiently similar in practically all cases.

2.2 Electrical Characterization and Calibration of Solar Cells



Figure 2.2: Schematic representation of DSR-measurement outcomes for a linear (black line) and a nonlinear (red line) solar cell. (a) Bias dc current $j_{\rm b}$ plotted over bias irradiation level $E_{\rm bias}$, (b) STCweighted slopes of (a) according to Eq. (2.27). The reddish area in (b) represents $I_{\rm SC}^{\rm STC}$ and illustrates the integration according to Eq. (2.28) with $E_{\rm STC}$ as upper integration boundary.

carrier density or $j_{\rm b}$) and the trivial case $s(\lambda) = \tilde{s}(\lambda)$ is obtained. Consequently, the corresponding $I_{\rm SC}^{\rm STC}$ can be readily computed from Eq. (2.25) with $\tilde{s}(\lambda)$.

However, solar cells generally exhibit a more or less pronounced nonlinearity that is e.g. being caused by injection-dependent carrier lifetime effects (see Section 2.1.3). In these cases the differential cell response varies with bias irradiation level (see red line in Fig. 2.2(a)), thereby making above's trivial identity of differential and *absolute* responsivity invalid. Instead, a specific bias current level or cell operating condition has to be identified that corresponds to STC. For identifying that bias level, the measured differential spectral responsivities from Eq. (2.26) are weighted by the STC solar spectrum according to

$$\tilde{s}_{\rm STC} \left(E_{\rm bias} \right) = \frac{\int\limits_{0}^{\infty} \tilde{s} \left(\lambda, E_{\rm bias} \right) E_{\rm STC} \left(\lambda \right) d\lambda}{\int\limits_{0}^{\infty} E_{\rm STC} \left(\lambda \right) d\lambda}.$$
(2.27)

As these $\tilde{s}_{\text{STC}}(E_{\text{bias}})$ are equivalent to the slope of the $j_{\text{b}}(E_{\text{bias}})$ dependency for irradiation under STC (see Fig. 2.2(b)), a simple integration along E_{bias} would yield

$$I_{\rm SC}^{\rm STC} = \int_{0}^{E_{\rm STC}} \tilde{s}_{\rm STC} \left(E_{\rm bias} \right) dE_{\rm bias}, \qquad (2.28)$$

with $E_{\rm STC} = 1000 \text{ W/m}^2$.

However, in practical applications the varying bias irradiation level E_{bias} is not known or rather difficult to assess. On the contrary, the dc current generated by the bias light, j_{b} , is readily available from dc current measurements. Therefore, instead of integrating along E_{bias} as in Eq. (2.28), it is rather integrated along j_{b} with

$$E_{\rm x} = \int_{0}^{I_{\rm x}} \frac{1}{\tilde{s}_{\rm STC} \left(j_{\rm b} = j_{\rm SC} \left(E_{\rm bias}\right)\right)} {\rm d}j_{\rm b}.$$
 (2.29)

The upper integration boundary I_x that yields $E_x = E_{STC} = 1000 \text{ W/m}^2$ is equivalent to the I_{SC}^{STC} .

Finally, the spectral responsivity $s(\lambda)$ is obtained from

$$s(\lambda) = I_{\rm SC}^{\rm STC} \left[\int_{0}^{I_{\rm SC}^{\rm STC}} \frac{1}{\tilde{s}(\lambda, j_{\rm b} = j_{\rm SC}(E_{\rm bias}))} \mathrm{d}j_{\rm b} \right]^{-1}.$$
 (2.30)

2.2 Electrical Characterization and Calibration of Solar Cells

The DSR-method is implemented into the laser-based measurement facility that has been developed in this work and is applied for measurements presented in Chapters 6 to 8.

Chapter 3

Ultrashort Laser Pulses & Nonlinear Optics

In this chapter the theoretical background on ultrashort pulses and nonlinear optics will be given that is relevant for the results and discussions in the remainder of this work. Starting from a brief introduction on the basic principles of ultrashort pulses, the origin of nonlinear optical effects and a theoretical framework for describing them will be given. This will be used subsequently to outline some second and third order nonlinear optical effects, followed by a short introduction to phasematching. Finally, the principle of supercontinuum generation from ultrashort laser pulses and photonic crystal fibers will be sketched concisely.

3.1 Basic Principles

In the course of this work a measurement system for solar cell spectral responsivities is developed that is being based on ultrashort laser pulses as illumination source. Owing to its quasi-monochromaticity this radiation source provides a significantly higher spectral power as available from typically used conventional white light sources (xenon or halogen lamps), thereby enabling considerable reductions in measurement uncertainties (see Chapter 6). Naturally, the high spectral powers required for such a purpose are also available from continuous wave (cw) or Q-switched¹ lasers. However, the spectral range and availability of lasers is intrinsically limited by the applied (gain) materials impeding their usage in the new measurement system that requires spectral tunability from the UV (approx. 300 nm) to the NIR (1200 nm or higher).

One way to overcome this bottleneck is to apply nonlinear optical effects for generating radiation in the entire spectral range. Ultrashort pulses are especially advantageous in this context, as their extremely high pulse peak powers allow for efficient nonlinear optical processes (see Section 3.2) at moderate pulse energies and average powers (thus, in a quasi- cw^2 manner). For instance, a laser emitting ultrashort pulses of 100 fs full-width-at-half-maximum (FWHM) pulse duration at 80 MHz repetition rate achieves a pulse peak power of more than 110 kW at an average output power of 1 W. In this section the basic principle of ultrashort laser pulse generation and typical properties of these pulses, that are important for the remainder of this work, will be briefly outlined. Readers interested in a detailed discussion of ultrashort pulses are referred to the textbook Ultrafast Optics by Andrew M. Weiner [25].

The underlying principle of ultrashort pulse generation is the superposition of longitudinal resonator modes of different frequencies resulting in pulse durations that are essentially limited by the transform limit according to [26, p. 10]

$$\tau_{\rm FWHM} \Delta \omega \ge 2\pi c_{\rm TBP},$$
(3.1)

with FWHM pulse duration $\tau_{\rm FWHM}$, FWHM frequency bandwidth $\Delta \omega = 2\pi \Delta \nu$ and the minimum possible time-bandwidth-product $c_{\rm TBP}$ that takes different values depending on the temporal pulse shape. E.g. for a Gaussian pulse ($c_{\rm TBP} = 0.441$) with $\tau_{\rm FWHM} = 100$ fs a frequency bandwidth

¹In Q-switching a repetitive variation of the cavity losses by means of mechanical or electro-optical shutters is used to achieve a very high population inversion in the laser gain medium (for closed shutters) that is depleted when the losses are turned off, thereby (typically) emitting ns-pulses.

²The importance of high repetition rates (*quasi-cw* operation) arises from the desired temporal continuity of the radiation used for the solar cell measurements and is addressed in Chapters 4 and 5.

of $\Delta \omega \geq 2.77 \cdot 10^{13}$ Hz (corresponding to $\Delta \lambda \geq 9.4$ nm at 800 nm center wavelength) is required. In a laser cavity such ultrashort pulses are generated by *mode-locking* longitudinal resonator modes so that they have a fixed phase relation to each other and constructively interfere in a periodic manner. Assuming a typical longitudinal mode spacing of 80 MHz (corresponding to approx. 1.9 m cavity length) the generation of an ultrashort pulse as given above requires more than 50.000 mode-locked longitudinal laser modes.

In practice, this mode-locking can be achieved by e.g. active or passive modulation of the laser resonator gain or loss. In active mode-locking, for example, an acousto-optic modulator (AOM) can be used to sinusoidally modulate the cavity losses at a frequency identical to the inverse of the cavity roundtrip time. Thereby longitudinal modes that are in phase with the modulation frequency are favored establishing mode-locking of all longitudinal modes. In analogy to the AOM, a saturable absorber can also be applied that bleaches with increasing intensity. As the cavity losses reduce with increased absorber transmission (thus, for a bleached absorber), the laser favors an operational mode where all longitudinal modes are in phase, thus mode-locked. However, as the losses are not actively controlled by the saturable absorber, this technique is referred to as passive mode-locking. Please note, that this is only a very short digest on mode-locking techniques. For a detailed discussion of mode-locking techniques and their theoretical treatment it is referred to [25, pp. 32-84].

3.2 Nonlinear Optics

The previously discussed strong temporal confinement of optical power by means of mode-locking leads to very high pulse peak powers in ultrashort laser pulses that possibly imply nonlinear optical phenomena. These phenomena are subject of various topics in this work ranging from the generation of new wavelengths or spectral components (e.g. Chapters 6 and 7) to the investigation of their influence on solar cell device characterization using ultrashort pulses (Chapter 4).

In this section a short digest on the theory of nonlinear optics will

be given introducing the basic principles of the phenomena utilized and investigated in the remainder of this work. Starting from a brief explanation on the origin of nonlinear phenomena, the second and third order nonlinear optical processes relevant for this work will be described. Afterwards a derivation of the nonlinear wave equation will be presented that is essential for concluding the coupled amplitude equations. The discussion of nonlinear optics and its associated phenomena follows most closely the textbooks by *Zernike* and *Midwinter* [27] as well as by *Boyd* [28]. Finally, a very short overview on the generation of a supercontinuum from nonlinear optical processes will be given, mostly based on the review paper by *Dudley* [29].

3.2.1 From Linear to Nonlinear Optics

The interaction of electromagnetic radiation and a dielectric medium (thus, a medium with no free charges or currents) is essentially governed by $\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P}$, relating the electric displacement³ \mathbf{D} to the electric field \mathbf{E} and the electric properties of the medium described by the polarization density \mathbf{P} . In linear optics, the polarization density of a homogeneous and nondispersive dielectric medium is given by $\mathbf{P} = \varepsilon_0 \hat{\chi} \mathbf{E}$ with vacuum permittivity ε_0 and the electric susceptibility⁴ $\hat{\chi}$. Thus, the dielectric displacement can be likewise written as $\mathbf{D} = \hat{\varepsilon} \mathbf{E}$ with $\hat{\varepsilon} = \varepsilon_0 \hat{\varepsilon}_r$, introducing the relative permittivity $\hat{\varepsilon}_r = 1 + \hat{\chi}$ that is related to the material's refractive index by $n_0 = \sqrt{\hat{\varepsilon}_r}$.

In case of high electric field strengths the linear relation of polarization density and electric field might not sufficiently describe the field-matter interaction anymore. In nonlinear optics it is therefore convenient to expand the polarization density into the power series

$$\tilde{\mathbf{P}}(t) = \varepsilon_0 \left[\hat{\chi}^{(1)} \tilde{\mathbf{E}}(t) + \hat{\chi}^{(2)} \tilde{\mathbf{E}}^2(t) + \hat{\chi}^{(3)} \tilde{\mathbf{E}}^3(t) + \dots \right]$$
(3.2)

with nonlinear optical susceptibilities $\hat{\chi}^{(2)}, \hat{\chi}^{(3)}, \ldots$

³Also being called electric flux density.

⁴The hat denotes that $\hat{\chi}$ is a tensor. In an isotropic medium $\hat{\chi}$ can be replaced by the scalar equivalent χ .

3.2 Nonlinear Optics

A rather intuitive way for explaining the origin of these nonlinearities is provided by adapting the Lorentz model to nonlinear processes. In the Lorentz model the one-dimensional displacement r of a bound electron of charge q and mass m from its equilibrium position as a response to an applied electric field $\tilde{E}(t) = \mathcal{E} \cos(\omega t - \varphi)$ is described by the driven harmonic oscillator function

$$\frac{\mathrm{d}^{2}\tilde{r}\left(t\right)}{\mathrm{d}t^{2}} + 2\gamma \frac{\mathrm{d}\tilde{r}\left(t\right)}{\mathrm{d}t} - \frac{F_{\mathrm{restore}}}{m} = -\frac{q}{m}\tilde{E}\left(t\right)$$
(3.3)

with damping constant γ and restoring force F_{restore} . In the branch of linear optics the restoring force is assumed to be linearly dependent on the displacement according to $F_{\text{restore}} = -m\omega_0^2 r$. With this and ansatz $\tilde{r}(t) = r \exp(-i\omega t) + c.c.$ Eq. (3.3) is solved⁵ by

$$\tilde{r}(t) = -\frac{q}{m} E \exp\left(-i\omega t\right) \frac{1}{\omega_0^2 - 2i\gamma\omega - \omega^2} + c.c.$$
(3.4)

with c.c. denoting the complex conjugate, complex notation for the electric field $\tilde{E}(t) = E \exp(-i\omega t) + c.c.$ with $E = \mathcal{E} \exp(i\varphi)/2$ and real amplitude \mathcal{E} .

As the (one-dimensional) polarization density is equivalent to the product of density of electrons N, electron displacement r and charge q it follows from Eqs. (3.2) and (3.4) that

$$\tilde{P}(t) = \varepsilon_0 \chi^{(1)}(\omega) E \exp(-i\omega t) + c.c., \qquad (3.5)$$

with

$$\chi^{(1)}(\omega) = \frac{Nq^2}{\varepsilon_0 m} \frac{1}{\omega_0^2 - 2i\gamma\omega - \omega^2}$$
(3.6)

demonstrating that $\tilde{P}(t)$ oscillates at the same frequency as the incident field, thereby emitting electromagnetic radiation at the same frequency.

In contrast to this linear case, the restoring force F_{restore} applied to Eq. (3.3) might incorporate nonlinear terms as well, resulting from nonparabolic potential energy functions $U(r) = -\int F_{\text{restore}} dr$ as shown

⁵The solution can be retraced in e.g. [27, p. 3].



Figure 3.1: Potential energy functions for (a) a noncentrosymmetric medium (red lines) and a (b) centrosymmetric medium (red line). The black lines represent parabolic and the red lines nonparabolic potential energy functions. Both figures are adapted from [28].

in Fig. 3.1. The parabolic potential function (black line in Figs. 3.1a and 3.1b) results in the linear F_{restore} applied above, whereas the nonparabolic functions (red lines) give nonlinear restoring forces (e.g. $F_{\text{restore}} = -m\omega_0^2 r - ma_2 r^2$ for a noncentrosymmetric medium as in Fig. 3.1a and $F_{\text{restore}} = -m\omega_0^2 r + ma_3 r^3$ for a centrosymmetric medium as in Fig. 3.1b, with a_2 and a_3 describing the strength of nonlinearity).

Both nonparabolic potential energy functions result in an anharmonic oscillator equation (compare Eq. (3.3)) that can be solved by means of perturbation theory with a power series expansion according to $\tilde{r} = \kappa \tilde{r}^{(1)} + \kappa^2 \tilde{r}^{(2)} + \kappa^3 \tilde{r}^{(3)} + \dots$ and perturbation strength κ . The striking result of this approach⁶ for solving the anharmonic oscillator equation is the existence of higher-order or difference- and sum-frequencies in the correction-terms $\tilde{r}^{(2)}, \tilde{r}^{(3)}, \dots$ For example, an incident electric field with two frequencies ω_1 and ω_2 results in the oscillation frequencies $\pm 2\omega_1, \pm 2\omega_2, \pm (\omega_1 - \omega_2), \pm (\omega_1 + \omega_2)$ and 0 for $\tilde{r}^{(2)}$. In analogy to Eq. (3.5) the polarization density oscillations resulting from the nonlinearities in the restoring force generate electromagnetic radiation at these

 $^{^{6}}$ In Appendix A the solving scheme for the anharmonic oscillator in case of a noncentrosymmetric potential energy function using the perturbation theory is outlined.

new frequencies. Similar to the linear susceptibility $\chi^{(1)}$, the nonlinear susceptibilities $\chi^{(2)}, \chi^{(3)}, \ldots$ can be determined from $\tilde{r}^{(2)}, \tilde{r}^{(3)}, \ldots$ which is exemplarily shown in Appendix A for second harmonic generation, thus, the $2\omega_{1,2}$ frequency.

After this short motivation on the origin and description of nonlinear optical effects, those relevant for the remainder of this work will be outlined in the subsequent section.

3.2.2 Survey on Nonlinear Optical Effects

3.2.2.1 Second Order Processes

Second Harmonic Generation

Assuming an electric field $\tilde{E}(t) = E \exp(-i\omega t) + c.c.$ incident on a noncentrosymmetric, loss- and dispersionless dielectric medium, Eq. (3.2) in scalar representation yields

$$\tilde{P}(t) = \varepsilon_0 \chi^{(1)} \left(E \exp\left(-i\omega t\right) + c.c.\right) + 2\varepsilon_0 \chi^{(2)} E E^* + \left(\varepsilon_0 \chi^{(2)} E^2 \exp\left(-i2\omega t\right) + c.c.\right)$$
(3.7)

with the asterisk denoting a complex conjugation. The first term on the right hand side of the equation is the previously discussed linear contribution to the polarization density re-emitting electromagnetic radiation at the same frequency. The second term is a non-oscillating field referred to as *optical rectification*,⁷ a steady polarization resulting from the anharmonic potential as shown in Fig. 3.1a. The third term is denoted as *second harmonic generation (SHG)* as the polarization density oscillates at 2ω and is, therefore, generating electromagnetic radiation at twice the frequency of the incident field. Within the scope of this work, SHG is mainly used for the generation of photons with higher energies $(E_{\rm ph} = \omega\hbar + \omega\hbar = 2\omega\hbar)$ and, thus, shorter wavelengths ($\lambda = hc_0/E_{\rm ph}$), as illustrated by the energy-level diagram on the right side of Fig. 3.2.

 $^{^{7}}$ Caused by the noncentrosymmetric potential energy the electron moved by the incident electric field is, on average, not at its equilibrium position. This results in a non-zero average polarization denoted as *optical rectification*.



Figure 3.2: Illustration of second harmonic generation (SHG). Left: incident electric field with frequency ω is partially converted into an electric field at twice the intial frequency 2ω . Right: energy-level diagram of SHG illustrating the generation of a photon with energy $E_{\rm ph} = \omega \hbar + \omega \hbar = 2\omega \hbar$ (adapted from [28]).



Figure 3.3: Illustration of sum-frequency generation (SFG). Left: incident electric fields with frequencies ω_1 and ω_2 generate a field at frequency $\omega_3 = \omega_1 + \omega_2$. Right: energy-level diagram of SFG illustrating the energy balance $E_{\rm ph}(\omega_3) = E_{\rm ph}(\omega_1) + E_{\rm ph}(\omega_2)$ (adapted from [28]).



Figure 3.4: Illustration of difference-frequency generation (DFG). Left: incident electric fields with frequencies ω_1 and ω_2 generate a field at frequency $\omega_3 = \omega_1 - \omega_2$. Right: energy-level diagram of DFG illustrating the generation of two photons at $E_{\rm ph}(\omega_2)$ and $E_{\rm ph}(\omega_3)$ from a single photon of $E_{\rm ph}(\omega_1)$ yielding an amplification of the incident ω_2 -field (adapted from [28]).

Sum- & Difference-Frequency Generation

Applying a field with two frequencies $\tilde{E}(t) = E_1 \exp(-i\omega_1 t) + E_2 \exp(-i\omega_2 t) + c.c.$ to the same medium as above gives (again with Eq. (3.2), but neglecting the linear susceptibility for clarity)

$$\tilde{P}^{(2)}(t) = \varepsilon_0 \chi^{(2)} \left[E_1^2 e^{-i2\omega_1 t} + E_2^2 e^{-i2\omega_2 t} + 2E_1 E_2 e^{-i(\omega_1 + \omega_2) t} + 2E_1 E_2^* e^{-i(\omega_1 - \omega_2) t} + c.c. \right] + 2\varepsilon_0 \chi^{(2)} \left(E_1 E_1^* + E_2 E_2^* \right).$$
(3.8)

Analogous to the previous case, the last term denotes optical rectification and the first two terms SHG of the ω_1 and ω_2 frequency, respectively. In addition to that, terms oscillating at frequencies ($\omega_1 + \omega_2$) and ($\omega_1 - \omega_2$), denoted as sum- (SFG) and difference-frequency generation (DFG) appear.

In SFG a higher frequency radiation ω_3 is generated by the summation of the two incident frequencies ω_1 and ω_2 (see Fig. 3.3). Within the scope of this work, SFG is used to generate the third harmonic (*third harmonic* generation (*THG*)) of an initial frequency ω_1 by a cascaded process of SHG (to generate $\omega_2 = 2\omega_1$) and SFG (to get $\omega_3 = \omega_1 + \omega_2 = 3\omega_1$).

In contrast to SFG, where one photon is created from two photons, in DFG two photons at $E_{\rm ph}(\omega_2)$ and $E_{\rm ph}(\omega_3)$ are produced from a single photon with $E_{\rm ph}(\omega_1)$ (see energy-level diagram on the right side of Fig. 3.4). In other words, the ω_2 -field is amplified by DFG⁸ causing the process of DFG also being called *optical parametric amplification (OPA)*. In the course of this work, DFG (or OPA) is used in an *optical parametric* oscillator (OPO) that is pumped at $\lambda_1 = 410$ nm and generates a so-called signal (with $\lambda_2 > \lambda_1$; $\omega_2 < \omega_1$) and idler radiation (with $\lambda_3 > \lambda_2 > \lambda_1$; $\omega_3 < \omega_2 < \omega_1$). The term oscillator denotes that the signal radiation is resonantly oscillating in the cavity of the OPO, thereby stimulating the process of OPA.

⁸DFG might also take place without ω_2 or ω_3 being initially present. However, as this process is not *stimulated* but *spontaneous* the generated fields are much lower.

3.2.2.2 Nonlinear Refractive Index

In centrosymmetric material (e.g. glasses or liquids), represented by the symmetric potential energy function shown in Fig. 3.1b, the $\chi^{(2)}$ term vanishes for symmetry reasons and the $\chi^{(3)}$ of Eq. (3.2) term becomes the first nonlinear contribution. Applying an electrical field $\tilde{E}(t) = E \exp(-i\omega t) + c.c.$ gives

$$\tilde{P}^{(3)}(t) = \varepsilon_0 \chi^{(3)} \left(E^3 e^{-i3\omega t} + 3EE^* Ee^{-i\omega t} + c.c. \right)$$
(3.9)

for the third order polarization density. The first term oscillates at frequency 3ω and represents THG and the second term oscillates at ω , thus, at the same frequency as the linear component of polarization density $\tilde{P}^{(1)}(t) = \varepsilon_0 \chi^{(1)} E \exp(-i\omega t) + c.c.$ (see Eqs. (3.5) and (3.6)). Thus, the entire polarization density oscillating at ω is given by

$$P(\omega) = P^{(1)}(\omega) + P^{(3)}(\omega) = \varepsilon_0 \left(\chi^{(1)} + 3\chi^{(3)}|E|^2\right)E$$
(3.10)

with $\tilde{P}^{(j)}(t) = P^{(j)}(\omega) \exp(-i\omega t)$. Recalling that $n_0^2 = \varepsilon_r = 1 + \chi^{(1)}$ in the linear case (see Section 3.2.1) yields for the third-order nonlinear case $n^2 = 1 + \chi^{(1)} + 3\chi^{(3)}|E|^2$ or

$$n^{2}(I) = n_{0}^{2} + 3\chi^{(3)} \frac{I}{2n_{0}\varepsilon_{0}c_{0}}$$
(3.11)

with intensity $I = 2n_0\varepsilon_0 c_0 |E|^2$. Introducing the nonlinear refractive index n_2 and approximating $n^2(I) = (n_0 + n_2 I)^2 \approx n_0 + 2n_0 n_2 I$ gives for the nonlinear refractive index

$$n_2 = \frac{3}{4n_0^2\varepsilon_0 c_0}\chi^{(3)}.$$
(3.12)

This nonlinear optical effect, being referred to as *Kerr effect*, results in an instantaneous change of the refractive index that, in turn, affects the propagation of the radiation itself by e.g. self-phase modulation.⁹

 $^{^{9}}$ Also, in the non-degenerate case, one frequency radiation can affect the propagation of another frequency radiation that is, for example, too weak to introduce

As this effect is of tremendous importance for the generation of optical supercontinua and the formation of temporal solitons more details are given in Section 3.2.5.

3.2.2.3 Multi-Photon Absorption

The previously discussed nonlinear processes are so-called parametric processes that incorporate virtual energy levels of the material. In contrast to these, multi-photon absorption is a nonparametric process, thus, it induces changes to the material properties by e.g. transferring population from one real level to another.

In multi-photon absorption processes this is accomplished by simultaneous interaction of two or more photons with a single electron. By simultaneous transfer of their energies to the electron a transition to a higher energy state can be reached. Naturally, the likeliness for such transitions increases with the photon rate and, thus, the transition cross section for two-photon absorption depends on the incident intensity I according to $\sigma = \sigma_{\beta} I$. Consequently,¹⁰ the transition rate for two-photon absorption reads

$$T_{\rm trans} = \frac{\sigma_\beta I^2}{h\nu} \tag{3.13}$$

and has a square-dependence on the intensity I.

3.2.3 Nonlinear Wave Equation

As detailed above new frequency components are generated by means of nonlinear optical effects that themselves act as new sources. In this section the theoretical framework to this will be given in terms of the nonlinear wave equation.

Starting from Maxwell's equation and assuming no free charges ($\rho = 0$),

refractive index changes to the material by itself. This nonlinear optical effect is called *cross-phase modulation*.

¹⁰Recalling that the transition rate for linear absorption equals $T_{\text{trans}} = \sigma_{\alpha} I / (h\nu)$.

no free currents ($\mathbf{j} = 0$) and nonmagnetic materials ($\mu = \mu_0$)

$$\nabla \times \nabla \times \mathbf{E} + \frac{1}{c_0^2} \frac{\partial^2}{\partial t^2} \mathbf{E} = -\frac{1}{\varepsilon_0 c_0^2} \frac{\partial^2}{\partial t^2} \mathbf{P}$$
(3.14)

is obtained from $\nabla \times \mathbf{E} = -\frac{\partial}{\partial t} \mathbf{B}$ and $\nabla \times \mathbf{H} = \frac{\partial}{\partial t} \mathbf{D} + \mathbf{j}$ with $\mathbf{B} = \mu_0 \mathbf{H}$, $\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P}$ and $\mu_0 \varepsilon_0 = c_0^{-2}$.

With the identity $\nabla \times \nabla \times \mathbf{E} = \nabla (\nabla \cdot \mathbf{E}) - \nabla^2 \mathbf{E}$ and the assumption that $\nabla \cdot \mathbf{E} \approx 0$ it follows

$$\nabla^{2}\mathbf{E} - \frac{1}{c_{0}^{2}}\frac{\partial^{2}}{\partial t^{2}}\mathbf{E} = \frac{1}{\varepsilon_{0}c_{0}^{2}}\frac{\partial^{2}}{\partial t^{2}}\left[\mathbf{P}^{(1)} + \mathbf{P}^{(\mathrm{NL})}\right].$$
 (3.15)

Considering loss- and dispersionless material, the subdivision of polarization density \mathbf{P} in its linear and nonlinear contribution is beneficial as it allows for a reformulation of Eq. (3.15) to

$$\nabla^{2}\mathbf{E} - \frac{\varepsilon_{r}^{(1)}}{c_{0}^{2}}\frac{\partial^{2}}{\partial t^{2}}\mathbf{E} = \frac{1}{\varepsilon_{0}c_{0}^{2}}\frac{\partial^{2}}{\partial t^{2}}\mathbf{P}^{(\mathrm{NL})},\qquad(3.16)$$

a driven wave equation clearly identifying the nonlinear polarization $\mathbf{P}^{(NL)}$ as source term for a wave.

Further it can be shown that including dispersion in Eq. (3.16) yields

$$\nabla^2 \mathbf{E}_j + \frac{\omega_j^2}{c_0^2} \varepsilon_0^{(1)}(\omega_j) \, \mathbf{E}_j = -\frac{\omega_j^2}{\varepsilon_0 c_0^2} \mathbf{P}_j^{(\mathrm{NL})}, \qquad (3.17)$$

which is valid for each frequency component ω_j .

3.2.4 Coupled Amplitude Equations and Phase-Matching

In the previous section the nonlinear wave equation Eq. (3.17) was derived, identifying the nonlinear polarization density - induced by nonlinear optical effects - as source term for a wave. As energy may flow back and



Figure 3.5: Illustration of phase-matching condition for SFG. The intensity of the field with frequency ω_3 after interaction (or crystal) length L varies with sinc $(\Delta kL/2)$ (adapted from [28]).

forth between fundamental and new frequencies, coupled amplitude equations can be derived that highlight the importance of phase-matching for efficiency in nonlinear optical effects.

For example, the coupled amplitude equations for SFG read¹¹

$$\frac{\partial A_1}{\partial z} = i \frac{2d_{\text{eff}}\omega_1^2}{k_1 c^2} A_3 A_2^* e^{-i\Delta kz}$$
(3.18a)

$$\frac{\partial A_2}{\partial z} = i \frac{2d_{\text{eff}}\omega_2^2}{k_2 c^2} A_3 A_1^* e^{-i\Delta kz}$$
(3.18b)

$$\frac{\partial A_3}{\partial z} = i \frac{2d_{\text{eff}}\omega_3^2}{k_3 c^2} A_1 A_2 e^{i\Delta kz}, \qquad (3.18c)$$

with amplitudes A_j , wave numbers $k_j = \frac{n_j \omega_j}{c_0}$, $n_j^2 = \varepsilon_r^{(1)}(\omega_j)$, effective nonlinearity¹² d_{eff} and the wave vector mismatch $\Delta k = k_1 + k_2 - k_3$.

The importance of Δk can be illustrated assuming the most trivial case of constant A_1 and A_2 along propagation direction z. It can be shown that

 $^{^{11}\}mathrm{For}$ a derivation of these coupled amplitude equations the reader is referred to [28, pp. 74-76].

¹²This is a typical notation for the strength of nonlinearity in an optical crystal. By several symmetry considerations, that are not further discussed here, and for fixed geometries the susceptibility χ (remember that χ is a tensor in general) is conveniently reduced to a single scalar value d_{eff} .

the intensity $I_3 = 2n_3\varepsilon_0c_0|A_3|^2$ varies with sinc $(\Delta kL/2)$ (see Fig. 3.5), where L denotes the interaction or crystal length, respectively.

Therefore, achieving phase-matching ($\Delta k = 0$) is critical for nonlinear optical interactions over extended regions of space. This phase-matching can e.g. be achieved in birefringent crystals that exhibit different refractive indices depending on the polarization direction of the electromagnetic wave. In such crystals the refractive indices of perpendicularly polarized ω - and 2ω -radiation (in case of SHG, for example) can be tuned to fulfill $n(\omega) = n(2\omega)$ by angle- or temperature-tuning of the crystal. In the course of this work, angle-tuning is applied for efficient SHG, THG (by SHG plus SFG, see Section 3.2.2.1) and DFG (as used in the OPO).

3.2.5 Supercontinuum Generation

The term *supercontinuum generation* denotes the generation of a broad spectrum from an initial narrow-band input radiation. Typically, supercontinuum generation is accomplished by propagating high intensity radiation through optical fibers (preferably, by taking advantage of the high peak powers of (ultra)short pulses, although continuous wave (cw) supercontinua were demonstrated as well [30]) and exploiting diverse nonlinear optical effects in order to accomplish the spectral broadening.

As a detailed discussion of the complex nature of supercontinuum generation, that depends on pulse duration, fiber dispersion, peak power and wavelength, is beyond the scope of this work, only the parameter domain relevant for this work (fs-pulses of nJ pulse energies in the anomalous group velocity dispersion $(\text{GVD})^{13}$ regime of a photonic crystal fiber (PCF)) is briefly discussed along the lines of the review paper by *Dudley* et al. [29]. Please note, that this outline intends to give the reader some general idea of the processes involved in supercontinuum generation rather than giving a profound theoretical background.¹⁴

In Fig. 3.6 (from [29]) the spectral and temporal evolution associated with supercontinuum generation in a PCF, under similar experimental

 $^{^{13}}$ The term GVD is clarified below.

 $^{^{14}}$ For readers interested in an in-depth discussion the review paper by *Dudley* et al. [29] appears to be an appropriate starting point.



Figure 3.6: Simulated spectral (left) and temporal evolution (right) of supercontinuum generation in a PCF for similar fiber and input pulse properties as used in this work. Graphic from [29].

conditions as applied in this work, is shown. A key condition for such a supercontinuum generation is the existence of *solitons* that are solutions to the nonlinear Schrödinger equation¹⁵

$$i\frac{\partial\tilde{a}_s}{\partial\zeta} = \frac{\operatorname{sign}\left(k_2\right)}{2}\frac{\partial^2\tilde{a}_s}{\partial t'^2} - N_{\operatorname{sol}}^2|\tilde{a}_s|^2\tilde{a}_s,\qquad(3.19)$$

with time and space variables t' and ζ , pulse envelope $a_s(t', \zeta)$ and soliton order N_{sol} . For $N_{\text{sol}} = 1$ the *fundamental* soliton solution

$$\tilde{a}_s(t',\zeta) = \operatorname{sech}(t') e^{i\zeta/2} \tag{3.20}$$

is obtained that depends only in its phase on the spatial variable ζ . Thus, the pulse remains unaltered in its temporal shape while propagating through the PCF, thereby maintaining the high peak power required for efficient nonlinear processes. The invariance of temporal shape is a remarkable feature as it represents the case where GVD compensates

 $^{^{15}\}mathrm{A}$ derivation of this equation is given in Appendix B.

self-phase modulation (SPM), two subsequently introduced optical effects affecting the pulse propagation.

GVD is a linear optical process related to the wavelength dependency of group velocity v_g that results in temporal broadening of pulses. Mathematically, GVD can be understood from the second term of a Taylor expansion of the propagation constant $k = n_0 \omega/c_0$ - obtained from solving the wave equation (e.g. Eq. (3.16) for $\mathbf{P}_j^{(\mathrm{NL})} \approx 0$) - given by [28]

$$k_2 = \left. \frac{\mathrm{d}^2 k}{\mathrm{d}\omega^2} \right|_{\omega=\omega_0} = \left(-\frac{1}{v_g^2} \frac{\mathrm{d}v_g}{\mathrm{d}\omega} \right)_{\omega=\omega_0}.$$
 (3.21)

For positive or normal GVD $(k_2 > 0) v_g$ increases with wavelength and red wavelengths propagate faster than blue wavelengths. Naturally, for negative or anomalous GVD $(k_2 < 0)$ the opposite is true.

In contrast to GVD, SPM is a nonlinear optical process. It results from the intensity dependent refractive index $n(t) = n_0 + n_2 I(t)$ introduced in Section 3.2.2.2 and causes a phase change of the pulse while it propagates through a dielectric material. In mathematical terms, the instantaneous frequency $\omega(t)$, defined as the temporal derivative of phase $\varphi(t)$, is introduced to describe SPM. As the time-dependent phase is given by $\varphi(t) = \omega_0 t - \omega_0 n_2 I(t) z/c_0$, the instantaneous frequency reads

$$\omega(t) = \omega_0 - \frac{\omega_0}{c_0} n_2 z \frac{\partial I(t)}{\partial t}.$$
(3.22)

Thus, with $n_2 > 0$, which is the case for e.g. fused silica, the frequencies at the leading edge of a pulse $(\partial I/\partial t > 0)$ are shifted to lower values (corresponding to a red shift in the wavelength domain) and those at the trailing edge $(\partial I/\partial t < 0)$ to higher values (blue shift in wavelength domain). Therefore, anomalous GVD is required for balancing of GVD and SPM and the creation of a fundamental soliton.¹⁶

Apart from fundamental solitons, higher-order solitons exist for $N_{sol} > 1$ in Eq. (3.19) that periodically change their temporal and spectral shape

¹⁶Without going into detail, this represents an important advantage of PCFs regarding supercontinuum generation as their zero-dispersion wavelength can be tuned in a quite broad range [29].

while propagating. In Fig. 3.6 the initial higher-order solition appearance is represented by the extreme spectral broadening within the first centimeter. After this initial spectral broadening the higher-order soliton is typically split into several fundamental solitons [29] (a process called *soliton fission*) that themselves continue propagating unaltered except for a self-frequency shift to longer wavelengths induced by stimulated Raman scattering (especially visible for the longest wavelength soliton in Fig. 3.6).¹⁷ These individual fundamental solitons can be observed to the best in the temporal domain representation of Fig. 3.6.

The continuous red-shift of the fundamental solitons also causes a variation in the GVD parameter k_2 resulting in temporal reshaping of the soliton as to maintain its order number $N_{\rm sol} = 1$. In addition to that, higher-order dispersion induces energy transfer from the soliton to resonant shorter wavelengths by *dispersive wave generation* that appear as sharp short wavelength limit in Fig. 3.6 [32].

A combination of the discussed processes yields an extreme spectral broadening even for comparably short interaction length (15 cm in this example) and low pulse energies as emitted from ultrashort pulse oscillators. Within the scope of this work, such a supercontinuum is generated from coupling ultrashort laser pulses of about 100 fs pulse duration and below 100 kW pulse peak power into a 12 cm long PCF in the anomalous GVD regime. The generated white spectrum, that exhibits a high spatial coherence, is spectrally shaped afterwards and used for advanced solar cell characterization methods (see Chapter 7).

¹⁷The fundamental solitons are of such broad spectral widths that the longer wavelengths are amplified at the cost of power at shorter wavelengths shifting the soliton to longer wavelengths [31].

Chapter 4

Interaction of Ultrashort Laser Pulses and Solar Cells

In this chapter the interaction of ultrashort laser pulses and solar cells regarding their electrical characterization is discussed. Firstly, a theoretical model is developed that describes this interaction and takes into account potential nonlinearities induced by the ultrashort laser pulses. Afterwards, this model is reduced and simplified by several approximations considering minority carrier lifetimes, pulse repetition frequencies and radiation intensities. Secondly, contribution of nonlinear absorption to the measured short circuit current in a silicon solar cell is experimentally demonstrated and applied for validation of the theoretical model. With this approach the most prominent, but experimentally hardly feasible, comparison of current generation for pulsed versus continuous illumination is conducted. The chapter is concluded by recommendations regarding the applicability of ultrashort laser pulses for highly accurate electrical characterization of solar cells. Parts of this chapter were published in a previous publication of the author of this work [33].

4.1 Introduction

The extraordinary features of ultrashort laser pulses triggered a remarkable progress in the development of this radiation source since the first reports of mode-locked ruby [34] and Nd:glass lasers [35] in the 1960s. The unprecedented temporal resolution achieved with ultrashort pulse durations in the picosecond(ps)-regime established the field of ultrafast spectroscopy [36]. Subsequent breakthroughs in that field were prompted by the development of colliding pulse mode-locked dye lasers in the 1980s [37], achieving pulse durations down to 6 femtoseconds(fs) [38], and selfmode-locking titanium-sapphire solid-state-lasers in 1991 [39], improving efficiency, stability and day-to-day practicality, thereby assisting commercialization of ultrashort pulse lasers [40].

In the last 25 years, further significant progress has been made in both development and application of ultrashort pulse lasers. The femtosecond barrier [41] was overcome and isolated attosecond(as)-pulses down to 67 as [42] were generated, marking another milestone in time-resolved spectroscopy that opened the field of attosecond physics [43]. Besides the striking temporal characteristics exploited for ultrafast spectroscopy and femtochemistry [44],¹ the intense peaks of ultrashort pulses exceeding 10^{18} W/cm² are used to study relativistic laser-plasma interactions [46, 47]. The combination of high peak power and short duration of ultrashort pulses is further extensively applied in precision machining [48–50]. Moreover, the spectral properties of ultrashort pulses are exploited in frequency metrology using frequency combs [51].^{2,3}

In the course of this work, the high peak power of ultrashort laser

¹In 1999 the Nobel Prize in Chemistry was awarded to *Ahmed H. Zewail* "for his studies of the transition states of chemical reactions using femtosecond spectroscopy" [45].

²In 2005 the Nobel Prize in Physics was awarded to John L. Hall and Theodor W. Hänsch "for their contributions to the development of laser-based precision spectroscopy, including the optical frequency comb technique" [52].

³This short introduction to the development and application of ultrashort laser pulses does not claim to be comprehensive. For a more profound introduction the reader is referred to the given literature, especially to [25, 36, 40].

pulses is exploited for efficient spectral conversion by nonlinear optical effects. Various nonlinear components are combined in a laser system that enables gap-free wavelength-tuning from 230 nm to 2500 nm (for details see Chapter 6). By taking advantage of higher average spectral powers provided by the ultrashort pulse laser system as compared to conventionally applied radiation sources, a new measurement facility for highly accurate electrical characterization of solar cells is developed [53]. Being advantageous regarding spectral power and coverage, usage of ultrashort pulses might cause nonlinear effects not being present under continuous illumination and impeding their usage for highly accurate measurements. In order to assess these effects the interaction of ultrashort pulses and semiconductors is of major interest and has to be addressed prior to a final setup configuration.

With the rise of ultrafast spectroscopy a broad variety of ultrashort pulse based time-resolved methods as e.g. pump-probe, four-wave-mixing (FWM), luminescence, interferometric or terahertz (THz) spectroscopy were successfully applied to study semiconductors. The entity of these works resulted in a profound understanding of carrier, exciton and phonon dynamics on time scales down to the fs-regime. A thorough review on the activities in that field is given by Shah in [54]. Moreover, ultrashort pulses enabled experimental evaluation of nonlinear optical properties like Kerr nonlinearity or two-photon absorption (TPA) (as e.g. in [55]). However, to the best of the author's knowledge, no work has yet been specifically addressing ultrashort pulse carrier generation and extraction in solar cells. Therefore, a theoretical, experimental and computational study has been conducted in the course of this work focusing on current extraction of short-circuited solar cells under ultrashort pulse excitation [33]. In this work, special attention is given to potential deviations in the extracted current for pulsed versus continuous illumination. As it is aimed for highest measurement accuracy, the analysis of these potentially occurring deviations is of tremendous importance for the remainder of this work.

The chapter is organized as follows: firstly, a theoretical model describing the interaction of ultrashort laser pulses and solar cells regarding their short circuit current density $(J_{\rm SC})$ is derived. Secondly, experimental evidence for the nonlinear contributions that are expected from the theoretical model will be presented. Afterwards, the experimental results are used to validate a computational approach for solving the differential equations of the theoretical model. Finally, the computational approach is applied to conduct the most prominent, but experimentally hardly feasible, comparison of pulsed versus continuous illumination regarding the generated $J_{\rm SC}$ in a silicon (Si) solar cell.

4.2 Modelling of Solar Cell Response Under Pulsed Irradiation

4.2.1 J_{SC} in Terms of Excess Carrier Density

The applicability of ultrashort laser pulses for highly accurate electrical characterization of solar cells is evaluated by analyzing the potential deviations in the $J_{\rm SC}$ for pulsed versus continuous illumination. The $J_{\rm SC}$ is a favorable photovoltaic parameter for this evaluation as it is of major importance regarding solar cell calibration (see Section 2.2).

For a theoretical modeling of the $J_{\rm SC}$ its proportionality to the current flow across the device junction can be exploited. This current flow can be modeled by the gradient of excess charge carrier density $\Delta n(z,t)$ across the device junction (see Section 2.1.2.2), thus,

$$J_{\rm SC} \propto \left. \frac{\mathrm{d}\Delta n}{\mathrm{d}z} \right|_{\substack{z=0\\S_0 \to \infty}} \tag{4.1}$$

for front junction solar cells with a front surface recombination velocity S_0 . The short circuit conditions are modeled by strongest current extraction taking place for S_0 approaching infinity. Every minority charge carrier reaching the interface (here *pn*-junction at $z \approx 0$) is immediately extracted. It is noteworthy, that the subsequently presented results are limited to $J_{\rm SC}$ ratios deduced from the comparison of different illumination characteristics. Therefore, no absolute $J_{\rm SC}$ values need to be determined and the proportionality given in Eq. (4.1) is sufficient.

4.2.2 Coupled Differential Equations for Radiation-Semiconductor Interaction

For a theoretical modeling of the excess charge carrier density $\Delta n(z,t)$ induced by ultrashort laser pulses a set of coupled differential equation is derived in this work. The spatio-temporal evolution of $\Delta n(z,t)$ is described by the well-known one-dimensional continuity equation (see also Section 2.1.2.1)

$$\frac{\partial \Delta n\left(z,t\right)}{\partial t} = D_{\rm a} \frac{\partial^2 \Delta n\left(z,t\right)}{\partial z^2} + G\left(z,t\right) - R\left(z,t\right). \tag{4.2}$$

The continuity equation takes into account diffusion processes via an ambipolar diffusion coefficient D_a as well as excess carrier generation rate G(z,t) and recombination rate R(z,t). The recombination rate might also be expressed as $R = \Delta n(z,t) / \tau (\Delta n(z,t))$ with injection-dependent minority carrier lifetime $\tau (\Delta n(z,t))$. Although a maximum number of excess charge carriers is extracted under short circuit conditions, recombination may still compete with carrier extraction through solar cell terminals.

In contrast to the typical notation of the generation rate $G(z,t) = \alpha_{\rm bb} \Phi_0(t) \exp^{-\alpha_{\rm bb} z}$, with band-to-band absorption coefficient $\alpha_{\rm bb}$ and initial photon flux⁴ Φ_0 , G(z,t) is extended to

$$G(z,t) = \left(\sigma_{\alpha} \left(N_0 - \Delta n\left(z,t\right)\right) + \sigma_{\beta} \left(N_0 - \Delta n\left(z,t\right)\right) \frac{I(z,t)}{2}\right) \frac{I(z,t)}{h\nu}$$

$$(4.3)$$

in this approach. Firstly, the absorption coefficient $\alpha_{\rm bb}$ is replaced by the product $\sigma_{\alpha} (N_0 - \Delta n (z, t))$ in Eq. (4.3), with linear band-to-band absorption cross section σ_{α} given in cm² and a maximum excess carrier density N_0 in cm⁻³. This takes into account a possible saturation of absorption for an excess carrier density $\Delta n (z, t)$ approaching N_0 . The second term considers two-photon absorption (TPA), a process that excites a single

 $^{^{4}}Photon flux$ denotes the number of photons incident per second and unit area. As the photon flux quantizes optical intensity or power to a package of photons, it serves clarity of many photon-matter interaction concepts.

electron-hole pair by the virtually simultaneous interaction of a carrier with two photons (see also Section 3.2.2.3). The corresponding nonlinear absorption cross section σ_{β} is given in cm⁴W⁻¹. The probability for TPA increases quadratically with the applied optical intensity I(z,t) that is given in Wcm⁻² and used throughout this chapter to improve clarity of the later discussions.⁵ Finally, the factor 1/2 in the nonlinear absorption term of Eq. (4.3) accounts for generation of a single electron-hole pair by the energy of two photons. It is noteworthy, that light trapping effects can be neglected in the generation term of this work as only $J_{\rm SC}$ or Δn ratios are considered for this analysis.⁶

As any generated carrier is naturally associated with a reduction in photon flux or optical intensity, I(z,t) varies while the radiation propagates through the semiconductor sample in z-direction. This circumstance can be expressed by the differential equation [56]

$$\frac{\partial I(z,t)}{\partial z} = -\left[\sigma_{\alpha} \left(N_{0} - \Delta n(z,t)\right) + \sigma_{\text{FCA}} \left(n(z,t) + p(z,t)\right)\right] I(z,t) - \sigma_{\beta} \left(N_{0} - \Delta n(z,t)\right) I^{2}(z,t) .$$

$$(4.4)$$

In addition to the previously introduced cross sections for linear and nonlinear band-to-band absorption, the free-carrier absorption (FCA) cross section σ_{FCA} appears in Eq. (4.4) that accounts for a reduction in I(z,t) resulting from the interaction of photons with already excited charge carriers [57, 58]. Therefore, the FCA cross section σ_{FCA} is multiplied by the sum of all free carriers $(n(z,t)+p(z,t) = N_{\text{D}}+N_{\text{A}}+2\Delta n(z,t),$ with N_{D} and N_{A} as semiconductor doping densities). As no additional excess charge carrier is excited by this process, no equivalent term occurs in Eq. (4.3).

⁵Dividing I(z,t) by the photon energy $h\nu$, with Planck constant h and radiation frequency ν , yields the photon flux Φ_0 .

 $^{^{6}}$ In Appendix C a detailed discussion on the negligence of light trapping in the course of this chapter can be found.

4.2.3 Reduction of the Set of Differential Equations

The high peak power of ultrashort laser pulses is associated with extreme optical intensities on short time scales followed by comparably long time durations of (almost) zero intensity. Considering several contributions that arise from this strong temporal confinement of the overall intensity, Eqs. (4.2) to (4.4) form a set of coupled differential equations that allows for an analysis of the interaction of ultrashort laser pulses and semiconductors. Nevertheless, a closer examination of the individual contributions reveals that some of them can be neglected for the further discussion. In this section several approximations will be presented that significantly simplify the set of differential equations.

Injection Dependence of Minority Carrier Lifetime au

The strong temporal confinement of photons in an ultrashort laser pulse hitting a solar cell at $t = t_0$ yields an intrinsic variation of $\Delta n(z,t)$ within a pulse period $T = t_1 - t_0$ (see Fig. 4.1a). With an injection-dependent minority carrier lifetime $\tau (\Delta n(z,t))$ this results in an effective lifetime, evaluated over an entire pulse period, that might differ significantly from the lifetime under continuous illumination. Consequently, deviations in the measured effective short circuit current may arise. However, if the deviation in $\Delta n(z,t)$, that is being caused by the injection dependence of τ (denoted as δ_{1-2}), is significantly smaller than the overall change of $\Delta n(z,t)$ within a pulse period (δ_0), the injection dependence of τ can be neglected. Thus, $\delta_{1-2} \ll \delta_0$ has to be valid for negligence of injection dependence of τ , which is equivalent to

$$\left|\exp\left(-\frac{T}{\tau_1}\right) - \exp\left(-\frac{T}{\tau_2}\right)\right| \ll 1 - \exp\left(-\frac{T}{\tau_1}\right).$$
(4.5)

Assuming that $\tau_{1,2} \gg T$ the exponential terms can be linearized and Eq. (4.5) can be reduced to

$$\left|\frac{\tau_1 - \tau_2}{\tau_2}\right| \ll 1. \tag{4.6}$$



Figure 4.1: (a) Illustration of the impact of different lifetimes τ_1 and τ_2 on excess carrier density evolution within a pulse period $T = t_1 - t_0$. (b) $(\tau_1 - \tau_2) / \tau_2$ plotted over the ratio τ_2 / τ_1 . For Eq. (4.6) being valid a ratio τ_2 / τ_1 close to unity is required.

Plotting $(\tau_1 - \tau_2)/\tau_2$ over the ratio τ_2/τ_1 (see Fig. 4.1b) emphasizes the requirement of $\tau_1 \approx \tau_2$ for validity of $\delta_{1-2} \ll \delta_0$. With above's basic assumption $\tau_{1,2} \gg T$, a generally very small change in δ_0 is obtained. As this is intrinsically tied to small variations in τ , $\tau_1 \approx \tau_2$ is readily achieved.

Thus, if the formulated criteria $\tau \gg T$ holds, negligence of injection dependence of τ is valid even under open circuit conditions, where injectiondependent recombination is much more relevant than under short circuit conditions. For indirect semiconductors like Si with typical lifetimes in the μ s or even ms regime [59], ultrashort pulse oscillators with repetition rates in the range of several tens of MHz readily fulfill $\tau \gg T$.

In contrast to this, much shorter minority carrier lifetimes are present in direct semiconductors due to the significantly increased radiative decay (e.g. in the ns to μ s regime for gallium arsenide (GaAs) [60]). As $\tau \gg T$ in such cases, a weaker approximation has to be applied that takes into account the short circuit condition provided that substantial injection dependence of minority carrier lifetime cannot be ruled out. With this precondition the transit time τ_{trans} can be introduced as a maximum average lifetime. τ_{trans} describes the average time it takes for a charge carrier to reach the opposite side of a device. Thus, after [20, pp. 240-241]

$$\tau_{\rm trans} = \frac{1}{D} \left(\frac{W^2}{2} - \frac{1}{\alpha_{\rm bb}^2} \right) \tag{4.7}$$

excited charge carriers will have passed the device junction and will have been electrically extracted from the device.

In Fig. 4.2a $\tau_{\rm trans}$ is plotted over device thickness W for various diffusion coefficients. These transit times represent a most extreme assumption for the maximum average lifetime of minority carriers under short circuit conditions as an extremely shallow⁷ generation profile is assumed ($\alpha_{\rm bb}W \gg 1$). As long as $\tau_{\rm trans} \ll \tau$, the carrier lifetime is virtually limited by the injection-independent transit time $\tau_{\rm trans}$, thus, achieving injection-independence in the overall carrier lifetime⁸ $(1/\tau_{\rm sum} = 1/\tau + 1/\tau_{\rm trans} \approx 1/\tau_{\rm trans})$.

Saturation in Carrier Generation

Owing to the constant lifetime approximation reasoned above, identical effective lifetimes for pulsed and continuous illumination can be assumed. However, the temporally averaged excess carrier density $1/T \int_0^T \Delta n (z, t) dt$ might still differ for the two illumination cases, if the virtually instantaneous excitation of charge carriers by ultrashort pulses (schematically shown in Fig. 4.1a) temporary violates the approximation $\Delta n \ll N_0$.

For an estimation of such an impact the additional carriers induced within an ultrashort pulse period are assumed to be excited instantaneously and non-depth averaged. The results of this most conservative treatment for a Si and GaAs sample are shown in Fig. 4.2b. In this first order estimation N_0 is approximated by the effective carrier density of states (DOS) in the conduction bands. From Fig. 4.2b it is apparent that

 $^{^7}Shallow$ denotes an excitation of excess charge carriers close to the front surface of a device or wafer.

⁸Please note that this is a rather untypical notation as τ_{trans} does not describe a recombination being associated with $R = \Delta n (z, t) / \tau$. Instead, the given equation rather illustrates the role of τ_{trans} in comparison with τ 's resulting from recombination effects.



Figure 4.2: (a) Excess carrier transit times $\tau_{\rm trans}$ over device thickness for various diffusion coefficients D. The resulting transit times are a maximum assumption for the average lifetimes of minority carriers under short circuit conditions, as very shallow excitation conditions at the opposite side of the device junction are assumed. (b) Nondepth averaged linear carrier generation in Si and GaAs by ultrashort laser pulses. The plotted values correspond to the additional dose of excess charge carriers induced by a single ultrashort pulse with a given average intensity $I_{\rm av}$ at 12.5 ns pulse period. The energy of the exciting photons is normalized by the band gap energies of Si $(E_{\rm g} = 1.12 \text{ eV})$ and GaAs $(E_{\rm g} = 1.424 \text{ eV})$, respectively. Linear absorption data from [61] (Si) and [62] (GaAs).

the validity of approximating $N_0 - \Delta n (z, t) \approx N_0$ depends on both photon energy and illumination intensity, but holds true for broad spectral ranges and illumination intensities. Thus, the validity of this approximation depends on the applied radiation source and will be evaluated individually for the specific situations considered in the later part of this chapter.

Reduced Set of Differential Equations

If the previously discussed approximations regarding minority carrier lifetime and saturation of carrier generation are valid, the set of differential equations (Eqs. (4.2) and (4.4)) can be simplified to

$$\frac{\partial \Delta n\left(z,t\right)}{\partial t} = D_{a} \frac{\partial^{2} \Delta n\left(z,t\right)}{\partial z^{2}} + \left(\alpha_{bb} + \beta \frac{I\left(z,t\right)}{2}\right) \frac{I\left(z,t\right)}{h\nu} - \frac{\Delta n\left(z,t\right)}{\tau}$$
(4.8a)
$$\frac{\partial I\left(z,t\right)}{\partial z} = -\left[\alpha_{bb} + \sigma_{FCA}\left(n\left(z,t\right) + p\left(z,t\right)\right)\right] I\left(z,t\right) - \beta I^{2}\left(z,t\right)$$
(4.8b)

with $\alpha_{\rm bb} = \sigma_{\alpha} N_0 \approx \sigma_{\alpha} \left(N_0 - \Delta n \left(z, t \right) \right), \ \beta = \sigma_{\beta} N_0 \approx \sigma_{\beta} \left(N_0 - \Delta n \left(z, t \right) \right)$ and constant minority carrier lifetime τ .

4.2.4 Quasi-Steady-State Approximation

Owing to the previously discussed approximations and the accompanying simplifications resulting in the above set of differential equations (Eq. (4.8)), free carrier absorption (FCA) is the only term left that causes coupling of Eqs. (4.8a) and (4.8b). If significant changes in $\Delta n (z, t)$ take place at time scales of a pulse transit time through the sample, these changes affect the pulse propagation itself and require an alternating step-wise computation of Eqs. (4.8a) and (4.8b).

As this coupling involves a tremendous computational effort, a quasisteady-state approximation (QSS-approximation) is introduced that assumes invariance of $\Delta n (z, t)$ for each single pulse, thereby yielding a decoupling of Eqs. (4.8a) and (4.8b). The two fundamental constraints of this approximation can be stated as follows: firstly, carrier motion within a pulse transit time through the sample is negligible and, secondly, any variations in $\Delta n (z, t)$ that are induced by a single ultrashort pulse do not affect the intensity of the pulse itself. Hence, each pulse incident on the semiconductor sample interacts with a previously known excess carrier distribution $\Delta n (z, t)$ that is invariant for the pulse itself.⁹ If this QSS-approximation holds, n (z, t) and, likewise, p (z, t) in Eq. (4.8b) become temporally invariant for the time the pulse propagates through the

⁹Naturally the pulse induces changes in Δn , however, they are not *seen* by the pulse itself but by all subsequent pulses.
semiconductor. Consequently, Eq. (4.8b) can be directly solved and applied to Eq. (4.8a) resulting in a single differential equation describing the interaction of ultrashort pulses and semiconductors.

For the QSS-approximation being a valid assumption, the following two conditions have to hold:

- 1. Carrier redistribution due to diffusion within a pulse transit time is negligible.
- 2. Either the relative change in FCA induced by a single pulse or total FCA itself, as compared to band-to-band absorption, are negligible.

The first condition is readily achieved when considering typical diffusion coefficients and solar cell thicknesses (note, that the discussion is limited to one spatial dimension). Charge carriers move randomly while a pulse propagates through a sample. From the pulse transit time $\tau_{\rm p}$ through the device and the diffusion coefficient D the characteristic distance of this random motion can be determined as $\sqrt{\tau_{\rm p}D}$ (see Fig. 4.3a). For typical diffusion coefficients and device thicknesses this diffusion length is less than 0.1 μ m, thus, even less than typical spatial discretizations in numerical approaches. Therefore, significant carrier redistribution can be readily neglected for the time scale of a pulse transit through a semiconductor sample.

The second condition requires two effects to be considered: firstly, the variation in FCA induced by a single ultrashort pulse (denoted as $\delta \alpha_{\rm FCA}$) and, secondly, the impact of FCA relative to the band-to-band absorption processes. Starting with $\delta \alpha_{\rm FCA}$, FCA itself is defined by [57, 58, 63]:

$$\alpha_{\rm FCA} = \sigma_{\rm FCA}^{\rm n} \left(\lambda/\mu {\rm m}\right)^{x_{\rm n}} n + \sigma_{\rm FCA}^{\rm p} \left(\lambda/\mu {\rm m}\right)^{x_{\rm p}} p, \qquad (4.9)$$

with FCA cross sections $\sigma_{\text{FCA}}^{n,p}$ and wavelength powers $x_{n,p}$ for electrons $(n = N_{\text{D}} + \Delta n)$ and holes $(p = N_{\text{A}} + \Delta n)$. As n and p depend on $\Delta n(z, t)$, knowledge of the change in $\Delta n(z, t)$ induced by a single pulse (denoted as $\delta \Delta n$) is required for computation of $\delta \alpha_{\text{FCA}}$. From Eq. (4.8a), $\delta \Delta n$ can



Figure 4.3: (a) Diffusion length of charge carrier movement over device thickness for various diffusion coefficients. The considered time frame for the statistical movement is the time it takes for an ultrashort pulse to travel through a device of respective thickness. (b) Ratio of FCA to band-to-band absorption $(\alpha_{\rm FCA}/(\alpha_{\rm bb} + \beta I_{\rm peak}))$ over change in FCA by a single pulse $(\delta \alpha_{\rm FCA})$ for various average illumination intensities $I_{\rm av}$, normalized photon energies $E_{\rm ph}/E_{\rm g}$ and initial excess carrier distributions Δn_i in the *n*-type Si sample with $N_{\rm D} = 5 \cdot 10^{15} \,{\rm cm}^{-3}$. The assumed ultrashort pulses have a pulse duration of $\tau_{\rm FWHM} = 100$ fs and are emitted at a pulse period of T = 12.5 ns.

be described as^{10}

$$\delta\Delta n \approx \left(\alpha_{\rm bb} \frac{I_{\rm peak}}{h\nu} + \beta \frac{I_{\rm peak}^2}{2h\nu} - \frac{\Delta n_{\rm i}}{\tau}\right) \tau_{1/{\rm e}^2},\tag{4.10}$$

with $I_{\text{peak}} = I_{\text{av}} / (f_{\text{rep}}\tau_{1/e^2})$ as a rectangular equivalent to a Gaussianshaped ultrashort pulse peak intensity with pulse repetition rate $f_{\text{rep}} = 1/T$ and pulse duration τ_{1/e^2} (discussed in Appendix C on page 219). Δn_i denotes the excess carrier density in the semiconductor immediately before the considered ultrashort pulse hits. With Eqs. (4.9) and (4.10)

¹⁰For simplicity, an infinitely thin semiconductor is assumed here. This makes any consideration of diffusion or depth-averaging of carrier generation redundant. Also, this represents a very conservative treatment as $\delta \alpha_{\rm FCA}$ is proportional to $\delta \Delta n$ (see Eq. (4.11)), which is most pronounced for non-depth-averaged excess carrier generation.

 $\delta \alpha_{\rm FCA}$ is given by

$$\delta \alpha_{\rm FCA} = \frac{\left(\sigma_{\rm FCA}^{\rm n} \left(\lambda/\mu m\right)^{x_{\rm n}} + \sigma_{\rm FCA}^{\rm p} \left(\lambda/\mu m\right)^{x_{\rm p}}\right) \delta \Delta n}{\sigma_{\rm FCA}^{\rm n} \left(\lambda/\mu m\right)^{x_{\rm n}} \left(N_{\rm D} + \Delta n_i\right) + \sigma_{\rm FCA}^{\rm p} \left(\lambda/\mu m\right)^{x_{\rm p}} \left(N_{\rm A} + \Delta n_i\right)}.$$
(4.11)

For FCA having a significant impact on the set of differential equations given in Eq. (4.8), not only the induced change of FCA ($\delta \alpha_{\rm FCA}$) but also the FCA itself must be of significant magnitude. In Fig. 4.3b $\alpha_{\rm FCA}/(\alpha_{\rm bb} + \beta I_{\rm peak})$ is plotted over $\delta \alpha_{\rm FCA}$ for an idealized *n*-type Si sample ($N_{\rm D} = 5 \cdot 10^{15} {\rm cm}^{-3}$) illuminated with ultrashort pulses of $\tau_{\rm FWHM} =$ 100 fs pulse duration and $f = 1/T = 80 {\rm MHz}$ pulse repetition rate. In this idealized consideration no defect-states and, thus, no SRH-recombinations occur. Therefore, the minority carrier lifetime τ is purely intrinsically limited (including Auger and radiative recombination) and approximated from the parametrization published by *Richter* et al. in [22] using $\Delta n_{\rm i}$.

In contrast to the marginal influence of the initial excess carrier density $\Delta n_{\rm i}$ (given as solid and dotted/dashed lines of same color) the average illumination intensity $I_{\rm av}$ affects $\delta \alpha_{\rm FCA}$ significantly, hence, shifting the lines to the right (see highlighted points in Fig. 4.3b, where identical symbols denote specific photon energies). Furthermore, there is a quite distinct dependence on the energy of the incident photons. Those photons, that are close to and below the band gap energy ($E_{\rm ph}/E_{\rm g} \approx 1$) are most likely to violate the QSS-approximation. This results from a continuous reduction in band-to-band absorption with longer wavelengths, while there are still excess carriers being excited giving rise to significance of $\delta \alpha_{\rm FCA}$.¹¹ However, for $I_{\rm av} \leq 100 \,{\rm Wcm}^{-2}$ the presented approximation is working quite well as either $\delta \alpha_{\rm FCA}$ or $\alpha_{\rm FCA}/(\alpha_{\rm bb} + \beta I_{\rm peak})$ is less than 1%.

If the QSS-approximation discussed above is considered to be valid, any incident pulse interacts with a sample of known excess carrier density $\Delta n(z,t)$ and changes induced by the pulse do not significantly affect the pulse itself. Thus, there is no relevant time-dependency in $\Delta n(z,t)$ when calculating the intensity I(z,t) of a single pulse and $\Delta n(z,t) \rightarrow \Delta n(z)$.

¹¹Please note that a reconsideration of this argumentation might be necessary for extreme pulse peak powers I_{peak} as nonlinear absorption might outperform the linear band-to-band absorption.

Consequently, Eq. (4.8b) can be solved directly by

$$I(z,t) = \frac{I(t)\alpha_{\text{tot}}(z)\exp\left(-\alpha_{\text{tot}}(z)z\right)}{\alpha_{\text{tot}}(z) + \beta I(t)\left(1 - \exp\left(-\alpha_{\text{tot}}(z)z\right)\right)},$$
(4.12)

with $\alpha_{\text{tot}}(z) = \alpha_{\text{bb}} + \sigma_{\text{FCA}}^{n} (\lambda/\mu m)^{x_{n}} n(z) + \sigma_{\text{FCA}}^{p} (\lambda/\mu m)^{x_{p}} p(z).$

4.2.5 Final Modeling Equations

In Sections 4.2.3 and 4.2.4 several approximations were detailed that yield significant simplifications of the theoretical model describing the interaction of ultrashort laser pulses and semiconductors. In this section, the approximations and the resulting set of equations are recapitulated very briefly, starting from the most general set of differential equations

$$\frac{\partial \Delta n\left(z,t\right)}{\partial t} = D_{a} \frac{\partial^{2} \Delta n\left(z,t\right)}{\partial z^{2}} - \frac{\Delta n\left(z,t\right)}{\tau\left(\Delta n\left(z,t\right)\right)} + \frac{I\left(z,t\right)}{h\nu} \\
\cdot \left(\sigma_{\alpha}\left(N_{0} - \Delta n\left(z,t\right)\right) + \sigma_{\beta}\left(N_{0} - \Delta n\left(z,t\right)\right) \frac{I\left(z,t\right)}{2}\right) \\
\frac{\partial I\left(z,t\right)}{\partial z} = -\left[\sigma_{\alpha}\left(N_{0} - \Delta n\left(z,t\right)\right) + \sigma_{\text{FCA}}\left(n\left(z,t\right) + p\left(z,t\right)\right)\right] I\left(z,t\right) \\
- \sigma_{\beta}\left(N_{0} - \Delta n\left(z,t\right)\right) I^{2}\left(z,t\right).$$
(4.13)

If $N_0 \gg \Delta n(z,t)$ and if the injection dependence of $\tau (\Delta n(z,t))$ can be neglected, Eq. (4.13) can be reduced to

$$\frac{\partial \Delta n\left(z,t\right)}{\partial t} = D_{a} \frac{\partial^{2} \Delta n\left(z,t\right)}{\partial z^{2}} - \frac{\Delta n\left(z,t\right)}{\tau} + \frac{I\left(z,t\right)}{h\nu} \left(\alpha_{bb} + \beta \frac{I\left(z,t\right)}{2}\right) \\
\frac{\partial I\left(z,t\right)}{\partial z} = -\left[\alpha_{bb} + \sigma_{FCA}\left(n\left(z,t\right) + p\left(z,t\right)\right)\right] I\left(z,t\right) - \beta I^{2}\left(z,t\right).$$
(4.14)

Finally, if previously given conditions hold and the QSS-approximation

is valid, the equations can be combined to

$$\frac{\partial \Delta n\left(z,t\right)}{\partial t} = D_{a} \frac{\partial^{2} \Delta n\left(z,t\right)}{\partial z^{2}} - \frac{\Delta n\left(z,t\right)}{\tau} + \frac{I\left(t\right) \alpha_{\text{tot}}\left(z\right) \exp\left(-\alpha_{\text{tot}}\left(z\right)z\right)}{h\nu\left[\alpha_{\text{tot}}\left(z\right) + \beta I\left(t\right)\left(1 - \exp\left(-\alpha_{\text{tot}}\left(z\right)z\right)\right)\right]} \\ \cdot \left(\alpha_{\text{bb}} + \frac{\beta I\left(t\right) \alpha_{\text{tot}}\left(z\right) \exp\left(-\alpha_{\text{tot}}\left(z\right)z\right)}{2\left[\alpha_{\text{tot}}\left(z\right) + \beta I\left(t\right)\left(1 - \exp\left(-\alpha_{\text{tot}}\left(z\right)z\right)\right)\right]}\right), \tag{4.15}$$

with $\alpha_{\text{tot}}(z) = \alpha_{\text{bb}} + \sigma_{\text{FCA}}^{n} (\lambda/\mu \text{m})^{x_{\text{n}}} n(z) + \sigma_{\text{FCA}}^{p} (\lambda/\mu \text{m})^{x_{\text{p}}} p(z).$

In conclusion, it has to be emphasized that in contrast to saturation and injection dependence of lifetime, excess charge carrier generation by TPA cannot be readily neglected and remains as nonlinear contribution in Eqs. (4.13) to (4.15).

4.3 Experimental Study of TPA Impact on $J_{\rm SC}$

The previous discussion on the interaction of ultrashort laser pulses and solar cells was concluded by the statement that carrier generation by TPA remains as the only nonlinear contribution in the theoretical model, regardless of all given approximations. Thus, for finding experimental evidence of deviations in short circuit current density $(J_{\rm SC})$ when comparing pulsed and continuous illumination, it is favorable to focus on that effect.

Although the theoretical framework is generally valid for any semiconductor material, the experimental analysis of TPA has been limited to a Si solar cell. In the following the experimental approach and the measurement outcomes will be discussed along the lines of a previous publication by the author of this work [33].

4.3.1 Linear Versus Nonlinear Carrier Generation

In order to identify potential spectral regions and illumination intensities of significant TPA, nonlinear and linear excess carrier generation are compared in analogy to Eq. (C.6) with $I = I_{\text{peak}}$ (for a definition of I_{peak} see

4.3 Experimental Study of TPA Impact on $J_{ m SC}$



Figure 4.4: Ratio of nonlinear to linear carrier generation rates in intrinsic silicon for various wavelengths and average illumination intensities. The ultrashort pulses assumed here have pulse durations of $\tau_{\rm FWHM} = 100$ fs at 80 MHz pulse repetition rate.

Eq. (C.7) on page 219). Results of this analysis for intrinsic silicon are shown in Fig. 4.4, with $\tau_{\rm FWHM} = 100$ fs, $f_{\rm rep} = 80$ MHz and material data from *Bristow* et al. [55] and *Green* [61]. The results demonstrate that evidence of an impact of nonlinear absorption should be examined in the vicinity of the band edge of Si, if functionality of the QSS-approximation is eligible for later computations (thus, $I_{\rm av} < 100 \,{\rm Wcm}^{-2}$, see Section 4.2.4).

4.3.2 Experimental Setup

4.3.2.1 Laser Source

For achieving ultrashort laser pulses in the vicinity of the silicon band edge, an optical parametric oscillator (OPO) (*Inspire*/Newport Spectra-Physics) pumped by the frequency-doubled radiation of a titaniumsapphire ultrashort pulse laser (*MaiTai*/Newport Spectra-Physics) is used. The OPO emits ultrashort pulses spectrally tunable between 900 nm and 2500 nm, thereby covering the important spectral regions for demonstration of TPA impact in current generation of solar cells. In Fig. 4.5 an autocorrelation trace of an ultrashort pulse emitted by the OPO at 1000 nm center wavelength is shown. The full-width-at-halfmaximum (FWHM) of the Gaussian fit to the measured trace equals



Figure 4.5: Autocorrelation trace of an ultrashort pulse emitted by the OPO at 1000 nm center wavelength. The width of the Gaussian fit (red line) equals 223 fs, thus, the FWHM duration of the pulse equals $\tau_{\rm FWHM} = 223 \, {\rm fs}/\sqrt{2} \approx 158 \, {\rm fs}.$

 $\tau_{\rm AC} = 223$ fs, hence, the pulse duration is $\tau_{\rm FWHM} = 223 \, {\rm fs}/\sqrt{2} \approx 158$ fs. Pulse duration measurements have been carried out from 900 nm to 1100 nm (upper sensitivity threshold of the applied autocorrelator). Although $\tau_{\rm FWHM}$ slightly fluctuates within a range of approximately ± 10 fs when scanning the OPO wavelengths, no distinct wavelength dependency was observed. Thus, the pulse duration of any OPO wavelength output is approximated by $\tau_{\rm FWHM} \approx 160 \, {\rm fs}$.

4.3.2.2 Double-Ring-Resonator Setup

The contribution of TPA to the excess carrier generation in solar cells via ultrashort laser pulses is directly related to the temporal shape of these pulses, especially to their pulse peak intensity (see e.g. Eq. (4.10)). Hence, experimental evidence of a significant TPA-contribution to the $J_{\rm SC}$ induced by an ultrashort laser pulse can be deduced from two measurements with deviating temporal shape and otherwise identical characteristics (e.g. average intensity, wavelength, geometry, direction etc.). If this condition is fulfilled, any variations in the measured $J_{\rm SC}$ between the two measurements can be attributed to TPA.

For this purpose an experimental setup with two ring cavities has been



Figure 4.6: (a) Schematical drawing of the experimental setup for temporal shaping of ultrashort laser pulses by two ring cavities prior to their interaction with a target solar cell. Monitor diode, autocorrelator and camera are used to check the functionality of the experimental setup. Published in [33]. (b) Computed temporal pulse shapes emitted by the experimental setup representing characteristic situations for temporal alignment of the cavities using the translation stages: both cavities are aligned to half of the OPO pulse period (top, green, $\Delta t_{\rm ring}^{1,2} = T/2$); both cavities are identically misaligned (middle, blue, $\Delta t_{\rm ring}^1 = \Delta t_{\rm ring}^2 \neq T/2$); both cavities are differently misaligned (bottom, red, $\Delta t_{\rm ring}^1 \neq \Delta t_{\rm ring}^2 \neq T/2$). The horizontal axis is normalized to the OPO pulse period T, the vertical axis to the peak power of the original ultrashort pulses.

developed that is schematically shown in Fig. 4.6a. Incident ultrashort pulses from the OPO are partially transmitted and reflected at a beam splitter. The transmitted beam is delayed with respect to its reflected complement by the roundtrip time $\Delta t_{\rm ring}$ of a first ring cavity. After this roundtrip, the pulse is again partially transmitted and reflected. The cavity roundtrip time can be adjusted by a translation stage that varies the cavity length. If the cavity roundtrip time is adjusted to match half of the OPO pulse period ($\Delta t_{\rm ring} = T/2$), any incident pulse that completed two cavity roundtrips will temporally coincide with the subsequently emitted OPO pulse ($2\Delta t_{\rm ring} = T$), thereby doubling the pulse repetition rate of the pulse train emitted by the OPO ($f = 1/\Delta t_{\rm ring} = 2/T = 2f_{\rm rep}$). By choosing an appropriate beam splitter ratio the respective pulses emitted by the first ring cavity can be equalized in their amplitudes.

As the pulses emitted from the first ring cavity consist of multiple temporally overlapping pulses, any change to the cavity length induces a distortion of the resulting temporal pulse shapes. In order to increase the effect of pulse shape distortion an identical second ring cavity is implemented into the setup. In Fig. 4.6b computed temporal shapes emitted by the experimental setup are shown for three characteristic cases: firstly, both cavity roundtrip times are adjusted to half of the OPO pulse period (top, green, $\Delta t_{\rm ring}^{1,2} = T/2$); secondly, both cavity lengths are equally misaligned (middle, blue, $\Delta t_{\rm ring}^1 = \Delta t_{\rm ring}^2 \neq T/2$); thirdly, both cavity lengths are differently misaligned (bottom, red, $\Delta t_{\rm ring}^1 \neq \Delta t_{\rm ring}^2 \neq T/2$). From the different pulse peak intensities a variation in $J_{\rm SC}$ is expected if there is a significant contribution of TPA.

The experimental setup is completed by an optical lens that focuses the pulse train onto the test cell, thereby reaching average illumination intensities in the range of several tens of Wcm⁻², as well as a chopper wheel to enable highly sensitive differential $J_{\rm SC}$ measurements. Transimpedance and lock-in amplifiers (not shown in Fig. 4.6a) are used to keep the target cell in short circuit conditions and capture the differential current increase induced by the chopped ultrashort pulses.

For checking the geometrical alignment of the experimental setup one portion of the focused beam is sampled onto a CMOS-camera. Furthermore, a flip mirror can be implemented for a convenient verification of temporal cavity alignment using an autocorrelator. Finally, a monitor diode is used to track laser intensity fluctuations.

In Fig. 4.7 autocorrelation traces for different temporal alignments of the experimental setup are shown. To the left the cavities are temporally misaligned resulting in various pulses measured with the autocorrelator.



Figure 4.7: Autocorrelation traces for different temporal alignment configurations from misaligned (left) to aligned (right), denoted by the arrow on top. The autocorrelation signal merges from multiple pulses into a single pulse when the cavities are aligned. The insets show camera images of the focused pulses for the misaligned and the aligned configuration (after [33]).

When aligning the cavities (from left to right, indicated by the arrow on top) the pulses merge into a single pulse. Furthermore, two insets show camera images of the focused ultrashort laser pulses for temporally misaligned (left) and aligned (right) cavity lengths. Besides the apparently good geometrical alignment, as the individual pulses overlap to a single spot that is close to a Gaussian, fringes appear when the cavities are temporally aligned (note, that they are not appearing in the misaligned case). These are interference fringes resulting from the temporal overlap of multiple pulses and are a strong evidence for a neat temporal alignment of the setup.¹² Likewise, the significant increase of the background noise in the most right autocorrelation trace can be attributed to interference

¹²The temporal coherence of ultrashort laser pulses, as a measure for their ability to interfere with each other, results from the mode-locking principle that can be envisioned by a single pulse oscillating in the laser cavity. Although the coherence function decays with the number of considered pulse periods, a distinct fringe visibility is expected for the closely spaced pulses interfering in this case [64].

(note the small but distinct peaks next to the main pulse that disappear in the misaligned autocorrelation traces).

4.3.3 Experimental Results

The experimental setup presented and validated in the previous section has been applied to measure $J_{\rm SC}$ s of an *n*-type Si solar cell of 200 μ m thickness (doping concentration $N_{\rm D} = 5 \cdot 10^{15} \,\mathrm{cm}^{-3}$). The test cell has been illuminated with a spot size diameter of approximately 150 μ m (estimated from camera images) and an average power of 5 to 20 mW.

The resulting J_{SC} at a single wavelength, but for varying cavity alignments, is shown in Fig. 4.8a. The result clearly demonstrates a dependency of the measurement outcome on the temporal pulse shapes shown in the inset and indicates an impact of carrier generation due to nonlinear



Figure 4.8: (a) Variations in the measured $J_{\rm SC}$ for varying cavity alignments and resulting temporal pulse shapes shown in the inset (compare Fig. 4.6b). (b) Measured $J_{\rm SC}$ for aligned (green triangles) and misaligned cavities (red squares) over wavelength. The lines denote linear (solid) and nonlinear absorption (TPA, dashed) in Si with data from [55, 61]. The nonlinear absorption has been computed by $\beta I_{\rm peak} t_{\rm fac}/2$ with $I_{\rm peak}$ from Eq. (C.7) and $\tau_{\rm FWHM} = 160$ fs, $f_{\rm rep} = 160$ MHz, $I_{\rm av} = 20$ mW/ π (75 μ m)² and $t_{\rm fac} = \tau_{\rm FWHM} f_{\rm rep}$ which approximately reflects that conditions for temporally aligned ring cavities. Published in [33].

absorption. Furthermore, both robustness and reproducibility of the experimental approach are evident from the presented results as a constant $J_{\rm SC}$ is measured while the cavities remain unchanged and as previous $J_{\rm SC}$ values can be accurately restored.

In Fig. 4.8b measured $J_{\rm SC}$ s for the aligned (green triangles) and misaligned (red squares) cavity configuration are shown over a wide range of wavelengths. Also, linear (solid line) and nonlinear absorption coefficients (dashed line) are shown (with data from [55, 61]). The misaligned configuration yields consistently less $J_{\rm SC}$ than the aligned configuration, pointing at nonlinear contributions to the $J_{\rm SC}$. Furthermore, the relative trend of the $J_{\rm SC}$ over wavelength changes from a steep to a rather flat reduction in the measured $J_{\rm SC}$, thus, imitating the wavelength dependencies of linear (for shorter wavelengths, solid line) and nonlinear absorption (for longer wavelengths, dashed line). From this change in the relative trend of the $J_{\rm SC}$ a significant contribution of TPA can be deduced.

4.4 Simulation of TPA Impact on $J_{\rm SC}$

4.4.1 Validation of Simulation Approach

The experimental results presented in the previous section demonstrated that nonlinear contributions in the measured J_{SC} are detectable. A comparison of the relative trend of the measured J_{SC} with linear and nonlinear absorption in Si pointed out that the detected nonlinear contributions can be attributed to TPA.

However, the presented experimental approach is rather limited regarding its temporal variations induced to the ultrashort pulse train (Fig. 4.6b). E.g. it does not allow for generation of continuous radiation and, thus, lacks comparability of pulsed versus continuous illumination, which is the most prominent comparison regarding applicability of ultrashort laser pulses in solar cell characterization. Therefore, a simulation approach based on the theoretical model derived in Section 4.2 will be presented in this section. Modeling the experimental conditions from Section 4.3 yields a validation of the simulation approach and justifies its application for a comparison of pulsed to continuous illumination. The experimental conditions in Section 4.3 allow for application of the mostly reduced differential equation given in Section 4.2. In Section 4.2.3 it was demonstrated that injection dependence of τ and saturation in carrier generation can be readily neglected for Si and the given spectral range (see Fig. 4.2b). Furthermore, the assumed illumination intensities allow for application of the QSS-approximation detailed in Section 4.2.4. Thus, Eq. (4.15) can be applied for a simulation of the experimental conditions.

Modeling the short circuit current requires incorporation of an extremely high front surface recombination velocity S_0 for simulating the carrier extraction (see Eq. (4.1)). As discussed e.g. by *Giesecke* in [20, pp. 244-245], this yields a crucial bottleneck in finite element approaches for solving Eq. (4.15), since a strongly confined spatial modification of carrier densities, by e.g. absorption or recombination, requires a fine spatial discretization Δz . In turn, such narrow spatial discretization involves a fine temporal discretization for accurate finite element modeling as well $(\Delta t \ll \Delta z^2/D_a)$, resulting in tremendous computational effort.

One way to circumvent this bottleneck is the integration of a Green's function that allows for an analytical description of the diffusion process. Taking into account the less explicit but powerful approximation, that $\alpha_{\text{tot}} \approx \alpha_{\text{bb}} + \sigma_{\text{FCA}}^n (\lambda/\mu m)^{x_n} N_D$ (which is only valid for low-injection conditions $\Delta n \ll N_D$),¹³ the excess carrier density does not affect any of the pulses incident onto the solar cell. Thus, the generation term is unaltered for all pulses and can be written as

$$G(z,t) = \frac{I(t) \alpha_{\text{tot}} \exp(-\alpha_{\text{tot}} z)}{h\nu \left(\alpha_{\text{tot}} + \beta I(t) \left(1 - \exp(-\alpha_{\text{tot}} z)\right)\right)} \cdot \left(\alpha_{\text{bb}} + \frac{\beta I(t) \alpha_{\text{tot}} \exp(-\alpha_{\text{tot}} z)}{2 \left(\alpha_{\text{tot}} + \beta I(t) \left(1 - \exp(-\alpha_{\text{tot}} z)\right)\right)}\right).$$
(4.16)

¹³This low-injection approximation is much weaker than the similar QSSapproximation (see Section 4.2.4) as not only the changes induced by a single pulse are considered, but the contribution of FCA in the steady-state condition of radiationsemiconductor interaction. Especially for wavelengths below 1150 nm the low-injection condition might be violated in Si.



Figure 4.9: Ratios of measured $J_{SC}s$ for aligned and misaligned ring cavities (black squares, filled and open symbols denote measurements at different days) and simulated ratios (red line) versus wavelengths (after [33]). The error bars indicate the standard deviations of the measurement. The reddish area surrounding the simulated values denotes the standard deviation of the simulation accounting for the limited precision of temporal ring cavity alignment.

With this approximation Eq. (4.15) can be written as

$$\frac{\partial \Delta n\left(z,t\right)}{\partial t} = D_{\rm a} \frac{\partial^2 \Delta n\left(z,t\right)}{\partial z^2} + G\left(z,t\right) - \frac{\Delta n\left(z,t\right)}{\tau} \tag{4.17}$$

and the Green's function approach from [20, pp. 56-60] can be applied.¹⁴

In Fig. 4.9 measured and simulated ratios of $J_{\rm SC}$ for aligned versus misaligned ring cavities are plotted over excitation wavelength. The measurements are given as black symbols (open and filled squares denote measurements on different days), including error bars as standard deviations. The simulation result is given as red line. The generation term G(z,t)(Eq. (4.16)) has been modeled from temporal pulse train shapes I(t)for the aligned and misaligned setup configuration (compare Fig. 4.6b) and material parameters from [55, 58, 61]. The remaining parameters in Eq. (4.17) are given as follows: the ambipolar diffusion coefficient has

 $^{^{14}}$ In Appendix D the approach is sketched along the lines of [20, pp. 56-60].

been set to $D_{\rm a} = 11.6 \,{\rm cm}^2 {\rm s}^{-1}$,¹⁵ the pulse duration to $\tau_{\rm FWHM} = 160$ fs (see Section 4.3.2.1) and the minority carrier lifetime to $\tau = 10 \,\mu {\rm s}$.¹⁶

The reddish area surrounding the simulation result denotes a standard deviation of the simulation. This results from the assumed normal distribution of translation stage alignment with 10 μ m standard deviation. It considers the limited accuracy of mechanical stage alignment and alignment control by the autocorrelation signal. The red line represents the mean value after 1000 randomized simulations.

Simulation and experiment demonstrate a very good agreement up to 1350 nm. Especially the rising edge at shorter wavelengths is imitated very well. For longer wavelengths the measurements suffer from noise due to comparably low signals. Although some experimental data points are not covered, the general tendency is reproduced by the simulation. In conclusion, the presented results demonstrate that the simulation approach is capable of a comparison of $J_{\rm SC}$ s under different temporal characteristics of the illuminating radiation.

4.4.2 Pulsed Versus Continuous Illumination

As the simulation results presented in Fig. 4.9 demonstrate a good agreement to the measurements, theoretical model and simulation approach appear to reflect the interaction of ultrashort laser pulses and short-circuited Si solar cells quite well. From this conclusion simulations covering the most prominent comparison of pulsed versus continuous illumination can be conducted.

For a comparison of $J_{\rm SC}$ s from pulsed and continuous illumination ultrashort laser pulses with $\tau_{\rm FWHM} = 100$ fs and $f_{\rm rep} = 80$ MHz are assumed. The average intensity $I_{\rm av}$ is varied in order to reflect various

 $[\]overline{{}^{15}D_{\rm a}} = \frac{D_{\rm n}D_{\rm p}(n+p)}{D_{\rm n}n+D_{\rm p}p} \approx D_{\rm p} \text{ with electron and hole diffusion coefficients } D_{\rm n,p} \text{ and}$ $\Delta n \ll N_{\rm D} [20, \text{ pp. 40-41}]. \text{ With hole mobility } b_{\rm p} \approx 450 \,\mathrm{cm}^2 \mathrm{V}^{-1} \mathrm{s}^{-1} [65] \text{ the hole diffusion coefficient is given by } D_{\rm p} = b_{\rm p} k_{\rm B} \mathcal{T}/q \approx 11.6 \,\mathrm{cm}^2 \mathrm{s}^{-1}.$

¹⁶The exact value of minority carrier lifetime is of minor importance as carrier extraction rates are much higher than recombination rates in that lifetime range.



Figure 4.10: Deviation of the $J_{\rm SC}$ for pulsed versus continuous illumination at various average illumination intensities for a Si solar cell (published in [33]). Ultrashort laser pulses with $\tau_{\rm FWHM} = 100$ fs and $f_{\rm rep} = 80$ MHz are assumed for the simulations.

illumination conditions (e.g. high average intensities for localized characterizations and concentrator applications or low intensities for large area differential current response measurements).

In Fig. 4.10 deviations of simulated $J_{\rm SC}$ s for pulsed versus continuous illumination are plotted over photon energy (normalized by band gap energy $E_{\rm g} = 1.12$ eV) and wavelength. The presented results emphasize the impact of TPA that is increasing with wavelengths and illumination intensities. It can be deduced that for standard measurement applications (monochromatic illumination intensity $I_{\rm av} < 0.1 \,{\rm Wcm^{-2}}$ and $\lambda < 1200 \,{\rm nm}$) the contribution of TPA is negligible. However, for highly concentrating measurement applications, as e.g. in local characterizations or for concentrator cell measurements, TPA might significantly affect the measurement outcome.

4.5 Conclusions

In this chapter a theoretical, experimental and computational analysis of the interaction of ultrashort laser pulses and solar cells was conducted. A special focus was placed on short circuit conditions, as this is most relevant for the calibration of solar cells.

Firstly, a theoretical model of the interaction of ultrashort pulses and solar cells was developed. In detailed discussions relevant approximations were introduced regarding carrier generation and minority carrier lifetime for a significant simplification of the model. Furthermore, a decoupling of the model's set of differential equations was achieved with a quasi-steadystate approximation. It was concluded from the theoretical discussions that TPA remains the only significant cause to variations in measured $J_{\rm SCS}$. This contribution of TPA has been experimentally demonstrated afterwards by $J_{\rm SC}$ measurements with varying pulse shapes. The pulse shape variations were accomplished by a double-ring-resonator setup that temporally redistributed the pulse energy in an ultrashort pulse train. As the degree of temporal variation of the pulse train was limited in this approach, a simulation was set up solving the previously derived theoretical model. The simulation was validated by the experimental outcomes. Subsequently, the computational approach was applied to conduct the most prominent, but experimentally hardly feasible, comparison of pulsed versus continuous illumination.

From these simulations very minor effects of nonlinearities induced by ultrashort pulses were found for standard applications. However, going beyond these applications in e.g. highly localized or concentrator measurements, significant contributions of TPA are present in the vicinity of the silicon solar cell's band gap. It was concluded, that care has to be taken regarding the temporal shape of the laser pulses when previously mentioned measurement configurations are applied.

Furthermore, the ongoing advances in solar cell technology that are e.g. trying to harvest a broader spectral range of the sun by upconversion [66] or Si-multi-junction devices [67] as well as the development of new solar cell materials with (partially) unknown optical properties, stress the importance of caution when applying ultrashort laser pulses for their electrical characterization. Current generated from TPA in the silicon part of an advanced cell concept might appear as an artifact and might be misinterpreted as an improved functionality of the device itself.

In conclusion, the results presented in this chapter demonstrate that ultrashort laser pulses might be readily applied for standard measurement applications (e.g. silicon solar cell, $\lambda \leq 1200$ nm, $I_{\rm av} \leq 0.1 \,{\rm W cm}^{-2}$). On the contrary, special measurements (highly localized, concentrator PV) or characterization of new materials or device concepts might be biased by TPA as nonlinear contribution to the measured current. Thus, a temporal shaping of the ultrashort laser pulses, yielding a significant reduction of the associated pulse peak powers, is highly recommended prior to their usage in highly accurate solar cell characterization or calibration facilities.

Chapter 5

Temporal Shaping of Ultrashort Laser Pulses

In this chapter temporal manipulation of ultrashort laser pulses will be discussed. Starting from a brief overview on several techniques for temporal shaping of ultrashort laser pulses, pulse stretching in multimode fibers will be identified as most promising for the intended application in this work. Therefore, the theory of impulse responses of step-index multimode fibers will be discussed. Afterwards a novel characterization method for multimode fiber properties will be presented that has been developed in the course of this work [68], followed by a discussion of how a step-index multimode fiber affects non-temporal radiation properties. Finally, an innovative monolithic fiber device for spatio-temporal shaping of ultrashort laser pulses will be presented, that has been developed for the special demands of the new measurement facility.

5.1 Strategies for Temporal Shaping

The results presented in Chapter 4: Interaction of Ultrashort Laser Pulses and Solar Cells demonstrated that the electrical characterization of solar cells using ultrashort laser pulses might suffer from nonlinearities biasing the measurement outcome. The chapter was concluded by recommending temporal shaping of the laser pulses in order to reduce or even eliminate this effect. In this chapter a convenient way for this shaping-task will be presented, starting from a short overview on pulse shaping techniques in this section.

5.1.1 A Brief Overview on Temporal Shaping Approaches

Several approaches for temporal shaping of laser pulses were developed in the past that might be classified in spectral and non-spectral methods. The first class, that takes advantage of the spectral composition of laser pulses, is best represented by Fourier synthesis pulse shaping of ultrashort pulses [69]. In these methods wavelength components of an ultrashort pulse are dispersed in space, individually delayed or filtered (by amplitude or phase masks) and recombined to a single beam. Major characteristics of this technique are the tight control of the actual temporal pulse shape as well as its reversibility (if no amplitude masks are used). Another approach exploiting the spectral composition of ultrashort pulses is material dispersion that introduces wavelength-dependent phase terms resulting in a temporal broadening of a previously transform-limited pulse [70, pp. 352-354].

The second class of pulse shaping techniques, the non-spectral class, rather generates various sub-pulses (independent of their spectral composition) and delays them with respect to each other prior to recombining them into a single beam (see e.g. [71, pp. 205-213] for a short summary of such methods). If no active components are applied, this process is irreversible and exhibits a much lower degree of controlling the output waveform as compared to the spectral methods. Thus, it is more often referred to as pulse *stretching* instead of pulse *shaping*.



Figure 5.1: Pulse duration over FWHM bandwidth for Gaussian shaped transform-limited ultrashort pulses with center wavelength λ_0 .

5.1.2 Temporal Shaping Strategy in This Work

In the present work, transform-limited ultrashort laser pulses of 100 to 200 fs pulse duration emitted at 80 MHz pulse repetition rate (thus, 12.5 ns pulse period) need to be converted into a continuous wave (cw) signal in order to definitely eliminate previously discussed nonlinearities (see Chapter 4). As this requires a temporal overlap of successive pulses, the extreme duty cycle of approximately 10^{-5} represents the main challenge of this task. In the following, prospects of some approaches for achieving pulse stretching from fs- to ns-scale will be briefly discussed.

Owing to the relation of pulse duration and spectral width of transformlimited pulses (compare Eq. (3.1)) a spectral reduction of the radiation represents one approach for pulse broadening. Moreover, this spectral reduction is, to some degree, anyway required for the development of the measurement facility introduced in Chapter 6. However, considering the desired standard full-width-at-half-maximum (FHWM) bandwidth of approximately 5 nm for the final setup, the achievable increase in pulse duration by this approach is rather limited (see Fig. 5.1). Apart from the fact that the FWHM bandwidths of 100 fs pulses below 580 nm center wavelength are readily narrower than 5 nm, the maximum achievable transform-limited pulse duration for 5 nm equals 1.76 ps (at $\lambda_0 =$ 2500 nm), thus, still four orders of magnitude lower than the pulse period. In addition to the spectral reduction, the ultrashort pulses could be dispersed by application of varying group velocities to the respective spectral components (see e.g. Eq. (3.21)). Two approaches might be distinguished in that: firstly, material dispersion can be exploited [70, pp. 352-354] or, secondly, Fourier synthesis methods might be applied [69]. Regarding Fourier synthesis methods there are no general restrictions in the amount of introduced pulse spreading given that an arbitrary large phase term can be experimentally added to certain frequency components. However, such setups can become quite challenging regarding alignment control and might be practically limited when scaling fs- to ns-pulses. Material dispersion, on the other hand, is more promising as it is experimentally quite conveniently implementable by coupling light into optical fibers. A major drawback of material dispersion is, however, that extensive material or fiber lengths of more than 1 km are required to convert a 100 fs transform-limited pulse at 870 nm center wavelengths into a ns-pulse.¹

Turning the discussion towards non-spectral pulse stretching, most approaches yield difficulties comparable to Fourier synthesis methods regarding alignment and complexity [71, pp. 205-213]. Especially those setups consisting of various mirror, beam splitter and prism combinations can result in tremendous alignment effort. A significantly more feasible approach is represented by multimode optical fibers. In addition to the previously discussed material dispersion, these fibers exhibit modal dispersion which enhances pulse stretching capabilities substantially [70, pp. 351-352]. For example, 100 m of a multimode fiber with numerical aperture NA = 0.22 is sufficient to achieve impulse responses in the ns-range. Moreover, physical delay lines can be realized when fibers of different lengths are combined, thereby enhancing the duty cycle of the ultrashort pulse train by several factors [72, 73]. Applying this prior to the actual pulse broadening by modal dispersion results in a significant reduction of the necessary fiber lengths and, hence, a significant reduction of overall material absorption. This argumentation exposes that multimode optical fibers are an excellent choice for a pulse-to-cw-conversion of the ultrashort pulse trains utilized in this work.

¹Assuming a silica-glass fiber with a group velocity dispersion parameter of $D_{\lambda} \approx -80 \text{ ps/km/nm}$ [70, p. 353].

Hereafter, the physical concept of pulse stretching in step-index² multimode optical fibers, a new method for characterizing their properties and further effects of the fibers on the radiation properties will be discussed. Finally, a monolithic fiber concept for pulse-to-cw-conversion will be presented that has been developed in the course of this work.

5.2 Impulse Response of Step-Index Multimode Optical Fibers

5.2.1 Introduction to Step-Index Fiber Optics

In this section a very brief introduction to step-index optical fibers will be given in order to introduce properties, parameters and concepts relevant for the subsequent sections of this chapter. A neat review of waveguide and fiber optics can be found in the textbook *Fundamentals of Photonics* by *Saleh* and *Teich* [70, pp. 289-364]. A more profound discussion of waveguide theory is given by *Snyder* and *Love* in the textbook *Optical Waveguide Theory* [74]. The following outline of optical fiber theory is mainly based on [70, pp. 289-364].

Generally spoken, step-index optical fibers are three-dimensional dielectric waveguides consisting of a low-loss core region with refractive index $n_{\rm co}$ and a cladding region with a smaller refractive index $n_{\rm cl}$. A ray hitting the core-cladding boundary is entirely reflected at the interface by means of total internal reflection (TIR) if its incidence angle exceeds the critical angle $\theta_{\rm c} = \arcsin(n_{\rm cl}/n_{\rm co})$.³ From this TIR condition the acceptance angle $\theta_{\rm a} = \arcsin\left(\sqrt{n_{\rm co}^2 - n_{\rm cl}^2}\right)$ with respect to the waveguide's axis can be determined. Rays incident within this acceptance angle onto a dielectric waveguide or an optical fiber will be guided by TIR. Another

 $^{^2 \}rm Step-index$ fibers exhibit much larger modal dispersion as graded-index fibers. The latter ones are therefore omitted in this chapter.

³Please note that the incidence angle is the complementary angle of the propagation angle θ as defined in Fig. 5.2.

term for the acceptance angle is the so-called numerical aperture

$$NA = \sqrt{n_{co}^2 - n_{cl}^2} \tag{5.1}$$

of the waveguide.

Although guidance of radiation can be explained in terms of geometrical optics, electromagnetic theory is necessary to describe the modal character of radiation propagation through waveguides, resulting in discrete properties of the radiation fields. This becomes clear from Fig. 5.2 that shows a most simple⁴ planar waveguide consisting of two ideal mirrors surrounding a medium of thickness 2a with refractive index n. A plane wave propagating in the waveguide is shown with its wavefronts indicated by the lines orthogonal to the ray's propagation direction. For being self-consistent the plane wave needs to reproduce itself after two reflections. It can be shown that this does only hold for distinct propagation angles [70, pp. 291-292]

$$\theta_m = \arcsin\left(\frac{m\lambda}{4an}\right) \tag{5.2}$$

being characteristic for the modes m = 0, 1, 2, ... of a planar waveguide. Naturally, these distinct propagation angles result in specific group velocities $v_m = c_0 \cos(\theta_m) / n$ of the different modes. A low order mode (small θ_m) passes a waveguide faster than a high order mode (high θ_m). This difference in propagation velocities results in temporal stretching and is called *modal dispersion*.

In the same manner as above discrete properties of modes propagating in cylindrical dielectric optical waveguides, namely fibers, can be derived.⁵ A significant simplification to the various possible modal solutions

⁴The equivalent situation in dielectric waveguides is slightly more complex as the phase term added to the propagating plane wave by each reflection is not always π but varies from 0 to π depending on the incidence angle [70, pp. 292].

⁵The modes either exhibit a purely transversal electric or magnetic field component (TE or TM modes) or they are of hybrid nature with neither field component being purely transversal (EH or HE modes) [70, pp. 331-348], [74, pp. 248-263]. The precise characteristics of the modes is irrelevant for the results in this section. Therefore, the TEM or LP simplification above is sufficient.



Figure 5.2: Schematic illustration of the formation of modes in a planar waveguide consisting of two ideal mirrors framing a material with refractive index n (adapted from [70, pp. 292]). An incident plane wave is only self-consistent for particular propagation angles θ_m .



Figure 5.3: Spatial intensity distributions and propagation angles with respect to the fiber axis for some LP_{lm} modes of a fused silica stepindex optical fiber with NA = 0.22 and $a = 50 \ \mu m$ at 632 nm radiation wavelength.

in fibers is obtained if weak guiding is assumed $(n_{\rm co} \approx n_{\rm cl})$. In that case both, the electric and magnetic field component are of purely transversal nature and the modes are called *transversal electro-magnetic* (TEM) modes, also referred to as *linearly polarized* (LP) modes. In contrast to the two-dimensional waveguide above, the LP modes in cylindrical dielectric waveguides are classified by two order numbers as several modal solutions m = 0, 1, 2, ... exist for a single azimuthal index l. In Fig. 5.3 spatial intensity distributions and propagation angles with respect to the fiber axis for some LP_{lm} modes of a step-index optical fiber are shown. Equivalent to the waveguide example above, the increase in propagation angles with mode number lm causes an increase in group velocity of that mode, resulting in temporal stretching of an input pulse when launched at different fiber modes.

5.2.2 Mode Coupling in Optical Fibers

In an ideal fiber, the modes, as e.g. shown in Fig. 5.3, will propagate unaltered through the waveguide. In contrast, fiber bending, imperfections and irregularities yield coupling of the individual modes in real fibers. In a ray optical picture this can be understood as an abrupt change of the rays' propagation angle resulting in a change of the fiber mode. Due to modal dispersion (compare Eq. (5.2)) mode coupling becomes a significant contribution to the overall pulse stretching in multimode optical fibers and needs to be considered when computing fiber impulse responses.⁶

Power coupling of modes can be described by [78]

$$\frac{\mathrm{d}P_{l'm'}}{\mathrm{d}z} = \sum_{l} \sum_{m} \kappa_{l'm',lm} \left(P_{lm} - P_{l'm'} \right) \exp\left(-\frac{D_{\mathrm{cl}}}{2} \left(\beta_{l'm'} - \beta_{lm} \right) \right)^2,$$
(5.3)

with coupling coefficients $\kappa_{l'm',lm}$ and $(P_{lm} - P_{l'm'})$ as difference of modal powers. Further, the degree of power coupling depends on a constant D_{cl} , that describes the correlation length of the waveguide disturbances inducing the coupling, and on the difference of modal propagation constants with $\beta_{lm} = n_{co} 2\pi \cos(\theta_{lm}) / \lambda$. This accounts for an increased coupling of neighboring modes due to their similar propagation constants. For a rigorous discussion of mode coupling along propagation direction z, Eq. (5.3) needs to be solved for all $LP_{l'm'}$ modes. Even those modes that are initially not excited potentially carry power after a certain distance z as

⁶This short introduction to mode coupling is restricted to *power* coupling between modes. It has to be mentioned that more recent work on exploiting the nature of multimode fibers by means of mode-division multiplexing (for e.g. enhanced data transmission rates in optical networks) gave rise to advanced coupling models [75], termed *field coupling models*, including e.g. polarization effects [76] and so-called *principal modes* [77].

they are excited by mode coupling processes. Furthermore, it is noteworthy that mode coupling can also induce additional losses by coupling of LP modes to e.g. radiation modes leaving the fiber (not incorporated in Eq. (5.3)).

5.2.3 Gloge's Time-Dependent Power Flow Equation

As detailed above, mode coupling contributes to the temporal properties of a multimode fiber, hence, each mode needs to be considered individually in a rigorous impulse response computation, even if not all modes are initially excited. As in case of highly multimode fibers the number of modes can be in the range of millions,⁷ such rigorous computation involves tremendous computational effort. A significant reduction of computational effort can be achieved if the distinct modes and their associated amplitudes and properties are expressed as continuous functions over propagation angle θ . A suchlike approach was presented by *Gloge* in 1972 [79]. As this approach is essential for the novel multimode fiber characterization method that has been developed in the course of this work (see Section 5.3), it is briefly outlined subsequently.

The first fundamental assumption underlying *Gloge*'s approach is, that the LP_{lm} modes of an optical fiber can be grouped by a single mode number q according to $q = 2m + l.^8$ With this approximation, identical propagation angles for all LP_{lm} modes grouped into mode number qare obtained and, in analogy to a two-dimensional waveguide (compare Eq. (5.2)), are given by

$$\theta_q = \frac{q\lambda}{4an}.\tag{5.4}$$

Consequently, the difference in propagation angles of two neighboring modal groups q and q + 1 is

$$\Delta \theta = \frac{\lambda}{4an}.\tag{5.5}$$

⁷For example: 1.08 million modes are supported by a multimode fiber with NA = 0.39 and core radius $a = 400 \ \mu \text{m}$ at 600 nm wavelength (with number of modes $M = 4/\pi^2 (2\pi a \text{NA}/\lambda)^2$ [70, pp. 334, 338]).

⁸Please note that a different notation for the modes is used than in [79].

Also, *Gloge* takes into account that mode coupling is predominantly present between neighboring modes in multimode fibers $(\beta_{l'm'} - \beta_{lm} \approx 0)$, hence, being equivalent to a diffusion process in which neighboring modal sections exchange optical power. From Eq. (5.3) follows with these assumptions, that the incremental power change of the q-th mode dP_q within an incremental distance dz can written as [79]

$$q\frac{\mathrm{d}P_q}{\mathrm{d}z} = -q\alpha_q P_q + qd_q \left(P_{q+1} - P_q\right) + (q-1)d_{q-1} \left(P_{q-1} - P_q\right). \quad (5.6)$$

The first term denotes power losses due to attenuation and incorporates both coupling to radiation modes and material absorption. The coupling coefficients or rates are given as so-called diffusion coefficients d_q and d_{q-1} . The factors q and (q-1) take into account that the total number of modes within the modal group q and q-1 actually equals q and q-1, respectively. In [80] *Gloge* extends his work and includes the temporal domain in dP according to

$$\mathrm{d}P = \frac{\partial P}{\partial z}\mathrm{d}z + \frac{\partial P}{\partial t}\mathrm{d}t.$$
 (5.7)

With Eq. (5.7) and $dt/dz = n/(c_0 \cos(\theta))$, Eq. (5.6) is modified to

$$\frac{\partial P_q}{\partial z} = -\alpha_q P_q - \frac{n}{c_0 \cos(\theta)} \frac{\partial P}{\partial t} + d_q \left(P_{q+1} - P_q \right)
+ (q-1) \frac{1}{q} d_{q-1} \left(P_{q-1} - P_q \right).$$
(5.8)

Finally, Eq. (5.8) can be transformed into a continuum by introducing

$$\frac{P_{q+1} - P_q}{\theta_{q+1} - \theta_q} = \frac{\mathrm{d}P_q}{\mathrm{d}\theta} \tag{5.9}$$

and a functional dependency of α and d on θ . This yields the timedependent power flow equation [80]

$$\frac{\partial P(\theta, z, t)}{\partial z} = -\alpha(\theta) P(\theta, z, t) - \frac{n}{c_0 \cos(\theta)} \frac{\partial P(\theta, z, t)}{\partial t} + \frac{1}{\theta} \frac{\partial}{\partial \theta} \left(\theta d(\theta) \frac{\partial P(\theta, z, t)}{\partial \theta}\right).$$
(5.10)

Given that attenuation $\alpha(\theta)$, mode coupling $d(\theta)$ and power distribution $P(\theta, z_0, t)$ at $z = z_0$ are known, the power distribution at $z_1 = z_0 + \Delta z$, $P(\theta, z_1, t)$, can be computed from Eq. (5.10).

5.2.4 Numerical Solution to the Power Flow Equation

Incorporating modal diffusion and attenuation in a most general approach by taking into account the modal dependency of both, no analytical solutions to Eq. (5.10) are available.⁹ Thus, a finite element scheme is used to solve the time-dependent power flow equation and retrieve impulse responses of step-index multimode fibers. For the fiber characterization method detailed below (see Section 5.3) a method published by *Mateo* et al. has been adapted that expresses Eq. (5.10) in a matrix form, thereby boosting computation speed [83]. Subsequently, the approach will be sketched along the lines of [68, 83].

In order to eliminate the temporal partial derivative in Eq. (5.10) a Fourier transform is applied, yielding

$$\frac{\partial p(\theta, z, \omega)}{\partial z} = -\left(\alpha(\theta) + \frac{n}{c_0 \cos(\theta)}i\omega\right)p(\theta, z, \omega) + \frac{1}{\theta}\frac{\partial}{\partial\theta}\left(\theta d(\theta)\frac{\partial p(\theta, z, \omega)}{\partial\theta}\right).$$
(5.11)

Eq. (5.11) is then transformed into finite differences by a first-order

⁹For analytical estimates of Eq. (5.10) several constraints were implemented in the past including constant mode coupling and Gaussian input power distribution [80–82].

forward difference for the z-derivative

$$\frac{\partial p\left(\theta, z, \omega\right)}{\partial z} \approx \frac{p\left(\theta, z + \Delta z, \omega\right) - p\left(\theta, z, \omega\right)}{\Delta z} \tag{5.12}$$

and first- and second-order central differences in θ

$$\frac{\frac{\partial p\left(\theta, z, \omega\right)}{\partial \theta}}{\frac{\partial^2 p\left(\theta, z, \omega\right)}{\partial \theta^2}} \approx \frac{p\left(\theta + \Delta \theta, z, \omega\right) - p\left(\theta - \Delta \theta, z, \omega\right)}{2\Delta \theta} \\ \frac{\frac{\partial^2 p\left(\theta, z, \omega\right)}{\partial \theta^2}}{\frac{\partial \theta^2}{\partial \theta^2}} \approx \frac{p\left(\theta + \Delta \theta, z, \omega\right) - 2p\left(\theta, z, \omega\right) + p\left(\theta - \Delta \theta, z, \omega\right)}{\Delta \theta^2}.$$
(5.13)

With Eqs. (5.11) to (5.13) $p(\theta, z + \Delta z, \omega)$ is given by

$$p(\theta, z + \Delta z, \omega) = \left(1 - \left(\alpha(\theta) + \frac{n}{c_0 \cos(\theta)}i\omega\right)\Delta z\right)p(\theta, z, \omega) + \frac{\Delta z}{2\Delta\theta}\left(\frac{d(\theta)}{\theta} + d'(\theta)\right)\left(p(\theta + \Delta\theta, z, \omega)\right) - p(\theta - \Delta\theta, z, \omega) - \frac{2d(\theta)\Delta z}{\Delta\theta^2}p(\theta, z, \omega) + \frac{d(\theta)\Delta z}{\Delta\theta^2}\left(p(\theta + \Delta\theta, z, \omega)\right) + p(\theta - \Delta\theta, z, \omega),$$
(5.14)

with first derivative of $d(\theta)$ after θ denoted as $d'(\theta)$. Rewriting p as a vector with (N'+1) elements representing discretized angles $\theta = k\Delta\theta$, with k = 0, 1, 2, ..., N', Eq. (5.14) can also be expressed in matrix form as

$$\mathbf{p}(z_2,\omega) = \left(\tilde{\mathbf{A}}(\omega) + \tilde{\mathbf{D}}\right)^w \mathbf{p}(z_1,\omega).$$
 (5.15)

In this representation, $w = (z_2 - z_1) / \Delta z$ denotes the number of steps performed in z-direction while solving the power flow equation from z_1 to z_2 . The diagonal matrix

$$\tilde{A}_{k,k}(\omega) = \exp\left(-\alpha \left(k\Delta\theta\right)\Delta z - \frac{\Delta zn}{c_0}i\omega \left(\frac{1}{\cos\left(k\Delta\theta\right)} - 1\right)\right)$$
(5.16)

represents both attenuation (first part; modulating the amplitude) and

5.2 Impulse Response of Step-Index Multimode Optical Fibers

propagation (second part; modulating the phase).¹⁰ It is noteworthy that the phase term is dependent on the angle as this represents the modal dispersion causing pulse stretching in multimode fibers (compare Eq. (5.2)). The mode coupling, or diffusion process, respectively, is implemented by the tri-diagonal matrix

$$\tilde{D}_{k,k-1} = \left(d\left(k\Delta\theta\right) - \frac{d\left(k\Delta\theta\right)}{2k} - \frac{d'\left(k\Delta\theta\right)\Delta\theta}{2} \right) \frac{\Delta z}{\Delta\theta^2} \\ \tilde{D}_{k,k} = -2d\left(k\Delta\theta\right) \frac{\Delta z}{\Delta\theta^2}$$

$$\tilde{D}_{k,k+1} = \left(d\left(k\Delta\theta\right) + \frac{d\left(k\Delta\theta\right)}{2k} + \frac{d'\left(k\Delta\theta\right)\Delta\theta}{2} \right) \frac{\Delta z}{\Delta\theta^2}.$$
(5.17)

In order to avoid power loss due to diffusion the boundary conditions

$$d(\theta) \left. \frac{\partial p}{\partial \theta} \right|_{\theta=0} = 0$$

$$d(\theta) \left. \frac{\partial p}{\partial \theta} \right|_{\theta=N'\Delta\theta} = 0$$

$$(5.18)$$

are formulated as

$$\tilde{D}_{0,0} = -4d(0)\frac{\Delta z}{\Delta\theta^2} \qquad \tilde{D}_{0,1} = 4d(0)\frac{\Delta z}{\Delta\theta^2}
\tilde{D}_{N',N'-1} = 2d(N')\frac{\Delta z}{\Delta\theta^2} \qquad \tilde{D}_{N',N'} = -2d(N')\frac{\Delta z}{\Delta\theta^2}.$$
(5.19)

As $\tilde{\mathbf{A}}(\omega)$ and $\tilde{\mathbf{D}}$ contain all information on the fiber properties once the matrix $\mathbf{W}(\omega) = \left(\tilde{\mathbf{A}}(\omega) + \tilde{\mathbf{D}}\right)^w$ is determined any output power distribution $\mathbf{p}(z_2, \omega)$ can be computed in a very fast manner. In Fig. 5.4 (a)-(c) angular-temporal power distributions for a typical step-index plastic optical fiber of different lengths L are shown. The two-dimensional representations are obtained from inverse Fourier transforms of $\mathbf{p}(z = L, \omega) \equiv$

 $^{^{10}}$ In contrast to [83], a constant phase term has been added to Eq. (5.16) that takes into account phase changes, or temporal delays, respectively, common to all propagating modes.



Figure 5.4: Angular-temporal power distributions ((a) to (c)) and resulting pulse shapes ((d) to (f)) for a highly multimode plastic optical fiber with NA = 0.39 after 50 m ((a), (d)), 75 m ((b), (e)) and 100 m ((c), (f)) fiber length computed with Eq. (5.15), Eq. (5.20) and Eq. (5.21). As the input pulse has been a Dirac delta pulse, the resulting pulse shapes can be referred to as impulse responses.

 $p\left(\boldsymbol{\theta}, \boldsymbol{z} = \boldsymbol{L}, \boldsymbol{\omega}\right)$ according to

$$P(\theta, z = L, t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} p(\theta, z = L, \omega) \exp(i\omega t) d\omega.$$
 (5.20)

The resulting pulse shapes H(L,t) shown in Fig. 5.4 (a)-(c) are computed from an integration over angle θ

$$H(L,t) = \int_{0}^{\pi/2} \sin(\theta) P(\theta, z = L, t) d\theta.$$
 (5.21)

5.3 Characterization of Multimode Fibers

As demonstrated in the previous section, impulse responses of step-index multimode fibers can be computed from *Gloge*'s time-dependent power flow equation given that input modal power distribution and fiber properties are known. Therefore, knowledge of fiber attenuation $\alpha(\theta)$ and mode coupling (or diffusion) $d(\theta)$ is important for predicting temporal performances of multimode optical fibers.

Several approaches for a determination of these fiber properties were presented within the last decades. Whereas modal attenuation $\alpha(\theta)$ has been measured directly [84, 85], mode coupling has been determined from farfield fiber outputs, thus, from the angular characteristics involved in Eq. (5.10) [86–89]. In the course of this work a novel characterization approach has been developed, that, in contrast to previous methods, estimates mode-dependent fiber attenuation and mode coupling based on impulse responses of a step-index multimode fiber (Estimation of Fiber Properties from Impulse Responses, EFPIR) [68].

Subsequently, the EFPIR-method will be outlined along the lines of the author's publication [68]. Utilizing the numerical approach presented in Section 5.2.4 to solve the time-dependent power flow equation (Eq. (5.10)), an optimization algorithm will be presented that retrieves the fiber properties by applying an intuitive downhill simplex scheme [90]. Finally, the computational approach will be validated against simulated impulse responses and measured bandwidths from different step-index plastic optical fibers.

5.3.1 EFPIR-Method

5.3.1.1 General Program Flow

With Eq. (5.21) (by taking into account Eqs. (5.15) and (5.20)) the temporal response of a step-index multimode fiber to any input angular-temporal power distribution $P(\theta, z = 0, t)$ can be calculated given that fiber length as well as attenuation and diffusion properties are known. In turn, knowing the impulse response of a multimode fiber, its length and the input power distribution $P(\theta, z = 0, t)$, a determination of the fiber's attenuation and diffusion characteristics should be possible.

However, as attenuation and diffusion have a quite similar impact on the impulse response of a step-index multimode fiber¹¹ the simultaneous optimization of both parameters, seeming advantageous at first sight, might be yielding erroneous results. Therefore, the determination of attenuation and diffusion is decoupled within the EFPIR-method by using two reference impulse responses at different fiber lengths but identical launching conditions.¹²

The general program flow of the EFPIR-method is schematically shown in Fig. 5.5. From a constant attenuation γ and the initial power distribution $P(\theta, z = 0, t)$ that is coupled into the fiber under test, the computed impulse response (shown as black dashed line) is adapted to a reference impulse response (green solid line; known e.g. from impulse response measurements) by changing the diffusion or mode coupling term $d(\theta)$ (top box) in Fig. 5.5). As a measure for the similarity of the impulse responses their root-mean-square deviation (RMSD) is computed. When the RMSD converges to a minimum, it is assumed that a best diffusion term $d(\theta)$ with respect to the current attenuation properties is found. This diffusion term is then passed on to the next algorithm step that optimizes the attenuation properties $\alpha(\theta)$ (bottom box) likewise and, in turn, passes on its best attenuation term to a next diffusion term optimization (again top box, with $\alpha(\theta)$ taken from the latest $\alpha(\theta)$ -optimization). This procedure is repeated until the RMSD for both, attenuation and diffusion optimization, stays unaltered within an individual roundtrip.

5.3.1.2 Downhill Simplex Method

For a successful application of the EFPIR-method illustrated in Fig. 5.5, an accurate optimization of attenuation $\alpha(\theta)$ and diffusion properties $d(\theta)$

¹¹For example, a low amplitude in the trailing edge of a pulse can be accomplished by both, a high absorption for large propagation angles or a strong diffusion directed towards lower angle modes.

¹²Please note that other solutions to the problem of wrong convergence in diffusion and attenuation, while simultaneously optimizing both, might be successfully applied as well.



Figure 5.5: Schematic illustration of program flow used in the EFPIRapproach. Attenuation $\alpha(\theta)$ and diffusion properties $d(\theta)$ are deduced from alternating optimization of simulated pulse shapes with respect to reference pulse shapes (measured or computed ones). For computation of the simulated pulse shapes the input power distribution $P(\theta, z = 0, t)$ is required.

is vital. However, as scanning the entire parameter range that is attributed to the respective functions $\alpha(\theta)$ and $d(\theta)$ is too time-consuming, the downhill simplex method proposed by *Nelder* and *Mead* is implemented [90].¹³

The basic purpose of the downhill simplex approach is a minimization of any given objective function in any given N-dimensional parameter space. Instead of scanning the entire parameter space with an Ndimensional grid, the downhill simplex approach compares the objective function output at (N + 1) different parameter sets. Following distinct

¹³It has to be mentioned, that faster, more elegant and potentially more suitable optimization routines might exist, that e.g. incorporate derivatives of the objective function, thereby gaining more information on its evolution in the parameter space at shorter time scales. However, the downhill simplex scheme, as an intuitive and conveniently implementable approach, has been working excellently in the course of this work. Still, the EFPIR-method might be speed up significantly by the implementation of an optimized function minimization approach.


Figure 5.6: Illustration of function minimization using the downhill simplex method [90] for an arbitrary objective function in a twodimensional parameter space. By comparing the simplex vertices and consecutively substituting the worst parameter set, the simplex moves towards the function minimum, where it finally contracts to find the (potentially) best parameter set.

rules, the parameter set providing the worst value of the objective function is consecutively substituted by another parameter set, thereby yielding a steady improvement of the (N+1) objective function outputs. In Fig. 5.6 a simple example for an arbitrary objective function in a two-dimensional parameter space (N = 2) is shown. The N+1 = 3 initial data points being compared to each other are forming a simplex (which is a triangle in the two-dimensional case) displayed by the solid lines. As, in this example, the aim is to find the minimum of the objective function, the worst parameter set (most red one) is substituted by a reflection of its parameter set at the centroid of the remaining data points. With this new parameter set an updated simplex is formed, illustrated by the dashed lines, and being treated equivalently to the previous one. Besides the reflection operation, the simplex can be expanded, contracted or reduced (for further details it is referred to [90]). With these operations, the simplex *moves* within a given parameter space towards the minimum of the objective function yielding the (potentially) best parameter set.

5.3.1.3 Parametrization of Attenuation and Diffusion

The objective function of the downhill simplex algorithm in the EFPIRmethod is the RMSD of reference and computed pulse shapes (compare Fig. 5.5). The parameter space is given by the parameters of the attenuation $\alpha(\theta)$ or diffusion function $d(\theta)$, respectively. For the validation of the EFPIR-method, presented in [68] and outlined subsequently in Section 5.3.2, the functions

$$d(\theta) = \begin{cases} d_0 \cdot \left(\frac{\theta_c}{|\theta|}\right)^{2 \cdot d_q}, & \text{if } \theta \neq 0\\ d(\Delta \theta) - \Delta \theta \cdot \left|\frac{\partial d(\theta)}{\partial \theta}\right|_{\theta = \Delta \theta}, & \text{if } \theta = 0 \end{cases}$$
(5.22a)

$$\alpha\left(\theta\right) = \alpha_0 + \alpha_1 \theta^2 + \alpha_2 \theta^4, \qquad (5.22b)$$

have been assumed with free parameters d_0 , d_q , α_1 and α_2 . The remaining parameter α_0 is given according to

$$\alpha_0 = \gamma - \frac{\sum_{k=0}^{k_c} \left(\alpha_1 \cdot (k \cdot \Delta \theta)^2 + \alpha_2 \cdot (k \cdot \Delta \theta)^4 \right)}{(k_c + 1)}, \qquad (5.23)$$

with $k_{\rm c} = \theta_{\rm c} \Delta \theta$ and the constant attenuation coefficient γ , known from literature or measured independently.¹⁴ Eq. (5.23) ensures that the overall attenuation of the fiber equals γ . Additionally, Eq. (5.22) is constrained by the physical restriction that $\alpha(\theta) \ge 0$ as well as $d(\theta) > 0$ as well as $d_{\rm q} > 0$ to ensure positive diffusion that decreases with propagation angle θ [91].

5.3.2 Validation of EFPIR-Method

The previously presented EFPIR-method (see Section 5.3.1 and [68]) enables an estimation of modal diffusion and attenuation in step-index multimode fibers from impulse responses. In this section, the validation of the EFPIR-method will be briefly recapitulated [68].

 $^{^{14}}$ The models for fiber attenuation and diffusion given in Eq. (5.22) were used in previous publications as well [79, 82, 91].



Figure 5.7: Evolution of impulse responses (a) and fiber properties ((b), (c)) over iterations in the EFPIR-method for 100 m PGU fiber. Blue lines indicate the reference data with properties from [89], red lines indicate the data obtained from the EFPIR-method. Published in [68].

For validation of the EFPIR-method typical step-index plastic optical fibers have been used, namely, ESKA-PREMIER GH4001 from Mitsubishi (denoted as GH subsequently) and PGU-FB1000 (PGU) from Toray. From previous publications by other authors [83, 89] the modal attenuation and diffusion characteristics of these fibers are well-known. With the fiber properties given by *Mateo* et al. in [89] and an initial angular-temporal power distribution

$$P(\theta, z = 0, t) = \delta(t) \exp\left(-4\ln(2)\left(\frac{\theta}{\theta_0}\right)^2\right), \qquad (5.24)$$

with $\theta_0 = 7.3^{\circ}$, temporal responses of the fiber types at 75 m and 150 m have been computed and have been taken as reference pulse shapes $H_{\rm ref} (z = 75 \,\mathrm{m}, t)$ and $H_{\rm ref} (z = 150 \,\mathrm{m}, t)$. As Eq. (5.24) is a Dirac delta pulse $(\delta(t))$, the temporal responses $H_{\rm ref} (z = 75 \,\mathrm{m}, t)$ and $H_{\rm ref} (z = 150 \,\mathrm{m}, t)$ are denoted as impulse responses hereafter.

In Fig. 5.7 the evolution of impulse responses (a) and fiber properties

((b), (c)) with iteration steps of the EFPIR-method are shown for 100 m PGU fiber (blue lines denote the reference shapes and properties, red lines the results of the EFPIR-algorithm).¹⁵ Although not being used as reference pulse shape, the shape obtained after several iterations of the EFPIR-method shows an excellent agreement to the impulse response at 100 m fiber length. This is caused by the very good conformity of modal attenuation $\alpha(\theta)$ and diffusion $d(\theta)$ (see Fig. 5.7b and Fig. 5.7c).

Naturally, the fiber properties deduced from the EFPIR-method cannot imitate the reference properties perfectly, as different functional dependencies are assumed.¹⁶ This becomes apparent from Fig. 5.8 that displays $\alpha(\theta)$ and $d(\theta)$ for the GH (top row) and PGU fiber (bottom row). Within the constraints given by the deviations of the assumed modal functions the EFPIR-method reproduces the reference properties of the GH and PGU fiber quite well. This is as well highlighted by the dashed lines that represent least squares fits to the reference properties for the modal dependencies assumed in Eq. (5.22). From this consideration, remaining deviations of the EFPIR results to the reference properties can be mainly attributed to the intrinsic differences in the assumed modal dependencies.

However, the retrieved fiber properties using the EFPIR-method are still very well suitable for an estimation of temporal fiber characteristics, as it is demonstrated in Fig. 5.9 and Fig. 5.10. In Fig. 5.9 impulse (a) and frequency responses (b) of the PGU fiber at different lengths are shown for both the reference fiber properties (blue lines) and the properties determined by the EFPIR-method (red lines). The close match over all considered fiber lengths demonstrates the applicability of the EFPIR-method for a determination of fiber properties.

A different respresentation of the results shown in Fig. 5.9 is given in Fig. 5.10b showing the FWHM pulse durations and bandwidths of the

¹⁵It is noteworthy, that a proper simplex initialization prior to each roundtrip of the EFPIR-algorithm is important for avoiding a convergence to local instead of global minima in the downhill simplex optimization of attenuation and diffusion. For a detailed discussion of simplex initialization the interested reader is referred to [68].

¹⁶In the reference properties much more free parameters are included. This would yield a tremendous increase of computational time in the EFPIR-method and might also induce stability problems as a lower number of free parameters yields a less complex parameter space for the downhill simplex optimization.



Figure 5.8: Comparison of reference fiber properties from [89] (blue line) to the properties retrieved from the EFPIR-method (red solid line) for the GH (top row) and PGU fiber (bottom row). The dashed red lines indicate least squares fits to the reference data with the modal functions for attenuation and diffusion assumed in Eq. (5.22). Published in [68].

PGU fiber over fiber length. Again, red lines show the result from the EFPIR-method, blue symbols denote computed values based on the reference fiber properties. The black symbols are measurements of the fiber from [83]. The close match of computed values, based on both sets of fiber properties (the reference properties and the once obtained with EFPIR), and measured values emphasize once more that the EFPIR-method is a suitable approach for an accurate determination of fiber properties. In Fig. 5.10a the same is shown for the GH fiber yielding the same interpretations as discussed above.



Figure 5.9: Impulse responses (a) and normalized frequency responses (b) of the PGU fiber over fiber lengths. The responses based on the reference fiber properties from [89] are shown in blue, the ones based on the fiber properties retrieved by the EFPIR-method in red. Published in [68].



Figure 5.10: Comparison of pulse durations and bandwidths of the GH (a) and PGU fiber (b). Red lines indicate those values obtained from the EFPIR-method, blue symbols those from the reference fiber properties [89]. The black symbols are experimental values from [83]. Published in [68].

In this section a novel characterization method for attenuation and mode coupling properties of step-index multimode fibers was presented and validated, that has been developed in the course of this work [68]. The EFPIR-method allows for an accurate determination of the mentioned fiber properties form impulse responses. It represents, to the best of the author's knowledge, the first such approach that is purely based on impulse responses and does not require any measurements of spatial fiber output patterns.

5.4 Further Effects on Laser Pulses by Multimode Optical Fibers

5.4.1 Effects on Spatial Intensity Distribution

Apart from the temporal stretching of pulses in multimode optical fibers (discussed in Sections 5.2 and 5.3), the redistribution of energy within the fiber modes also yields a modification of the spatial intensity distribution that is given by the weighted sum over all excited fiber modes (see Section 5.2.1). In this section, this effect of multimode fibers will be briefly discussed.

Given that not all modes are initially launched the degree of modification in the spatial intensity distribution is intrinsically tied to the fiber length, as the modal distribution evolves with elongation of the fiber (see also Fig. 5.4). However, after a certain fiber length, that depends on the specific fiber properties, mode coupling and modal attenuation balance and an equilibrium mode distribution is achieved resulting in a stationary intensity distribution [79]. In step-index multimode fibers, this stationary intensity distribution typically resembles the shape of a top-hat profile, hence, exhibiting a rather flat intensity distribution. This modification of the spatial intensity distribution is exploited in various applications, especially in laser materials processing [92, 93]. In the context of optoelectronic device characterization it might be advantageous as well, as



Figure 5.11: Measured near-field intensity distributions at a multimode fiber end facet for various fiber lengths. The graph displays radially integrated amplitudes of the intensity distributions shown above the plot. For this experimental demonstration laser radiation at 800 nm has been coupled into a FT200EMT/Thorlabs multimode fiber (100 μ m core radius, NA = 0.39) of various lengths.

any input intensity distribution might be transformed to a rather uniform intensity distribution at low optical losses.¹⁷

In Fig. 5.11 this transformation is exemplified for a step-index multimode fiber with 200 μ m core diameter and NA = 0.39. For these experiments an ultrashort laser pulse at 800 nm center wavelength has been centrally launched into the fiber at an input numerical aperture of NA_{input} ≈ 0.117 . While keeping the coupling conditions unaltered, the

¹⁷Please note that, for this being a useful feature, measurements in the near-field of the fiber end facet or an imaging of this near-field in the measurement plane is required.

fiber has been shortened continuously. The images on top of the graph show near-field intensity distributions at the fiber end facet at the given fiber lengths (measured with a 4f imaging configuration). The graph below displays the radially integrated cross sections of above intensity distributions. The results clearly demonstrate the effect of mode coupling with increasing fiber length. While the central launch conditions at comparably narrow propagation and, thus, modal angles are apparent for short fiber lengths, an elongation of the fiber continuously increases uniformity of the near-field fiber output caused by enhanced contribution of all fiber modes. Further, after a specific length (in this case at about 25 m) no significant variations in the spatial intensity distributions are observed as the fiber is elongated further.¹⁸ It has to be noted that the intensity (apparently) exceeding the fiber core radius in the given graph is partially caused by the limited imaging capabilities of the optical setup (e.g. limited sharpness, back-reflections etc.) and partially caused by cladding and radiation modes that are not addressed in this chapter.

5.4.2 Effects on Polarization and Coherence

Apart from the spatial homogenization of radiation in a multimode optical fiber, the statistical mode coupling processes also yield a reduction of the degree of polarization [94] and a reduction of the radiation's coherence [95]. Although a detailed discussion of these effects is beyond the scope of this work, the appearance of these effects will be briefly exemplified by measurements that have been conducted within this work.

A convenient description for the degree of polarization of an electromagnetic wave is given by the Stokes parameters fragmenting the polarization into its linear and circular polarization contributions [96, pp. 373-379]. In Fig. 5.12 the Stokes parameters for radiation emitted by 2 m multimode optical fiber with NA = 0.22 (M25L02/Thorlabs) are given in a spatially resolved manner. The measurements have been conducted

¹⁸Please note that this fiber length, marking invariance of spatial intensity distribution for further elongation of the fiber, is not meant to be generalized; instead, it only holds for the specific conditions applied in this illustrative example. Any other launch conditions might yield a significantly different evolution of spatial intensity distribution over fiber lengths.

5.4 Further Effects on Laser Pulses by Multimode Optical Fibers



Figure 5.12: Spatially resolved Stokes parameters of radiation emitted from a 2 m long multimode fiber with NA = 0.22 (M25L02/Thorlabs). (a) denotes the degree of linear polarization at 0° (S_1), (b) the degree of linear polarization at 45° (S_2), (c) the degree of circular polarization (S_3) and (d) the overall degree of polarization (V_{Stokes}). The color coding denotes the amplitudes and orientations (-1 and 1, respectively) of the measured polarization contributions.

by coupling linearly polarized ultrashort laser pulses into the fiber and analyzing the emitted radiation field by a 4f imaging setup, a quarterwaveplate and a linear polarizer.¹⁹ The spatial information has been obtained by imaging the radiation field transmitted by the described optical setup onto a CMOS-camera, that has been calibrated pixel-wise regarding its polarization dependent current response prior to the measurements. In Fig. 5.12a the amount of linear polarization oriented at 0° or 90° is shown (S_1) , in (b) the amount of linear polarization oriented at 45° or 135° (S_2) and in (c) the amount of circular polarization (S_3) . The color-coding ranging from -1 (blue) to 1 (red) denotes the amplitude and orientation of the particular polarization contributions (e.g. blue indicates 90° polarization and red 0° polarization in Fig. 5.12a or clockwise (blue) and counterclockwise (red) circular polarization in (c)). Fig. 5.12d gives the

¹⁹Details of the measurement procedure are e.g. given in [96, p. 374].

degree of polarization computed from

$$V_{\rm Stokes} = \frac{\sqrt{S_1^2 + S_2^2 + S_3^2}}{I} \tag{5.25}$$

with overall intensity I (measured without polarizer or waveplate). $V_{\text{Stokes}} = 0$ denotes unpolarized and $V_{\text{Stokes}} = 1$ polarized radiation.

Taking into account that the degree of polarization without multimode fiber (pure laser pulse) has been measured to be approximately 0.95, the reduction in V_{Stokes} to (roughly) 0.5 after only 2 m of multimode fiber highlights the strong impact of the multimode fiber on the radiation's polarization properties. Furthermore, it is noteworthy, that the polarization properties are spatially uniform across the fiber end facet.

Remaining variations in Fig. 5.12d are mainly due to speckle patterns that are intensity variations caused by statistical interference effects across a given surface. For highly coherent radiation these patterns can be quite obvious, however, for lower coherence lengths they eventually vanish. Thus, the contrast of speckle patterns is an appropriate indicator for the coherence of radiation [97]. In multimode fibers the coherence of radiation propagating through the fiber is reduced due to the incoherent superposition of the intensity distributions of all present modes [95]. Therefore, multimode fiber approaches are also exploited in projector applications [98].

Speckle reduction in multimode fibers is demonstrated by the results in Fig. 5.13b. The measurements have been conducted by coupling ultrashort laser pulses with 100 fs pulse duration and 800 nm center wavelength into a multimode fiber with NA = 0.39 (FT200EMT/Thorlabs) of various lengths. For an evaluation of the speckle patterns the output facet of the multimode fiber has been imaged onto a CMOS-camera with a 4f-setup and the speckle contrast has been computed according to [98]

$$C = \frac{\sqrt{\langle P_{\rm i}^2 \rangle - \langle P_{\rm i} \rangle^2}}{\langle P_{\rm i} \rangle},\tag{5.26}$$

with P_i as intensity of the i-th camera pixel and $\langle \rangle$ denoting spatial averaging over all camera pixels.



Figure 5.13: (a) Enlargement of the spatial intensity distribution at the end facet of the FT200EMT/Thorlabs multimode fiber after 2 m (i) and 175 m (ii) fiber length. (b) Speckle contrast of a FT200EMT/Thorlabs fiber (NA = 0.39) at various lengths for ultrashort laser pulses of 100 fs pulse duration and 800 nm center wavelength. Please note, that the absolute values for speckle contrast are potentially biased by significant measurement uncertainties in this uncalibrated measurement.

The results demonstrate the general tendency of speckle contrast and, hence, coherence reduction while the radiation propagates through the multimode optical fiber. For compensating the ongoing variation of spatial intensity distribution with fiber length (see Fig. 5.11), the fiber output images have been normalized by their radially averaged intensity distribution prior to computation of the speckle contrast. However, the absolute values of speckle contrast might still suffer from measurement uncertainties in this uncalibrated measurement so that rather the general tendency shown in Fig. 5.13b should be noted instead of the absolute values of C. In Fig. 5.13a two enlargements of sections of the fiber end facet are shown for 2 m and 175 m fiber lengths. In the top image (2 m fiber lengths) speckle features of approximately 5 to 10 μ m size can be observed. For 175 m fiber length the speckles vanish and only fluctuations caused by noise remain. Both effects, the randomization of polarization and reduction of coherence, are welcomed side effects regarding the application of ultrashort laser pulses for characterization of current responses of solar cells. Besides temporal stretching and spatial homogenization of radiation inside a multimode optical fiber a further randomization of the distinct laser radiation properties in the polarization and coherence domain is greatly appreciated as this serves an improved imitation of the sun's radiation characteristics.

5.5 Fiber Concept for Conditioning of Ultrashort Pulses

The preceding sections demonstrated that spatial, temporal, polarization and coherence transformations of ultrashort laser pulses can be accomplished in a multimode optical fiber. In addition, multimode fibers can be readily implemented and provide a robust and efficient way of shaping the ultrashort pulses. Therefore, multimode fibers appear to be an ideal choice for the pulse-to-cw-conversion of an ultrashort pulse train, which is required for elimination of undesired nonlinearities in solar cell measurements (see Chapter 4).

Within the course of this work a fiber concept has been developed that conducts this conversion at minimum losses of optical power (see Fig. 5.14). Similar to previous approaches the fiber device utilizes many fibers of different lengths to split a single pulse into temporally equidistant sub-pulses for externally enhancing the pulse repetition rate of an incident pulse train [72, 73]. In addition to that, single multimode fibers are applied before and after the sub-pulse generation. These enable both, improved temporal mixing of the respective sub-pulses by additional pulse stretching and spatial homogenization yielding uniformity of sub-pulse generation and device output. The full fiber approach allows incorporation of above features in a monolithic tool, thus, enhancing applicability and robustness of the method. Furthermore, the spatially confined but homogeneous device output can be exploited for new measurement approaches as e.g. demonstrated in Chapter 8: EQE-Measurement of CPV Modules. Subsequently, the fiber concept and its design principles are presented. 20



Part 1: Multimode fiber Part 2: Fiber bundle delay lines Part 3: Multimode fiber

Figure 5.14: Schematic illustration of the monolithic pulse-to-cwconverter consisting of three parts merged by splicing technology. Part 1: multimode optical fiber for spatial averaging, Part 2: bundle of 19 fibers with individual lengths for generation of sub-pulses, Part 3: multimode fiber for spatial mixing and temporal broadening of the 19 sub-pulses. The evolution of the temporal pulse shape is illustrated by the four graphs on top showing estimated temporal shapes of the ultrashort pulse train at the respective positions of the pulse-to-cwconverter. The close-ups show a fiber bundle (bottom left) illuminated from the back and the bundle-to-fiber splice (bottom right) (image courtesy of S. Böhme, who was responsible for the fabrication of the pulse-to-cw-converter at Fraunhofer IOF, Jena, Germany)

²⁰At the time this thesis has been written, the pulse-to-cw-converter has still been under construction at the Fraunhofer IOF, Jena. However, a prototype version of the converter has been created beforehand demonstrating its functionality and has been successfully applied for the measurements presented in Chapter 8.

5.5.1 General Concept

The monolithic pulse-to-cw-converter is shown in Fig. 5.14 and consists of three fundamental parts.

Firstly, the ultrashort laser pulses are coupled into a multimode optical fiber that spatially homogenizes the radiation. The fiber length is chosen such that an appropriate homogenization at the distal end of the fiber is achieved (compare Section 5.4.1).

This end is then spliced to a fiber bundle of the same diameter consisting of 19 multimode fibers (see bottom left close-up in Fig. 5.14 that shows such a fiber bundle illuminated from the back, image courtesy of S. Böhme who was responsible for fabrication of the pulse-to-cw-converter at Fraunhofer IOF, Jena, Germany). The bundle is created by threading the 19 fibers in a capillary tube and subsequent tapering of the tube (reduction of its diameter) to ensure a firm connection of all fibers and to enable good cleaving of the bundle. Attributing different lengths to each of these 19 fibers, an incoming pulse is split into 19 sub-pulses that are temporally delayed with respect to each other (as e.g. in [72, 73]). The length differences ΔL of the 19 fibers are chosen in a way that the sub-pulses are equally distributed within the pulse period T of the ultrashort pulse train (length difference $\Delta L = Tc_0/n/19 \approx 13.55$ cm), thereby enhancing the pulse repetition rate of the pulse train by a factor of 19. The previous spatial homogenization in the first multimode fiber ensures that each of these sub-pulses carries the same optical power.

Finally, the 19 fibers are merged at their distal end to a fiber bundle and spliced to a single multimode fiber that recombines the individual sub-pulses spatially (see bottom right close-up in Fig. 5.14). Further, this highly multimode fiber is designed to temporally broaden the subpulses in a way that they temporally overlap with each other at the fiber's exit facet, thus, creating a continuous output signal (with a more or less pronounced amount of remaining fluctuations in the absolute amplitude along one pulse period, depending on the chosen fiber properties).

5.5.2 Design Principles

The realization of the previously described pulse-to-cw-converter requires consideration of several design rules or parameters that will be discussed subsequently.

Numerical Aperture

Firstly, the NA of a preceding fiber should not be larger than the NA of the next fiber components attached to this, thus,

$$NA_1 \le NA_2 \le NA_3. \tag{5.27}$$

If Eq. (5.27) is not fulfilled, optical power might be lost as some rays emitted by a large NA fiber exceed the total internal reflection angle of the next fiber with a smaller NA.

Besides that, the fiber NA is also an important design parameter regarding the temporal shaping abilities of the pulse-to-cw-converter, as a higher NA enhances temporal broadening [70, pp. 351-352]. Therefore, a high NA₃ should be chosen to increase the rate of temporal broadening and extend the range of possible NA's for NA₁ and NA₂.

Geometrical Properties

In addition to the impact of the fiber's NA, temporal broadening is also affected by the geometrical properties, thus, core and cladding diameters, of the fibers being used. This relation results from the dependence of small irregularities in a fiber, causing mode coupling (microbends), on the core and cladding diameter of the optical fiber. As e.g. shown by *Fermann* [99] smaller core diameters result in smaller coupling coefficients.

Regarding the impact of the mode coupling coefficient on a fiber's impulse response, it can be generally stated, that a low mode coupling coefficient yields increased temporal pulse broadening [100]. Phenomenologically this can be explained by considering two rays launched into a multimode fiber at different angles. In an ideal fiber without any mode coupling each ray maintains the same propagation angle, thus, their propagation time through an optical fiber will be different (and will scale with fiber length L [74, p. 53]). For mode coupling rates approaching infinity both rays will instead have identical propagation times as both have been partially propagating in all available fiber modes.²¹ Thus, for enhanced pulse broadening in multimode optical fibers reduced mode coupling is advantageous. This, not necessarily intuitive, feature can be demonstrated by simple numerical simulations based on the approach presented in Section 5.2.4. The results shown in Fig. 5.15a represent the impulse responses of a 500 m long multimode fiber with NA = 0.39 and a modal independent, thus, constant diffusion or mode coupling rate. For simplicity absorption has been neglected and an initially uniform modal distribution (uniform launch conditions) is assumed. The results clearly demonstrate that a reduced diffusion coefficient yields a significantly broader impulse response.²²

This simulated impact of the mode coupling coefficient is consistent with the experimental results shown in Fig. 5.15b. Here, impulse responses of identical fiber types with varying core diameters and 100 m length have been compared (FTx00EMT/Thorlabs). The experiments have been conducted with a 4f-setup imaging the fiber output onto a high speed photodiode (818-BB-45/Newport, 12.5 GHz bandwidth) connected to a 33 GHz oscilloscope (DSOX93204A/Agilent). As the launch conditions for all three measurements were very similar, the results demonstrate that a smaller core diameter, hence, a lower mode coupling coefficient [99], yields a broader impulse response.

Besides the impact of core diameter on impulse broadening in multimode optical fibers, the diameters of the respective pulse-to-cw-converter fiber components need to be adapted to each other. Naturally, the diameter 2a of any part of the converter should not be smaller than that of the preceding part, thus,

$$2a_1 \le 2a_2 \le 2a_3. \tag{5.28}$$

²¹The rate of pulse broadening scales with \sqrt{L} in that case [100].

²²It is noteworthy that for non-uniform modal conditions the relation of mode coupling coefficient and pulse broadening becomes significantly more complex. In fact, a higher mode coupling coefficient can result in broader impulse responses as the higher order modes, propagating at slower speeds through the fiber, are excited significantly earlier as for low mode coupling coefficients.



Figure 5.15: (a) Simulated impulse responses of a 500 m long multimode fiber with NA = 0.39 and varying diffusion or mode coupling properties. Reduced mode coupling leads to broader impulse responses. (b) Measured impulse responses of the FTx00EMT multimode fiber from Thorlabs with 200, 400 and 800 μ m core diameter and 100 m length each. As mode coupling reduces with core diameter, broader impulse reponses are obtained for smaller fibers.

Absorption Characteristics

Furthermore, the fibers should be chosen to minimize material absorption. This prevents damage to the fiber device and ensures a maximum output signal for the later measurements. Therefore, glass optical fibers seem to be a better choice than plastic optical fibers that typically exhibit significantly higher absorption.²³ A further reduction of absorption is achieved when fibers designed for distinct spectral ranges (UV, VIS or NIR) are used. In this work, two monolithic pulse-to-cw-converters are realized, one for the UV-to-VIS-region (270-520 nm) the other for the VIS-to-NIR-region (520-1800 nm). Details on their implementation in the Laser-SR-setup can be found in Section 6.2.4.

²³Liquid-core and photonic bandgap fibers are not considered here as their processibility is rather limited as compared to glass optical fibers.

Chapter 6

DSR-Setup Based on Ultrashort Laser Pulses

In this chapter the Laser-DSR measurement setup will be presented that has been developed in the course of this work. Starting from a short introduction motivating the application of lasers for measuring spectral responsivities (SRs) or external quantum efficiencies (EQEs), the ultrashort pulse laser system used within this work is presented. Afterwards, the strategies and concepts for spectral, temporal and spatial shaping of the radiation, making it applicable for solar cell measurements, will be outlined giving experimental and simulated results on the performance of the developed system. The optical losses going along with the shaping of the laser radiation will be analyzed subsequently, pointing out remaining potential for an enhancement in optical efficiency. Then, the measurement uncertainties of the DSR-setup will be discussed in detail demonstrating the capabilities of the new setup and revealing remaining potential for further reduction of uncertainties. Finally, the chapter is concluded by recapitulating its most relevant results and comparing the new facility to state-of-the-art DSR-measurement setups.

6.1 Introduction to DSR-Measurements

The SR of a solar cell, or equivalently used the EQE, is an important measurand for solar cell characterization and calibration. The absolute SR allows for a highly accurate determination of the solar cell's short circuit current ($I_{\rm SC}$), which is the major photovoltaic parameter for solar cell calibration (see Section 2.2). Furthermore, the (relative) SR enables a spectrally resolved analysis of solar cell features and a spectral mismatch correction (see Section 2.2.2.1).

The most widely spread and accepted method for measuring the SR or EQE of a solar cell is the differential spectral responsivity (DSR) method [14] (see Section 2.2.2.2). In this method, the test device is illuminated with a bias light source of variable irradiation level and chopped monochromatic radiation. Using transimpedance and lock-in amplifiers the test cell is kept at short circuit conditions and its differential current response to the chopped monochromatic light can be extracted. Not only that the DSR-approach provides the lowest reported measurement uncertainties, it also allows for detecting comparably low signals owing to the lock-in measurement technique. Moreover, the test cell's linearity can be determined simultaneously by applying different levels of bias irradiation, thus, exciting a different excess carrier density within the test cell.

Due to the importance of precise SR-measurements in calibration of solar cells and driven by economic aspects and technological advances, there is a steady development of new DSR-measurement facilities featuring reduced measurement uncertainties. In most state-of-the-art setups, broadband white light sources, like xenon or tungsten halogen lamps, are applied that are spectrally filtered by a (double-)grating-monochromator [101–105] or a combination of various filters [106, 107]. However, the intrinsic limitation in spectral power of the available white light sources often impedes a further reduction of measurement uncertainties in such setups. A promising approach to overcome this limitation is provided by application of spectrally narrow, ideally monochromatic, radiation sources. Although being spectrally still quite broad, the advent of light-emitting diodes (LEDs) and their nowadays availability over wide spectral ranges pushed the development of several LED-DSR-setups recently [108–110]. A spectrally more narrow alternative to LEDs is represented by lasers that are not only superior because of their spectral bandwidths, but also because of their availability in a spectrally tunable manner allowing for a free choice of center wavelength. Therefore, lasers were suggested [15, 16] and, very recently, been successfully applied for a further reduction of measurement uncertainties [17].

In the subsequent sections the development of a laser-based DSRmeasurement facility at Fraunhofer ISE CalLab is presented, that has been conducted in the course of this work.

6.2 Setup Description and Characterization

As concluded in the previous section the high spectral power of lasers as compared to conventional sources can be exploited for the development of novel DSR-facilities featuring significantly lower measurement uncertainties. However, the monochromatic nature of lasers also implies that either multiple laser sources at different wavelengths or tunable laser sources are necessary for covering the wide spectral ranges required in such setups. As, for convenience and for their enhanced application potential, tunable laser sources are typically favored over a multiple-source approach, wide range tunable lasers are required.

Unfortunately, wide range tunable lasers emitting continuous wave (cw) radiation are typically rather limited in their spectral range (e.g. laser diodes, solid-state lasers) or their handling (e.g. dye lasers) [111]. Taking advantage of nonlinear optical processes, as e.g. parametric amplification or frequency doubling (see Section 3.2.2), these limitations can be overcome. In fact, there has been some significant progress in the development of cw optical parametric oscillators (OPOs) recently, that are now able to (partially) cover the ultraviolet (UV) (e.g. [112]) and visible (VIS) (e.g. [111]) region, in addition to the near-infrared (NIR) and mid-infrared spectral region that were made available earlier (e.g. [113]). In such systems the comparably low nonlinear conversion efficiency of cw radiation is compensated for by rather sophisticated resonator layouts

(e.g. in doubly- or even triply-resonant scheme) and tight control of the oscillator and crystal properties.

In contrast to this, high efficiency in nonlinear optical processes is achieved when using pulsed laser systems and, especially, when using ultrashort pulses owing to their extreme pulse peak powers.¹ As this yields less demanding optical concepts being more robust and user-friendly, (automated) wide range tunable ultrashort pulse laser systems are commercially available. Although usage of such laser systems entails further steps for conditioning the radiation in its spectral and temporal properties, their advantages regarding spectral range coverage, automation and availability outperform cw systems these days, when it comes to the development of a Laser-DSR measurement facility.

For the reasons given above, a tunable titanium-sapphire ultrashort pulse laser (Ti:Sa fs-laser) has been combined with a frequency doubling (SHG) and tripling unit (THG) as well as an OPO for the development of a new Laser-DSR facility in the course of this work. Fig. 6.1 shows a schematic illustration of the new measurement setup. The laser radiation is generated by above mentioned components (further discussed in Section 6.2.1) and steered through the setup in a fully-automated manner by computer-controlled radiation sources and movable mirrors. Depending on the chosen wavelength and measurement configuration (details in Section 6.2.2), the radiation is steered to the appropriate spectral shaping element for filtering undesired spectral components (e.g. remaining fundamental radiation after frequency doubling) and/or for reducing the radiation bandwidth (discussed in Section 6.2.3). Afterwards, the radiation is (spatio-)temporally shaped by the fiber concept introduced in Section 5.5 and the optics of the measurement plane (further discussed in Section 6.2.4). For establishing the DSR-principle the cell under test is illuminated with steady broadband bias irradiation plus narrow band radiation, that is frequency-modulated by mechanical chopper wheels. Comparing the differential current increase induced by the chopped light to the differential increase from a known reference cell (substitution method), the DSR

¹Naturally, high energy nanosecond pulses also allow for efficient nonlinear processes, however, their pulse repetition rates impede the temporal conversion from pulsed to cw radiation as discussed in Chapter 5.



Figure 6.1: Schematic illustration of the Laser-DSR setup that has been developed in the course of this work. Ultrashort laser pulses (fs-pulses) are spectrally, temporally as well as spatially shaped and illuminate a solar cell for measuring its DSR.

of the test cell is achieved. Furthermore, the monitor method is realized by a monitor unit tracking intensity fluctuations of the incident radiation. The electrical equipment for conducting the DSR-measurements consists of transimpedance and lock-in amplifiers and is detailed in Section 6.2.5.

6.2.1 Radiation Sources

The main radiation source is a Ti:Sa laser (*MaiTai HP*/Newport Spectra-Physics) that generates ultrashort pulses of approximately 100 fs pulse duration tunable in the spectral range from 690 nm to 1040 nm. The ultrashort pulses are generated by means of regenerative mode-locking using an acousto-optical modulator driven by the cavity frequency measured with a photodiode. The fully-automated spectral tuning is accomplished by a wavelength-calibrated tuning slit placed between two prisms, that compensate for group velocity dispersion in the cavity.

The ultrashort pulses emitted by the Ti:Sa laser are steered to an OPO (*Inspire HF*/Newport Spectra-Physics) that is generating so-called *signal* and *idler* radiation at 490 nm to 750 nm and 900 nm to 2500 nm center wavelength by means of parametric amplification (see Fig. 3.4 in Section 3.2.2.1). For this, Ti:Sa pulses at 820 nm are frequency-doubled to 410 nm inside the OPO and used for pumping the parametric oscillator. By rotation of the nonlinear crystal for parametric amplification phasematching is achieved and specific signal and idler wavelengths are generated (for phase-matching see Section 3.2.4). Adjusting the OPO cavity length, resonance for the signal wavelength is realized enhancing nonlinear conversion efficiency. In contrast to the Ti:Sa, the fully-automated wavelength control of the OPO is not conducted by a calibrated slit but by an internal spectrometer.

For coverage of the UV to blueish spectral region the Ti:Sa pulses are steered to a SHG/THG unit (*HarmoniXX*/APE) (for SHG and THG see Section 3.2.2.1). The incident pulses (fundamental radiation) are focused onto a nonlinear crystal for generation of the second harmonic radiation (345 nm to 520 nm). Phase-matching of fundamental and second harmonic is achieved by tuning the angular position of the crystal. After passing a compensator plate (that corrects for the wavelength dependent refraction at the second harmonic crystal) and a delay compensator (that corrects for the wavelength dependent delay), a waveplate rotates the polarization of the depleted fundamental so that fundamental and second harmonic have identical polarizations. The fundamental and second harmonic are then focused onto another nonlinear crystal that creates the *third harmonic* (230 nm to 347 nm) by sum-frequency generation. Again, phase-matching is controlled via angular tuning of the crystal. The three wavelength components fundamental, second and third harmonic are subsequently collimated, separated by two pairs of dichroic mirrors and steered to individual outputs. For full coverage of the entire spectral range from 230 nm to 520 nm an appropriate dichroic mirror pair has to be chosen and placed in the device. In order to achieve a fully-automated device, the SHG/THG unit has been modified by placing the dichroic mirror pairs on an external, computer-controller translation



Figure 6.2: (a) Measured FWHM bandwidths and pulse durations as emitted from the ultrashort pulse laser system. (b) Average output power of ultrashort pulse laser system.

stage. Furthermore, a calibration routine has been developed that relates any desired wavelength output to certain positions of the motors for crystal rotation, waveplate tilt and dispersion compensation.

In Fig. 6.2a measured full-width-at-half-maximum (FWHM) bandwidths and pulse durations and in Fig. 6.2b measured average output powers of the ultrashort pulse laser system are shown. Due to the spectral limitations of the applied autocorrelator (PScout HR/Newport) no pulse duration measurements have been conducted for THG, SHG and signal radiation. The increase in spectral bandwidth with wavelength results from the nearly constant pulse duration and time-bandwidthproduct over wavelength (compare Eq. (3.1)). The presented results demonstrate that the applied ultrashort pulse laser system enables gapfree wavelength-tuning from 230 nm to 2500 nm. In spectral overlap regions, in which two systems might generate radiation, Ti:Sa and SHG are preferred over the OPO for power output and stability reasons.

6.2.2 Modes of Operation

Besides gap-free wavelength-tuning from 230 nm to 2500 nm the ultrashort pulse laser system also features a simultaneous DSR-measurement at two quasi-monochromatic wavelengths, thereby allowing for a significant reduction of measurement duration.

This feature mainly results from the intrinsic presence of at least two wavelengths when nonlinear effects are exploited (e.g. fundamental/second harmonic or signal/idler). Especially signal and idler wavelengths are always present at the same time and, hence, can be readily used simultaneously without any reduction of spectral power in either signal or idler radiation. When the second or third harmonic are generated, the remaining fundamental Ti:Sa radiation could be used likewise. The involved reduction of average Ti:Sa power could be endured, as significantly more power is provided by the Ti:Sa as compared to the other spectral ranges (see Fig. 6.2b). As it appeared to be technically more feasible, a beam splitter is applied to steer a portion of the Ti:Sa radiation directly to the measurement plane while the other portion is simultaneously used for SHG or THG, instead of using the remaining fundamental after nonlinear conversion (see also Fig. 6.1). Furthermore, the applied lock-in amplifiers allow for a simultaneous detection of two signals as long as they are modulated at different frequencies.

In summary, laser system and electrical equipment readily allow for an implementation of the so-called *dual reference* mode (in contrast to *single reference* mode), the simultaneous DSR-measurement at two different wavelengths. Therefore, the Laser-DSR system and its beam steering configuration has been designed to enable both operational modes. Switching between the modes is conveniently implemented into the user interface of the developed measurement software.²

6.2.3 Spectral Shaping of Ultrashort Laser Pulses

Although being quasi-monochromatic, the ultrashort pulse nature of the radiation emitted by the applied laser system yields significant radiation bandwidths that would increase the uncertainty of the DSR-measurements (compare Fig. 6.2a). Furthermore, undesired spectral components might be present in the laser radiation, especially after nonlinear conversion processes, that need to be blocked by filters or monochromators.

 $^{^{2}}$ The developed software is not discussed in this thesis.

6.2 Setup Description and Characterization

As a general guideline, the new Laser-DSR setup aims for bandwidths smaller than 5 nm throughout the available spectral range. Consequently, no further reduction of THG and SHG spectral bandwidths are required (compare Fig. 6.2a). In contrast, monochromators are required for the signal, Ti:Sa and idler radiation to reduce their spectral bandwidths. Furthermore, signal and idler need separate monochromators due to the desired dual reference mode configuration discussed above. This appears to be a conceptual disadvantage of the dual reference mode only on first sight, as the necessity of using two monochromators can be exploited by using both a prism and a grating monochromator, thereby taking advantage of their individual benefits for the VIS-NIR or NIR spectral range. Subsequently the spectral shaping approaches for the UV-VIS, VIS-NIR and NIR spectral range will be discussed.

UV-VIS (THG, SHG): Bandpass Filters

Although dichroic mirrors are used to separate the fundamental, second harmonic and third harmonic radiation at the output of the APE, some unwanted radiation remains in the respective beams. For (eventually) eliminating this unwanted radiation the THG and SHG output of the APE are equipped with 2mm thick UG11 and BG40 bandpass filters.

The transmission curve of the UG11 bandpass filter shown in Fig. 6.3 (black curve) demonstrates that it significantly absorbs wavelengths shorter than 270 nm, thereby reducing the overall available spectral range of the setup. However, the subsequent fibers and optics for spatio-temporal shaping of the radiation (as detailed in Section 6.2.4) reject wavelengths below approximately 270 nm anyway. Furthermore, wavelengths below 270 nm play a minor role in PV as the solar spectral irradiance in that regime is virtually negligible. Thus, a limitation of the available spectral range to wavelengths longer than 270 nm is reasonable. A way more substantial conclusion from Fig. 6.3 is the absorption of wavelengths from 405 nm to 520 nm (corresponding to the second harmonic wavelength range required for THG, see Section 3.2.2.1) and above 810 nm (corresponding to the fundamental wavelengths required for the respective THG), thereby blocking undesired radiation from the



Figure 6.3: Transmission of spectral filters added to the THG and SHG output of the APE. The UG11 is applied to the THG output and blocks the remaining SHG and fundamental radiation from the THG output, the BG40 is applied to the SHG output and blocks the remaining fundamental radiation from the SHG output.

THG beam significantly. For determination of the amount of undesired radiation after insertion of UG11 filter into the THG beam path, spectral measurements without the filter have been conducted. Multiplication of these results with the UG11 transmission curve shown in Fig. 6.3 revealed a relative optical power of undesired radiation in the THG radiation $< 10^{-6}$ (without filter the relative optical power was measured to be approximately 0.3%). Consequently, the THG output beam can be regarded as consisting purely of the desired third harmonic.

A very similar argumentation can be performed for the BG40 filter and the second harmonic radiation. However, as in case of SHG the third harmonic crystal is detuned so that no third harmonic is generated, only the depleted fundamental needs to be considered in the discussion. From equivalent spectral measurements as for the THG output, the relative optical power of the undesired fundamental wavelength in the SHG output is determined to be < 0.02% after inserting the BG40 filter. In contrast to the UG11 filer, the BG40 partially transmits relevant radiation from 690 to 700 nm (see red curve in Fig. 6.3), so that the value of 0.02% appears in the SHG spectral range from 345 to 350 nm. Above 350 nm the relative amount of undesired radiation drops to $< 10^{-5}$, which is similar to the blocking performance in THG.

Although being virtually negligible, the remaining undesired spectral components in the THG and SHG output are considered in Section 6.4.1.1 with respect to the spectral responsivity of the measured solar cells. As expected from the discussion in this subsection, the uncertainty analysis will point out that uncertainties due to wavelengths blocking are insignificant.

VIS-NIR (Signal, Ti:Sa): Prism Monochromator

As apparent from Fig. 6.2a the signal and Ti:Sa radiation (520 nm to 1040 nm) require spectral shaping for fulfilling the chosen 5 nm bandwidths condition. In the new Laser-DSR setup this spectral shaping is conducted by a prism monochromator that features a higher transmission efficiency as compared to a grating monochromator as all wavelengths are dispersed in a single order (see Fig. 6.4). Furthermore, the Brewster condition can be exploited as both signal and Ti:Sa are p-polarized, thereby significantly reducing reflection losses.³ For the conditions given in this work, the higher transmission efficiency outperforms the reduced dispersion of a prism as compared to a grating in this spectral range.⁴

The prism monochromator of the new Laser-DSR setup has been developed in the bachelor thesis of D. Lill under supervision of the author of this work [114]. In the following, the general concept of the setup and some performance results will be discussed.

The optical layout of the prism monochromator is equivalent to a 4fsetup (see Fig. 6.4). The apex of the first prism is placed in the front focal plane of a parabolic mirror so that the dispersed spectral components are collimated by the mirror and directed to the slit plane at the mirror's back focal plane. After the slit plane, a second mirror and a second prism are placed for recombining all wavelength components to a single

³The Brewster angle is given by $\theta_{\rm B} = \arctan(n_2/n_1)$ [70, p. 212]. Although only valid for a single wavelength, the overall reflection at the prism surface remains very low even when neighboring wavelengths are considered.

⁴Please note that prism dispersion drops for infrared wavelengths making grating monochromators more suitable in such spectral ranges.



Figure 6.4: Schematic illustration of the prism monochromator principle. The spectrally dispersed incident radiation is collected by a parabolic mirror that focuses each wavelength to a single spot in the slit plane. A movable slit with variable slit width selects a certain wavelength and bandwidth, that is collected by another parabolic mirror and prism subsequently.

beam facilitating further usage of the radiation. As identical wavelengths propagate parallel after the first prism, each wavelength is focused in the slit plane (as illustrated in Fig. 6.4). Thus, by laterally moving the slit, a desired wavelength at high spectral resolution can be chosen. This 4fdesign is specifically advantageous as only a single movable component, the slit, is required for the entire spectral range making the developed prism monochromator very robust.

The spectral resolution of the prism monochromator is defined by the linear dispersion and focal spot size in the slit plane. As both, linear dispersion in the slit plane

$$\frac{dx}{d\lambda} = f \frac{d\theta}{d\lambda} \tag{6.1}$$

and focal spot size

$$w_f \approx \frac{\lambda f}{\pi w_0} \tag{6.2}$$

scale linearly with focal length of parabolic mirror f, the spectral resolution is solely sensitive to the angular dispersion of the prism $d\theta/d\lambda$ and the collimated beam radius w_0 . Ray optical simulations have shown



Figure 6.5: (a) FWHM bandwidth over slit width of the prism monochromator for various wavelengths. (b) FWHM bandwidth after the prism monochromator as well as power loss at center wavelength and total power loss of integrated peak by the prism monochromator at 200 μ m slit width.

that N-SF11 glass type prisms are most appropriate for the considered spectral range. Achromatic lens telescopes enlarge the beam diameters of both signal and Ti:Sa radiation prior to hitting the first prism surface. The slit width can be varied from 0 to 5000 μ m in 10 μ m step sizes.

The impact of slit width is apparent from Fig. 6.5a that demonstrates the decrease in FWHM of the radiation for decreasing slit widths. The results demonstrate that bandwidths below 5 nm can be achieved for the entire spectral range when choosing a slit width of approximately 100 μ m. As such small slit widths are as well causing a reduction in spectral power of the peak (or central) wavelengths, a standard slit widths of 200 μ m is chosen yielding the performance shown in Fig. 6.5b. The apparent reduction in spectral power of the center wavelengths (filled blue symbols in Fig. 6.5b) are of intrinsic nature, as they are solely caused by the optical components used in the monochromator and not by the slit itself.⁵ As the 5 nm bandwidth condition is violated with a 200 μ m slit for wavelengths longer than 900 nm, the adjustable slit is not replaced by a fixed slit. This

⁵Please note that these losses are also shown in Fig. 6.8, incorporated in the prism, lens and beam pointing losses.

readily allows for reducing the slit width to 100 μ m (or even less) in order to achieve bandwidths smaller than 5 nm.⁶

NIR (Idler): Grating Monochromator

The results shown in Fig. 6.5 indicate the performance reduction of prism monochromators with increasing wavelength. Therefore, a commercially available grating monochromator (*iHR 320*/HORIBA) is applied to the idler radiation. The grating monochromator is optimized for usage with optical fibers at its input and output by application of mirror and lens adapters. The triple-grating turret is equipped with a 600 lines/mm and a 300 lines/mm grating enabling optimized measurements regarding repeatability, bandwidth and power throughput. Additionally, the output slit is motorized for convenient changes to the transmitted spectrum. The third turret position is equipped with a mirror that enables throughput of the entire optical power if desired.⁷

6.2.4 Spatio-Temporal Shaping of Laser Pulses

As illustrated in Fig. 6.1 the spectrally shaped laser pulses are subsequently shaped in their temporal and spatial properties, which will be discussed in this section.

From the model derived in Chapter 4: Interaction of Ultrashort Laser Pulses and Solar Cells it was concluded that the ultrashort laser pulses should be converted into continuous radiation for eliminating potential nonlinear effects in the measurements. In Chapter 5: Temporal Shaping of Ultrashort Laser Pulses and especially in Section 5.5 a fiber-based monolithic device was presented that enables such a transformation of the ultrashort pulses.⁸ It has also been demonstrated in Section 5.4.1 that the multimode fiber approach does not only yield temporal but also spatial

 $^{^{6}}$ In a further development stage of the prism monochromator, a step motor can be attached to the slit making a variation of the slit width even more convenient.

⁷An additional longpass filter ensures that undesired spectral components below 900 nm are blocked.

 $^{^{8}\}mathrm{As}$ this device was discussed in detail beforehand, the temporal shaping is not further addressed in this section.

homogenization of the radiation which is an important property of solar cell characterization tools. In Chapter 8: *EQE-Measurement of CPV Modules* one measurement approach is presented that exploits exactly this feature of multimode optical fibers.

However, the new Laser-DSR measurement setup aims at absolute SRmeasurements for which illumination of the entire solar cell surface with uniformly distributed monochromatic radiation is beneficial. Outshining large area solar cells with typical edge lengths of approximately 15.6 cm in so-called overfilled measurements, hence requires a rather difficult 550xmagnification of the fiber output surface (assuming a 400 μm fiber core diameter). Furthermore, the concept of imaging the homogeneous fiber output onto the measurement plane in combination with the dual reference mode would require that all wavelengths are emitted by a single fiber resulting in additional absorption losses. The discussed difficulties going along with a direct imaging of the multimode fiber output motivate the development of a refined imaging setup that is discussed subsequently.

For combining the radiation from the different pulse-to-cw-converters (compare Fig. 6.1) they are placed close to the input facet of a mixing rod with hexagonal cross section. Besides spatially combining the radiation from the different fiber outputs, the hexagonal cross section of the mixing rod and the rather high NA input from the fiber converters (compare Section 5.5.2) yield a spatially homogeneous intensity distribution at the exit facet of the mixing rod. Thus, the exit facet of the mixing rod is kind of an equivalent to the fiber converter outputs regarding the spatial irradiance uniformity, but is significantly larger in spatial dimension, thereby simplifying its imaging onto the measurement plane.

Starting from this general concept, optical simulations with the raytracing software OptiCAD[®] have been conducted for optimizing the properties of the optical components with respect to the size and uniformity of the illuminated area as well as its wavelength sensitivity. The final optical configuration composed of a hexagonal mixing rod, a lens, a flat tilted mirror and a parabolic mirror as well as simulated results of the optical setup are shown in Fig. 6.6. Fig. 6.6a illustrates the combination of different optical components to achieve a uniformly illuminated area of 18x18 cm². The close-up shows the mixing rod and the lens (the fibers,



Figure 6.6: Layout and simulated results of the optical setup for illuminating a 18x18 cm² area with spatially uniform irradiance distribution. (a) Screenshot of optical setup implemented in ray-tracing software. Blue to red areas denote the illumination intensity from low to high. The green rays illustrate the beam paths. (b) Simulated non-uniformity over number of rays considered in simulation. (c) Simulated intensity distribution of a 16x16 cm² area for 10 million rays resulting in a non-uniformity of approximately 3%.

not shown here, are placed at the left side of rod). Lens and steering mirror direct the radiation emitted from the rod onto a parabolic mirror. The combination of lens (short focal length) and parabolic mirror (large focal length) result in a magnified image of the mixing rod output facet in the measurement plane (note the hexagonal intensity structure around the 18×18 cm² aperture). The short focal length of the lens ensures a higher magnification and avoidance of kaleidoscopic effects from the rod's inner

surfaces. Furthermore, as the lens merely re-directs or collects the emitted rays in order to ensure that they hit the parabolic mirror, the optical layout is rather insensitive to chromatic aberrations induced by the lens. Fig. 6.6c shows the simulated centric $16x16 \text{ cm}^2$ intensity distribution for 10 million rays resulting in a non-uniformity of approximately 3%. For a selected $2x2 \text{ cm}^2$ area (typical reference solar cell area) a non-uniformity below 0.2% is achieved. The simulated non-uniformity strongly depends on the number of rays (see Fig. 6.6b), thus, less than 3% non-uniformity are expected for the final setup.⁹

In addition to the $18\times18 \text{ cm}^2$ illumination setup shown in Fig. 6.6a, the new Laser-DSR setup can be conveniently equipped with further optical components for uniformly irradiating a $5\times5 \text{ cm}^2$ area. This is especially advantageous when smaller devices are measured, as the monochromatic irradiance is increased by a factor of 13. For this, the fibers are plugged to a second hexagonal mixing rod and another combination of steering and parabolic mirror as well as aperture (see Fig. 6.7a, for convenience the other rod and lens - used for the large area setup - are not shown here).

The spatial intensity distributions obtained from (i) ray-tracing simulation and (ii) measurement shown in Fig. 6.7b demonstrate the high spatial uniformity being achieved with this optical setup (please note that the scale is identical to the scale in Fig. 6.6c). In fact, the best non-uniformity for a $2x2 \text{ cm}^2$ area is (i) 0.14% and (ii) 0.23% (for a $4x4 \text{ cm}^2$ area it is (i) 0.80% and (ii) 1.05%). Fig. 6.7b also indicates that the simulated intensity distribution resembles the measured one, especially regarding the tendency of a slight drop in intensity when approaching the edges of the measurement plane. Finally, Fig. 6.7c illustrates first results regarding the insensitivity of the setup to wavelength changes (here from 700 to 1000 nm, note that the scale is different as compared to Fig. 6.7b). Apart from the obvious similarity of both irradiance distributions, their nonuniformities at identical positions are very similar: for a 2x2 cm² area the non-uniformity for 700 nm equals 0.24% and for 1000 nm it equals 0.23%, for a $4x4 \text{ cm}^2$ area the respective non-uniformities are 0.93% (700 nm) and 1.05% (1000nm).

 $^{^{9}}$ At the time this thesis has been written the optical components for achieving a $18x18 \text{ cm}^2$ illuimination area have not been installed yet.


Figure 6.7: Optical layout and intensity distribution for illuminating a $5x5 \text{ cm}^2$ area with a spatially uniform irradiance distribution. (a) Screenshot of optical setup implemented in ray-tracing software. Blue to red areas denote the illumination intensity from low to high. The green rays illustrate the beam paths. Steering and parabolic mirror as well as $5x5 \text{ cm}^2$ aperture are easily implemented into the large area setup (compare Fig. 6.6a). For convenience rod and lens of the large area setup are not shown here. (b) (i) Simulated and (ii) measured intensity distribution in measurement plane at the same scale as Fig. 6.6c. Best non-uniformity for a $2x2 \text{ cm}^2$ area is (i) 0.14% and (ii) 0.23%, for a $4x4 \text{ cm}^2$ area it is (i) 0.80% and (ii) 1.05%. (c) Measured intensity distributions at (i) 700 nm and (ii) 1000 nm wavelength (please note the different scales as compared to (b)). Nonuniformities for a $2x2 \text{ cm}^2$ area at idential positions are (i) 0.24% and (ii) 0.23% as well as (i) 0.93% and (ii) 1.05% for a $4x4 \text{ cm}^2$ area.

The results presented in this subsection demonstrate the excellent uniformity achieved with the optical concept presented in Figs. 6.6a and 6.7a. Moreover, the measurement setup can be switched from uniform illumination of a larger (max. $18 \times 18 \text{ cm}^2$, with an expected 3% non-uniformity over a $16 \times 16 \text{ cm}^2$) to a smaller area (max. $5 \times 5 \text{ cm}^2$, with a 0.23% nonuniformity over a $2 \times 2 \text{ cm}^2$), thereby enhancing irradiance, hence, measurement signal by a factor of 13. During the completion of the large area setup, that was being conducted at the time this thesis has been written, a neat mechanical solution enabling a convenient change of the measurement setup has been developed. The entire setup conversion is accomplished by simply re-attaching the fibers to the second rod and sliding in a frame holding the parabolic and steering mirror as well as the aperture for the small area setup. Furthermore, first experimental results support the expected insensitivity of the chosen optical setup to wavelength variations. Not only that this feature reduces measurement uncertainties (being discussed in Section 6.4), the wavelength-insensitivity is as well of tremendous practical importance as it significantly reduces effort when calibrating the setup.

6.2.5 Peripheral Equipment

After the previous discussion covering generation and shaping of the monochromatic radiation, the peripheral equipment required for conducting DSR-measurements is given in this section.

For obtaining a broadband bias irradiation multiple halogen lamps of two different types are used (13095/Philips, ALU PRO 56 50W/Philips), that differ slightly in their spectral properties. As the ensemble of each lamp type is controlled individually by fully-automated power supplies $(TOE \ 8872-130/\text{Toellner})$, an improved spectral match of bias irradiation and AM1.5g spectrum can be achieved as compared to using only one halogen lamp type.

Monitor unit (consisting of a silicon and germanium diode, S1337/Hamamatsu and J16-P1-R10M-SC/Teledyne Judson Technologies) and test or reference solar cell are connected to two in-house manufactured transimpedance amplifiers ensuring zero voltage across the respective device junction. The monochromatic signal is chopped by three chopper wheels (MC2000/Thorlabs), that transmit their frequency to two lock-in amplifiers (7265/Ametek) detecting the modulated voltage drop across the feedback resistors of the transimpedance amplifiers. In dual reference mode, the chopper frequencies are set by the internal oscillator frequencies of the lock-in amplifiers.

6.3 Optical Efficiency and Loss Analysis

The optical components used for laser beam routing, spectral, temporal and spatial shaping of the ultrashort laser pulses (see Sections 6.2.2 to 6.2.4) induce losses to the laser radiation that are discussed in this section.

In Fig. 6.8 the total power loss (white line, referring to the left axis) and the relative losses resulting from the various components of the setup are shown. Please note, that losses related to spectral shaping are not included in this plot as they are of intrinsic nature in the Laser-DSR setup and cannot be avoided by choosing other optical components. Instead, Fig. 6.8 illustrates remaining efficiency potential, if a reduction in the total loss is required. For example, the figure displays that the major part of losses are related to fiber absorption that might be compensated for by improved optical fibers. Fiber coupling losses, not to be confused with fiber reflection losses caused by Fresnel reflection, are estimated from experimental measurements to be 10% at each fiber entrace facet. The mentioned lenses are used in the prism monochromator and also for coupling the radiation into the multimode fibers.

In Fig. 6.9 the optical output power of the Laser-DSR setup is shown. The drop from pure laser output power (dashed black line) to the single reference mode without spectral shaping (blue line) corresponds to the total loss curve shown in Fig. 6.8 (white line). Taking the bandwidth reduction down to 5 nm into account yields the green curve in single reference mode. As no monochromators are used for wavelengths below 520 nm, the output power stays unaltered in that spectral region. Changing the operational mode of the Laser-DSR setup from single to dual ref mode (both including spectral shaping), by inserting a beam splitter into the Ti:Sa beam path, yields the red curve. As the dual reference mode does not affect the OPO radiation, the output power of



Figure 6.8: Total loss (white line, referring to left axis) and the relative contributions to this loss from various components of the Laser-DSR setup for spatial and temporal shaping. Losses related to a reduction of the spectral bandwidths are not included.



Figure 6.9: (a) Output power of new Laser-DSR setup; single ref mode without spectral shaping indicates the maximum available optical power of the setup (blue line). The dashed line represents the output power of the laser itself (see also Fig. 6.2b). (b) Laser-DSR setup in dual reference mode compared to state-of-the-art DSR-measurement facilities being based on spectrally filtered xenon (black) and halogen lamps (blue lines). The top red area illustrates the changes in output power of the Laser-DSR setup with bandwidth variation.

signal (520 nm $< \lambda < 690$ nm) and idler ($\lambda > 1040$ nm) remain unchanged. In contrast to this, a significant drop in the output power of the Ti:Sa (690 nm $\leq \lambda \leq 1040$ nm) radiation is observed caused by the applied beam splitter and going along with a slight drop of power for THG (270 nm $\leq \lambda < 345$ nm) and SHG (345 nm $\leq \lambda \leq 520$ nm). However, as apparent from the figure the loss in optical power of Ti:Sa radiation can rather be regarded as a leveling of the overall output power.

Fig. 6.9b demonstrates that the new Laser-DSR facility yields a significantly higher output power as typical DSR-measurement systems based on spectrally filtered white light sources. In fact, a 10 to 500 times higher optical output power is achieved with the Laser-DSR setup that has been developed in the course of this work as compared to the filter monochromator system. Compared to a grating monochromator setup 100 to 10,000 times more power is achieved. As this increase yields a significantly higher measurement signal, measurement time and standard deviation will be considerably reduced. Furthermore, smaller bandwidths can be realized at sufficient optical powers regarding the measurement signal. Also, the available spectral range is enhanced which might be beneficial for new solar cell concepts being under development and becoming sensitive to the infrared spectral region.

6.4 Measurement Uncertainty Analysis

The DSRs (denoted by $\tilde{s}(\lambda)$)¹⁰ measured with the setup introduced above are conducted by a twofold measurement applying the substitution and the monitor method. Firstly, the differential current response of a reference cell (RC) of known \tilde{s}^{RC} is divided by the monitor diode differential current response to obtain $Q^{\text{RC}} = j^{\text{RC}}/j^{\text{M,RC}}$. Then, the reference cell is replaced by the test cell (TC) and likewise $Q^{\text{TC}} = j^{\text{TC}}/j^{\text{M,TC}}$ is measured. With these two measurements the test cell's differential spectral

¹⁰Please note that the DSR equals the SR, denoted by $s(\lambda)$, in case of linear cells or for an appropriate level of bias irradiation [115] (see also Section 2.2.2.2). Further, EQE $(\lambda) = s(\lambda) hc_0/(q\lambda)$ with $s(\lambda)$ in units of A/W (thus, generated current relating to radiant flux) or EQE $(\lambda) = s(\lambda) hc_0/(q\lambda A_{cell})$ with cell area A_{cell} and $s(\lambda)$ in units of A/W · m² (thus, generated current relating to irradiance).

6.4 Measurement Uncertainty Analysis

responsivity \tilde{s}^{TC} is obtained according to

$$\tilde{s}^{\mathrm{TC}} = \tilde{s}^{\mathrm{RC}} \frac{Q^{\mathrm{TC}}}{Q^{\mathrm{RC}}} = \tilde{s}^{\mathrm{RC}} \frac{j^{\mathrm{TC}}/j^{\mathrm{M,\mathrm{TC}}}}{j^{\mathrm{RC}}/j^{\mathrm{M,\mathrm{RC}}}}.$$
(6.3)

For an appropriate description of the measurement outcome, Eq. (6.3) needs to be expanded by several terms that allow for a correction of deviations from ideal measurement conditions and an analysis of the measurement uncertainties. Subsequently, these correction terms are introduced starting with those that are specifically relating to the new setup (see Section 6.4.1). Afterwards, further terms are described that arise from the reference and test object and the peripheral equipment of the measurement setup (see Section 6.4.2). In the final subsection 6.4.3, the combined measurement uncertainty is determined and discussed by means of a SRmeasurement conducted with the new Laser-DSR setup. The uncertainty discussion will be concluded by presenting the uncertainty in short circuit current measurements and a comparison of the new setup to other state-of-the-art measurement facilities.

6.4.1 Optical Measurement Uncertainties

6.4.1.1 Spectral Uncertainties

Spectral uncertainties in measuring $\tilde{s}^{\text{TC}}(\lambda_0)$ arise from the following contributions being discussed in this subsection:

- Deviations from the center wavelength λ_0
- Current generation from undesired spectral components λ_{bl}
- Limited bandwidth of the quasi-monochromatic radiation λ_{bw}

A deviation of the actual wavelength λ from the intended wavelength λ_0 causes discrepancies in j^{TC} and j^{RC} , as the differential spectral responsivity of test and reference cell are generally wavelength-dependent. For correcting an erroneous wavelength deviation, j^{TC} and j^{RC} might be expanded with a Taylor series up to its first order around the intended center wavelength λ_0

$$j^{\mathrm{TC,RC}}(\lambda) \approx j^{\mathrm{TC,RC}}(\lambda_0) + \frac{\partial j^{\mathrm{TC,RC}}(\lambda)}{\partial \lambda}\Big|_{\lambda=\lambda_0} (\lambda - \lambda_0).$$
 (6.4)

Thus, the corrected current response is achieved from $j^{\text{TC,RC}}(\lambda_0) = f_{\lambda 0} j^{\text{TC,RC}}(\lambda)$ with the correction term

$$f_{\lambda 0}^{\mathrm{TC,RC}} = 1 - \frac{\partial j^{\mathrm{TC,RC}}(\lambda)}{\partial \lambda} \Big|_{\lambda = \lambda_0} \frac{(\lambda - \lambda_0)}{j^{\mathrm{TC,RC}}(\lambda)}.$$
 (6.5)

Apart from any known deviation in center wavelength that can be corrected with $f_{\lambda 0}$ (given that the deviations are not too strong), each measurement suffers from intrinsic uncertainties of the center wavelength $\Delta \lambda_0$. Typically two different contributions to $\Delta \lambda_0$ are considered that are, on the one hand, resulting from the (wavelength-dependent) repeatability of the applied components $\Delta \lambda_{01}$ and, on the other hand, from a wavelength-independent offset $\Delta \lambda_{02}$ [116].

The wavelength-dependence of $\Delta\lambda_{01}$ in the previously presented Laser-DSR setup is predominantly caused by the variation of the output sources (either THG or SHG are used directly or a prism or grating monochromator is applied, see Section 6.2.3). For THG and SHG $\Delta\lambda_{01}$ directly depends on the laser repeatability that is estimated to be approximately ± 1 nm (corresponding to plus-minus one wavelength increment).¹¹ Consequently, $\Delta\lambda_{01} \approx \pm 0.33$ nm for THG and $\Delta\lambda_{01} \approx \pm 0.5$ nm for SHG. The wavelength repeatability of the grating monochromator is specified with ± 0.04 nm for $\lambda \leq 1200$ nm and ± 0.08 nm for $\lambda > 1200$ nm. The prism monochromator repeatability is determined by the dispersion characteristics of the prism and the repeatability of the motorized translation axis. In the prism monochromator configuration presented in Section 6.2.3 $\Delta\lambda_{01}$ varies from approximately 0.01 nm at 520 nm to 0.13 nm at 1040 nm.

The wavelength-independent offset is essentially given by the uncertainty in wavelength calibration of the setup and estimated to be $\Delta\lambda_{02} \approx \pm 1$ nm in this work. In contrast to the repeatability uncertainty $\Delta\lambda_{01}$, the

¹¹This estimation is based on manufacturer's data.

offset uncertainty $\Delta \lambda_{02}$ accounts for both, test and reference cell measurement in an identical manner given that the measurement system remains unchanged in between the two measurements. In order to account for this correlation, the correction terms for test and reference cell (see Eq. (6.5)) are combined to

$$f_{\lambda 0} = \frac{f_{\lambda 0}^{\mathrm{TC}}}{f_{\lambda 0}^{\mathrm{RC}}} = \frac{1 - \frac{\partial j^{\mathrm{TC}}(\lambda)}{\partial \lambda} \Big|_{\lambda = \lambda_0} \frac{\Delta \lambda_0}{j^{\mathrm{TC}}(\lambda)}}{1 - \frac{\partial j^{\mathrm{RC}}(\lambda)}{\partial \lambda} \Big|_{\lambda = \lambda_0} \frac{\Delta \lambda_0}{j^{\mathrm{RC}}(\lambda)}}.$$
(6.6)

The spectrally resolved standard measurement uncertainties¹² $u_{\lambda 0}$ resulting from $\Delta \lambda_0 = \Delta \lambda_{01} (\lambda) + \Delta \lambda_{02}$ are exemplified in Table 6.1 in Section 6.4.3 assuming rectangular probability density functions (PDFs) for both $\Delta \lambda_{01}$ and $\Delta \lambda_{02}$. In Table 6.2 in Section 6.4.3 their relative contribution to the combined standard measurement uncertainty is given.

In addition to the uncertainty in center wavelength, spectral components other than the desired quasi-monochromatic radiation¹³ and incident onto the reference or test cell contribute to $Q^{\text{TC,RC}}$ via $j^{\text{TC,RC}} = j_{\text{mono}}^{\text{TC,RC}} + j_{\text{bl}}^{\text{TC,RC}}$, with "mono" denoting the desired quasi-monochromatic and "bl" the undesired current response contribution ("bl" symbolizes that these spectral components are supposed to be blocked). Expressing the total spectral irradiance $E_{\lambda}^{\text{tot}}(\lambda)$ as the sum of quasi-monochromatic and undesired radiation, $E_{\lambda}^{\text{tot}}(\lambda) = E_{\lambda}^{\text{mono}}(\lambda) + E_{\lambda}^{\text{bl}}(\lambda)$, yields the correction term

$$f_{\lambda \text{bl}}^{\text{TC,RC}} = 1 - \frac{\int \tilde{s}^{\text{TC,RC}}(\lambda) E_{\lambda}^{\text{bl}}(\lambda) d\lambda}{\int \tilde{s}^{\text{TC,RC}}(\lambda) E_{\lambda}^{\text{tot}}(\lambda) d\lambda}.$$
(6.7)

As monochromators are used over wide spectral ranges in the new Laser-DSR setup, thereby blocking the undesired radiation, Eq. (6.7) only needs to be considered for THG and SHG radiation. However, the dichroic mirrors and filters used in the THG and SHG beam path (see discussion in Section 6.2.3) eliminate the undesired wavelengths in such a manner, that

¹²Please note that general uncertainties are denoted by capital U and that *standard* measurement uncertainties are denoted by lowercase u throughout this work.

¹³These spectral components might e.g. result from remaining fundamental radiation after nonlinear conversion processes.

a correction using $f_{\lambda bl}^{TC,RC}$ is not reasonable. Instead, the still remaining undesired spectral contributions are taken into account as measurement uncertainties¹⁴

$$U_{\lambda \rm bl} = \frac{\sum_{i} \tilde{s}^{\rm TC, RC} \left(\lambda_{\rm bl, i}\right) E_{\lambda} \left(\lambda_{\rm bl, i}\right)}{\tilde{s}^{\rm TC, RC} \left(\lambda_{\rm tot}\right) E_{\lambda} \left(\lambda_{\rm tot}\right)} \approx \frac{\sum_{i} \tilde{s}^{\rm TC, RC} \left(\lambda_{\rm bl, i}\right) E_{\lambda} \left(\lambda_{\rm bl, i}\right)}{\tilde{s}^{\rm TC, RC} \left(\lambda_{\rm mono}\right) E_{\lambda} \left(\lambda_{\rm mono}\right)}.$$
 (6.8)

The insignificance of $U_{\lambda bl}$ becomes apparent in Table 6.1 in Section 6.4.3 where the standard measurement uncertainties $u_{\lambda bl} \approx 0$ in the entire spectral range.

Finally, a further wavelength-related measurement uncertainty arises from the bandwidth of the monochromatic radiation that deviates from the idealized case of true monochromaticity. If the second-order derivative of $\tilde{s}^{\rm TC,RC}$ after λ is non-vanishing (non-zero), the impact of bandwidth of the monochromatic radiation is comparable to a deviation in the center wavelength as discussed above.¹⁵ This is expressed as a second-order Taylor expansion with vanishing first order

$$j^{\mathrm{TC,RC}}(\lambda) \approx j^{\mathrm{TC,RC}}(\lambda_0) + \frac{\partial^2 j^{\mathrm{TC,RC}}(\lambda)}{\partial \lambda^2} \Big|_{\lambda = \lambda_0} \frac{(\lambda - \lambda_0)^2}{2}.$$
 (6.9)

Instead of applying a correction using Eq. (6.9), the impact of the radiation bandwidth $\lambda_{\rm bw}$ might be considered as a measurement uncertainty that is subject to an additional uncertainty arising from potential variations $\delta \lambda_{\rm bw}$ induced by the laser or the monochromators. Thus, the entire bandwidth-related uncertainty reads $\Delta \lambda_{\rm bw} = \lambda_{\rm bw} \pm \delta \lambda_{\rm bw}$.

Similar to the previously introduced uncertainty due to a wavelength

¹⁴In Eq. (6.8) a monochromatic approximation is applied that is reasonable when considering the quasi-monochromatic nature of the laser radiation in both desired and undesired spectral components. The sum over i represents that more than a single undesired quasi-monochromatic wavelength component might be present as e.g. in case of THG, when remaining SHG and fundamental wavelength components might exist simultaneously.

¹⁵This becomes clear when considering e.g. a linearly decaying differential spectral responsivity $\tilde{s}(\lambda)$ around a center wavelength λ_0 . As long as $\partial \tilde{s}(\lambda) / \partial \lambda$ is a constant (which is the case for a linear decay as in this example and is equivalent to a vanishing second order derivative), the measurement outcome does not change with spectral width $\lambda_{\rm bw}$ of the quasi-monochromatic radiation.

6.4 Measurement Uncertainty Analysis

offset $\Delta \lambda_{02}$, the impact of λ_{bw} acts identically on both the reference cell and the test cell measurement. Thus, it becomes significant if the secondorder derivatives of test and reference cell SR differ. This correlation is expressed by the correction term

$$f_{\lambda bw} = \frac{1 - \frac{\partial^2 j^{\text{TC}}(\lambda)}{\partial \lambda^2} \Big|_{\lambda = \lambda_0} \frac{\Delta \lambda_{bw}^2}{2j^{\text{TC}}(\lambda)}}{1 - \frac{\partial^2 j^{\text{RC}}(\lambda)}{\partial \lambda^2} \Big|_{\lambda = \lambda_0} \frac{\Delta \lambda_{bw}^2}{2j^{\text{RC}}(\lambda)}}.$$
(6.10)

In Table 6.1 in Section 6.4.3 the standard measurement uncertainties $u_{\lambda bw}$ assigned to the spectral bandwidths are exemplified for the combination of a GaAs test and a Si reference cell and the spectral bandwidths of the presented setup. In a rather conservative estimation the uncertainty of the setup bandwidths is assumed to be $\delta \lambda_{bw} = \pm 2$ nm. In spectral ranges using the prism monochromator a rectangular PDF is assumed for $\delta \lambda_{bw}$ resulting from a convolution of the rectangular slit function with the much smaller Gaussian beam width in the slit plane. A triangular PDF is assumed for the grating monochromator that is obtained from a convolution of entrance and exit slit and, finally, a normal PDF is assumed for THG and SHG radiation that is not passing any monochromator.

6.4.1.2 Spatial Non-Uniformity

Although a high degree of uniformity in the monochromatic spatial irradiance distribution E(x, y) is achieved (compare Figs. 6.6 and 6.7), the remaining non-uniformity necessitates a correction of the measurement outcome, thereby introducing measurement uncertainties that are discussed in this section.

Firstly, the spatial non-uniformity of the monochromatic radiation field might result in a variation of the spatially averaged irradiance

$$\bar{E}^{\mathrm{TC,RC}} = \frac{1}{A^{\mathrm{TC,RC}}} \int_{A^{\mathrm{TC,RC}}} E(x,y) \,\mathrm{d}A,\tag{6.11}$$

if reference and test cell area $A^{\text{TC,RC}}$ are different. Given that E(x,y) is

known and the center positions of test and reference cell are identical, the resulting deviation in \bar{E}^{TC} with respect to \bar{E}^{RC} can be corrected for by¹⁶

$$f_{\rm Size} = \frac{\bar{E}^{\rm RC}}{\bar{E}^{\rm TC}}.$$
(6.12)

In order to account for deviating center positions of test and reference cell, Eq. (6.12) can be expanded into a Taylor series

$$\frac{\bar{E}^{\mathrm{RC}}}{\bar{E}^{\mathrm{TC}}(x)} \approx \frac{\bar{E}^{\mathrm{RC}}}{\bar{E}^{\mathrm{TC}}(x_0)} + \bar{E}^{\mathrm{RC}} \frac{\partial \left(\bar{E}^{\mathrm{TC}}(x)\right)^{-1}}{\partial x}\Big|_{x=x_0} (x-x_0) \qquad (6.13)$$

yielding the position correction term

$$f_{\rm Pos} = 1 - \bar{E}^{\rm TC}(x) \frac{\partial \left(\bar{E}^{\rm TC}(x)\right)^{-1}}{\partial x} \Big|_{x=x_0} (x - x_0),$$
 (6.14)

with reference cell center position x_0 and test cell center position x. For convenience, only the x-direction is considered in Eq. (6.14). Naturally, the same equations hold for y and y_0 . Please note that only the difference of reference and test cell center position ($\Delta x = x - x_0$) is relevant in this consideration and that the absolute center position of the reference cell does not contribute. Combining f_{Pos} and f_{Size} gives

$$f_{\text{PosSize}} = f_{\text{Pos}} f_{\text{Size}} = \frac{\bar{E}^{\text{RC}}}{\bar{E}^{\text{TC}}} \left(1 - \bar{E}^{\text{TC}} \left(x \right) \frac{\partial \left(\bar{E}^{\text{TC}} \left(x \right) \right)^{-1}}{\partial x} \Big|_{x=x_0} \left(x - x_0 \right) \right).$$

$$(6.15)$$

For achieving an estimate for the impact of test cell size and position the first-order Taylor term of Eq. (6.15) has been simulated for various cell sizes in both measurement areas.¹⁷ Evaluating the maximum deviation

¹⁶It is noteworthy that an uncertainty in f_{Size} arises from an uncertainty in area measurement of the test and reference solar cells. However, as this area uncertainty ranges from 0.05% to 0.3%, depending on the cell size, and $f_{\text{Size}} < 2\%$ owing to the high uniformity of the field, the assigned uncertainty of f_{Size} is well below 0.01% and can be readily neglected.

¹⁷The script for this simulation has been kindly provided by Ino Geisemeyer, Fraunhofer ISE.



Figure 6.10: Maximum deviation in spatially average irradiance after Eq. (6.11) over position $(x - x_0)$ and test cell area (shown next to the respective curves). Blue denotes results for large area (see Fig. 6.6), black for small area setup (see Fig. 6.7).

of average irradiance from a re-positioning of the test cell in eight directions surrounding x_0 (in spatial coordinates: (-x, 0), (-x, x), (0, x), ...)yields the dependencies of test cell size, center position $(x - x_0)$ and measurement area as shown in Fig. 6.10. Assuming a 2x2 cm² test cell and a rectangular PDF with $\Delta x = 2$ mm, a standard measurement uncertainty of $u_{\text{PosSize}} \approx 0.11\%$ is obtained for the small measurement area.

Apart from the impact of cell size and position on the measurement outcome in presence of irradiance non-uniformity, also a non-uniformity of the test cell itself might affect the measurement. As the non-uniformity characteristics of the test cells are typically unknown, any correction of this impact is infeasible. Instead, a worst case consideration is conducted to achieve an upper bound estimate for the standard measurement uncertainty assigned to cell non-uniformity. For this, the short circuit currents of a highly uniform and non-uniform¹⁸ test cell with identical spatially averaged EQEs are compared for the spatial irradiance distributions of the Laser-DSR setup (see Section 6.2.4). Even in this most extreme case the standard measurement uncertainty $u_{CellInh} < 0.05\%$. In the final

 $^{^{18}{\}rm The}$ non-uniform test cell has been modeled to suffer from a halving of its EQE at one half of its entire area.

model equation cell non-uniformity is considered by the correction term $f_{\text{CellInh}} = 1 - u_{\text{CellInh}}.$

As a concluding remark to this subsection, it has to be mentioned that the presented correction terms and uncertainties are expected to be independent on wavelength owing to the imaging concept presented in Section 6.2.4 (see e.g. Fig. 6.7).

6.4.1.3 Longitudinal Test Object Position

The last correction term introduced in this section arises from the variation of spatially averaged irradiance $\bar{E}(z)$ in longitudinal z-direction (thus, along the beam path). Similar to the discussions above, the correction term f_{dz} is obtained from a first-order Taylor series of $\bar{E}(z)$ around $z = z_0$ yielding

$$\bar{E}(z) \approx \bar{E}(z_0) + \frac{\partial \bar{E}(z)}{\partial z}\Big|_{z=z_0} (z-z_0)$$
(6.16)

and resulting in

$$f_{\rm dz} = 1 - \frac{\partial E\left(z\right)}{\partial z}\Big|_{z=z_0} \frac{\left(z-z_0\right)}{\bar{E}\left(z\right)}.$$
(6.17)

It is noteworthy that only the relative deviation of reference to test cell zposition is of relevance when correcting the measurement with Eq. (6.17).

Again, evaluating the first-order Taylor term yields an estimation of standard uncertainty u_{dz} . The z-dependence of $\bar{E}(z)$ has been simulated with ray tracing software for both the small and the large measurement area. Within an height deviation of ± 20 mm, that is at least one order of magnitude larger than the entire relevant uncertainty range (arising from e.g. uncertainties in adjustment, cell thickness and installation height), the variation in $\bar{E}(z)$ is found to be smaller than the standard uncertainty of the simulated mean values. Thus, no height dependence could have been retrieved from the simulations, pointing out that the height dependence is certainly less than 0.01 %/mm (which is the estimated detection threshold of the applied simulation). Thus, attributing a rectangular PDF to $\Delta z = z - z_0 = 2$ mm results in a standard measurement uncertainty of $u_{dz} < 0.012\%$.

6.4.2 Further Measurement Uncertainties

The previously discussed correction terms and uncertainties are directly related to the specific properties of the new Laser-DSR measurement setup that has been developed in the course of this work. In contrast to those, this section covers correction terms and measurement uncertainties that are not related to the optical properties of the new setup. Thus, from the point of view of developing a DSR-measurement facility with improved optical properties, the subsequently discussed uncertainty contributions are of intrinsic nature.

Firstly, reference and test cell current responses $j^{\text{TC,RC}}$ are dependent on the temperature \mathcal{T} of the pn-junction. In analogy to the above derivations, this dependency can be expressed as a Taylor series given by

$$j^{\mathrm{TC,RC}}\left(\mathcal{T}\right) \approx j^{\mathrm{TC,RC}}\left(\mathcal{T}_{0}\right) + \frac{\partial j^{\mathrm{TC,RC}}\left(\mathcal{T}\right)}{\partial \mathcal{T}}\Big|_{\mathcal{T}=\mathcal{T}_{0}}\left(\mathcal{T}-\mathcal{T}_{0}\right)$$
(6.18)

yielding the correction term

$$f_{\text{temp}}^{\text{TC,RC}} = 1 - \frac{\partial j^{\text{TC,RC}}\left(\mathcal{T}\right)}{\partial \mathcal{T}} \Big|_{\mathcal{T}=\mathcal{T}_0} \frac{\left(\mathcal{T}-\mathcal{T}_0\right)}{j^{\text{TC,RC}}\left(\mathcal{T}\right)}.$$
(6.19)

The uncertainty in test cell temperature is assumed to be $\Delta T = 0.5$ K, whereas the housing of the reference cells typically enables a better control of the temperature estimated as $\Delta T = 0.2$ K. Especially due to the temperature dependent band edge shift in semiconductors, the standard measurement uncertainty u_{temp} is strongly wavelength-dependent. In Table 6.1 in Section 6.4.3 u_{temp} is exemplified in a spectrally resolved manner. In Table 6.2 its relative contribution to the combined measurement uncertainty is given.

A further correction term is related to the phase of the lock-in amplifiers. Taking care that the phase difference of chopped monochromatic light and lock-in amplifier is zero ($\alpha = 0$), it suffices to read out the Xvalue, thus, the real part, of the lock-in signal for reference and test cell. Small range deviations from this phase-condition can be treated by the Taylor series expansion around $\alpha_0 = 0$ yielding $\cos(\alpha) \approx 1 - \alpha^2/2$ and the correction term

$$f_{\text{phase}}^{\text{TC,RC}} = 1 + \frac{\alpha^2}{2\cos\left(\alpha\right)}.$$
 (6.20)

Assuming a rectangular PDF of $\Delta \alpha = 1^{\circ}$ results in an assigned standard measurement uncertainty of

$$u_{\rm phase} = \sqrt{\left(u_{\rm phase}^{TC}\right)^2 + \left(u_{\rm phase}^{RC}\right)^2} \approx 0.012\%, \tag{6.21}$$

taking into account both reference and test cell.

Furthermore, the lock-in amplifier itself and the transimpedance amplifier contribute to the overall uncertainty by their non-linearity and temperature-dependence. From the uncertainty analysis of other setups using identical amplifiers, the assigned standard measurement uncertainty is given by $u_{\rm amps} = 0.1\%$. If reference and test cell differ significantly in their size, different feedback resistors are used yielding an additional uncertainty contribution of $u_{\rm res} = 0.09\%$, as known from other setups. The related correction term is given by $f_{\rm el} = 1 - u_{\rm el}$, with $u_{\rm el} = (u_{\rm amps}^2 + u_{\rm res}^2)^{1/2}$.

Finally, the uncertainty in the reference cell's differential spectral responsivity \tilde{s}^{RC} itself (u_{RC}) as well as the standard deviations of the measured mean values of Q^{TC} and Q^{RC} , u_{Q}^{TC} and u_{Q}^{RC} , contribute to the overall measurement uncertainty (compare Eq. (6.3)).

Apart from the uncertainties introduced in Sections 6.4.1 and 6.4.2, each measurement might suffer from additional uncertainties: e.g. nonlinearity of the test cell, interreflection of radiation, spatial and spectral properties of the bias irradiation, stability of monitor diode.... Although it is important to be aware of these uncertainties, they are negligible if their origin is tackled properly and, hence, not addressed further.

6.4.3 Combined measurement uncertainty¹⁹

Introducing the previously discussed correction terms and measurement uncertainties into Eq. (6.3) yields

$$\tilde{s}^{TC}(\lambda) = \tilde{s}^{RC}(\lambda) \frac{Q^{TC}(\lambda)}{Q^{RC}(\lambda)} + f_{\lambda 0} \frac{f_{\lambda bl}^{TC}}{f_{\lambda bl}^{RC}} f_{\lambda bw} f_{PosSize} f_{CellInh} f_{dz} \frac{f_{temp}^{TC}}{f_{temp}^{RC}} \frac{f_{phase}^{TC}}{f_{phase}^{RC}} f_{el}.$$
(6.22)

Applying the new Laser-DSR setup for measuring Q^{TC} and Q^{RC} of a linear GaAs test and Si reference solar cell (both of approximately $2x2 \text{ cm}^2$ cell area; with linearity: $\tilde{s}(\lambda) = s(\lambda)$) yields the results shown in Fig. 6.11a when using Eq. (6.22). The open black symbols in Fig. 6.11a are measured data points of the spectral responsivity (SR) of the GaAs test cell. The red line, denoting a SR-measurement of the same cell with another setup that is typically applied for solar cell calibration, demonstrates the excellent agreement of the Laser-DSR result and the independent reference measurement.

The dashed red line in Fig. 6.11a gives the expanded uncertainty of the applied Si reference cell. The dashed black line indicates the expanded uncertainty $U_{\text{tot}}^{\text{MC}}$ of the Laser-DSR measurement with a *coverage probability* of 95.45% (95.45% coverage probability corresponds to twice the standard deviation of a normal distribution) achieved from Monte Carlo (MC) simulations after [118] discussed in the Appendix E. Additionally, the thinner solid black line illustrates the equivalent expanded uncertainty after GUM $U_{\text{tot}}^{\text{GUM}}$ [117], not taking into account correlations in $\Delta\lambda_0$ and $\Delta\lambda_{\text{bw}}$ (discussed in Section 6.4.1.1). Computing the effective degree of

¹⁹Being frequently used in GUM ("Guide to the expression of uncertainty in measurement") framework [117] for expressing that a combination of various input uncertainties is considered, the term *combined* is redundant for Monte Carlo simulations [118, p. 7, 4.10]. However, the term is used for both, GUM and Monte Carlo, in this work to emphasize the currently discussed uncertainty aiming at a better legibility.



Figure 6.11: (a) Spectral responsivity (SR) of a GaAs solar cell measured with the new Laser-DSR setup (black symbols). The red solid line, denoted by reference measurement, indicates an independent SRmeasurement of the same solar cell with another SR-measurement setup. The dashed lines indicate the expanded measurement uncertainty of the Laser-DSR measurement (black) and of the reference solar cell (red) used for this measurement. The thinner solid black line gives the Laser-DSR measurement uncertainty after GUM. (b) Normalized SRs of reference cell (Si) and test cell (GaAs) used for the measurement presented in (a).

freedom $\nu_{\rm f, eff}$ from the Welch-Satterthwaite formula [117, p. 73, G.4.1]

$$\nu_{\rm f,eff} = \frac{\left(\sqrt{\sum_{i=1}^{N} u_i^2}\right)^4}{\sum_{i=1}^{N} \frac{u_i^4}{\nu_{\rm f,i}}} \gg 50, \tag{6.23}$$

taking into account the degrees of freedom $\nu_{\rm f,i}$ of the Type A uncertainties as given in Table 6.1, demonstrates that assuming a normal distribution for the combined measurement uncertainty is reasonable. To achieve the same coverage probability as in the Monte Carlo simulations, a coverage factor of $k_{\rm cov} = 2$ is chosen to compute $U_{\rm tot}^{\rm GUM} = k_{\rm cov} u_{\rm tot}^{\rm GUM}$.²⁰ The

 $^{^{20}}$ It is noteworthy that, in presence of nonlinear uncertainty contributions, the validity of the GUM framework is constricted if their respective PDFs are not normally

expanded uncertainties after GUM and MC are very similar apart from the short wavelength region, where a slight reduction in MC uncertainty results from the correlation in wavelength offset $\Delta \lambda_{02}$.

In Table 6.1 Type B (those, that were discussed in Sections 6.4.1 and 6.4.2) and Type A standard measurement uncertainties (standard deviations in measurement $u_{\rm Q}^{\rm TC}$ and $u_{\rm Q}^{\rm RC}$) are given for several characteristic wavelengths of the measurement result shown in Fig. 6.11a. In addition, the expanded uncertainties obtained from Monte Carlo and GUM, $U_{\rm tot}^{\rm MC}$ and $U_{\rm tot}^{\rm GUM}$, are given.²¹ In Table 6.2 the relative contributions of the individual uncertainties to the expanded measurement uncertainty are given. The color coding serves as a guide to the eye for identifying the most relevant uncertainty contributions. The relative contributions shown in Table 6.2 refer to the expanded uncertainty after Monte Carlo $U_{\rm tot}^{\rm MC}$, given in Table 6.1, that conveniently enables extraction of standard uncertainties from more complex PDFs consisting of various individual components as e.g. in $u_{\lambda 0}$ (see Section 6.4.1.1).

The results shown in Table 6.2 clearly demonstrate that in the spectral range from 500 to 800 nm the most significant contribution to the expanded uncertainty of the Laser-DSR measurement arises from the uncertainty of the reference cell itself $(u_{\rm RC})$. This conclusion is also apparent from the close vicinity of the dashed lines in Fig. 6.11a highlighting the comparably small additional uncertainty induced by the setup itself in the respective spectral range. Moreover, the dashed lines demonstrate that the reference cell remains to be the major uncertainty component up to approximately 860 nm. Apart from the reference cell uncertainty, the positioning of the cell $(u_{\rm PosSize}$, cells of same area have been compared here) and the electrical peripheral equipment $(u_{\rm el})$ are main contributors to the combined uncertainty. Whereas the latter contribution might be regarded

distributed [118, p. 13, 5.8.1 d)], [117, p. 19, 5.1.2 note]. Above 520 nm this is not given for $\Delta \lambda_{\rm bw}$ (see Section 6.4.1.1). However, comparing GUM to Monte Carlo (see Fig. 6.11a), that is not subject to this restriction, demonstrates applicability of GUM for the case considered here.

²¹Please note that $u_{\text{tot}}^{\text{GUM}}$ in Table 6.1 cannot be directly computed from the individual standard measurement uncertainties u_i , as these relate to the Monte Carlo simulation outcome and take correlations into account that are not being considered in this GUM representation.

Uncer- tainty	Type	Wavelength/nm									
		300	340	400	500	600	800	880	900	920	
$\overline{u_{\lambda 0}}$	В	2.75	2.40	0.35	0.14	0.01	0.01	0.97	6.73	7.48	
$u_{\lambda \mathrm{bl}}^{\mathrm{TC}}$	В					0.00					
$u_{\lambda \mathrm{bl}}^{\mathrm{RC}}$	В					0.00					
$u_{\lambda \mathrm{bw}}$	В	1.07	0.75	0.08	0.01	0.01	0.00	3.30	11.89	34.45	
$u_{\rm CellInh}^{\rm TC}$	В					0.01					
$u_{\rm CellInh}^{\rm RC}$	В					0.00					
u_{PosSize}	В					0.11					
$u_{\rm dz}$	В					0.01					
$u_{\rm temp}^{\rm TC}$	В	0.43	0.04	0.00	0.01	0.01	0.02	0.16	0.66	1.27	
$u_{\rm temp}^{\rm RC}$	В	0.01	0.01	0.00	0.00	0.01	0.00	0.01	0.01	0.02	
$u_{\rm el}$	В					0.10					
$u_{\rm phase}$	В					0.01					
$u_{ m RC}$	В	0.32	0.30	0.28	0.26	0.28	0.25	0.27	0.27	0.27	
$\overline{u_{\mathrm{Q}}^{\mathrm{TC}}}$	А	0.45	0.11	0.06	0.01	0.01	0.02	0.02	0.05	0.15	
$ u_{\mathrm{f},\mathrm{Q}}^{\mathrm{TC}}$						99					
$u_{\mathrm{Q}}^{\mathrm{RC}}$	А	0.04	0.08	0.01	0.02	0.02	0.01	0.01	0.02	0.03	
$ u_{ m f,Q}^{ m RC}$						99					
$u_{ m tot}^{ m GUM}$	M	4.19	3.39	0.98	0.53	0.35	0.32	3.60	13.84	36.34	
$U_{\rm tot}^{\rm GUM}$	GĽ	8.37	6.78	1.97	1.06	0.71	0.64	7.19	27.68	72.68	
$u_{ m tot}^{ m MC}$	U	3.03	2.54	0.48	0.33	0.32	0.29	3.45	13.72	35.39	
$U_{ m tot}^{ m MC}$	Μ	5.72	4.68	0.96	0.66	0.63	0.58	6.90	27.30	70.88	

Standard Measurement Uncertainty/%

Table 6.1: Spectrally resolved standard measurement uncertainties u_i of the new Laser-DSR setup obtained from Monte Carlo simulations; $u_{\text{tot}}^{\text{GUM}}$ denotes the total standard uncertainty after GUM. The expanded uncertainties $U_{\text{tot}}^{\text{MC}}$ and $U_{\text{tot}}^{\text{GUM}}$ are given for a coverage probability of 95.45%. The uncertainties refer to the given example of measuring a GaAs test cell against a Si reference cell.

Uncer- tainty	Wavelength/nm									
	300	340	400	500	600	800	880	900	920	
$U_{\lambda 0}$	82.08	89.10	52.63	17.46	0.03	0.04	7.85	24.06	4.46	
$U_{\lambda \mathrm{bl}}^{\mathrm{TC}}$					0.00					
$U_{\lambda \mathrm{bl}}^{\mathrm{RC}}$					0.00					
$U_{\lambda \rm bw}$	12.43	8.78	2.60	0.13	0.04	0.00	91.18	75.19	94.76	
$U_{\rm CellInh}^{\rm TC}$	0.00	0.00	0.04	0.09	0.10	0.12	0.00	0.00	0.00	
$U_{\rm CellInh}^{\rm RC}$					0.00					
$U_{\rm PosSize}$	0.13	0.18	5.12	10.71	11.85	13.83	0.10	0.01	0.00	
$U_{\rm dz}$	0.00	0.00	0.06	0.12	0.13	0.15	0.00	0.00	0.00	
$U_{\rm temp}^{\rm TC}$	2.05	0.03	0.00	0.04	0.14	0.37	0.23	0.23	0.13	
$U_{\rm temp}^{\rm RC}$	0.00	0.00	0.00	0.01	0.08	0.01	0.00	0.00	0.00	
$U_{\rm el}$	0.11	0.15	4.30	8.99	9.95	11.62	0.08	0.01	0.00	
$U_{\rm phase}$	0.00	0.00	0.07	0.14	0.15	0.18	0.00	0.00	0.00	
$U_{\rm RC}$	1.10	1.42	33.98	61.72	76.91	72.60	0.62	0.04	0.01	
$\overline{U_{\mathrm{Q}}^{\mathrm{TC}}}$	2.20	0.19	1.50	0.16	0.20	0.72	0.00	0.00	0.00	
$U_{ m Q}^{ m RC}$	0.02	0.10	0.04	0.45	0.28	0.22	0.00	0.00	0.00	
$\sum U_i$	100	100	100	100	100	100	100	100	100	

Relative Contributions to Combined Uncertainty/%

Table 6.2: Spectrally resolved relative contributions of the individual uncertainties to the combined measurement uncertainty $U_{\text{tot}}^{\text{MC}}$ as given in Table 6.1. The color coding is a guide to the eye assisting in identifying most relevant uncertainties in dependence of the spectral range (from green for low to red for high relative contribution).

as given on first sight, the positioning uncertainty could be tackled by an improved uniformity of the radiation field or a tighter control of the cell position (e.g. by developing a camera-based positioning approach).

For wavelengths shorter than approximately 520 nm the combined measurement uncertainty of the Laser-DSR setup rises, which is mainly due to the increased impact of center wavelength uncertainty $(u_{\lambda 0})$. This increase in $u_{\lambda 0}$ is caused by several reasons: firstly, the wavelength repeatability is reduced when no monochromators are applied (below 520 nm SHG and THG radiation is directly applied for the measurements, see Section 6.4.1.1); secondly, the slope, thus, wavelength sensitivity of the GaAs test cell increases (see Fig. 6.11b); and, thirdly, the slope of reference and test cell become different for lower wavelengths (see also in Fig. 6.11b). The first and second issue are simply related to an enhancement of $\Delta \lambda_{01}$ and $|\partial j^{\rm TC}/\partial \lambda|$ in e.g. Eq. (6.5).²² The third issue, in contrast, is related to the correlative nature of the wavelength-independent offset $\Delta \lambda_{02}$ and occurs only in case of different test and reference cell SR slopes (see Eq. (6.6)). Thus, applying a reference cell that spectrally matches the test cell, the impact of $\Delta \lambda_{02}$ is eliminated, whereas the uncertainties related to the slope of the test cell $(|\partial j^{\rm TC}/\partial \lambda|)$ and the wavelength repeatability $(\Delta \lambda_{01})$ remain (see Fig. 6.12). Although this demonstrates the impact of reference and test cell combination, the issue of choosing a specific reference cell is typically not addressed in SR-measurements. This results from the virtually negligible contribution to the overall short circuit current of a test cell from spectral ranges where the similarity of reference and test cell becomes relevant (see discussion in Section 6.4.4).

In addition to the center wavelength uncertainty the bandwidth-related uncertainty $u_{\lambda bw}$ becomes larger and more significant for shorter wavelength (see Tables 6.1 and 6.2). Similar to the correlation in wavelength offset $\Delta \lambda_{02}$, $u_{\lambda bw}$ is caused by the differences in test and reference cell SR, however, by the differences in their second derivatives after λ (see Eq. (6.10)). As the bandwidth-related uncertainty is way more important at the test cell's band edge, a detailed discussion of $u_{\lambda bw}$ follows in the respective paragraph below.

Concluding the discussion for short wavelengths, it can be stated that the center wavelength uncertainty $u_{\lambda 0}$ is the main contributor to the combined measurement uncertainty. For reducing $u_{\lambda 0}$ a closer look at the conservative assumption of ± 1 nm wavelength repeatability of the laser

²²In fact, the expanded uncertainty shown as black dashed line in Fig. 6.11a resembles the shape of the first derivative of the test cell's SR $s^{\text{TC}}(\lambda)$.



Figure 6.12: Selected standard uncertainties for measuring a GaAs test cell with a (a) Si or (b) GaAs reference cell with identical uncertainty $u_{\rm RC}$. If reference and test cell have identical SRs as assumed in case (b), the (partially) correlated center wavelength $u_{\lambda 0}$ and bandwidth uncertainty $u_{\lambda bw}$ are significantly reduced.

system itself might be worthwhile. Verifying a significantly better wavelength repeatability by external means would directly result in a substantial reduction of the combined measurement uncertainty in this spectral region.

Turning the discussion towards longer wavelengths a dramatic increase in the expanded uncertainty is observed in Fig. 6.11a for wavelengths longer than 860 nm. From Table 6.2 this increase is attributed to the dominating impact of bandwidth-related uncertainty $u_{\lambda bw}$ and center wavelength uncertainty $u_{\lambda 0}$. The reasoning for an enhanced contribution of $u_{\lambda 0}$ follows very closely the previous discussion for the short wavelength spectral region. Although the prism monochromator is applied in this spectral range, featuring a more than ten times better wavelength repeatability as assumed in the short wavelength range above, $u_{\lambda 0}$ contributes significantly due to the increased slope of test cell SR ($|\partial j^{TC}/\partial \lambda|$ in Eq. (6.5)). The rising impact of $u_{\lambda bw}$ at the test cell's band edge results from the substantial increase in the second-order derivative of the test cell's spectral responsivity ($|\partial^2 j^{TC}/\partial \lambda^2|$ in Eq. (6.10)). Assuming a reference cell of identical SR illustrates the correlative nature of this uncertainty contribution, as $u_{\lambda bw}$ vanishes (see Fig. 6.12).

Sticking to the idealized assumption of a perfectly adapted reference cell further demonstrates the necessity of controlling test and reference cell temperature. In fact, the remaining increase in u_{tot} up to 920 nm is caused by $u_{\text{temp}}^{\text{TC}}$ and $u_{\text{temp}}^{\text{RC}}$ (not shown in Fig. 6.12b). Above 920 nm the spectral responsivity approaches zero (see Fig. 6.11b) and the standard deviations of the measurements (u_{Q}^{TC} and u_{Q}^{RC}) become the dominating uncertainty contributions.

At least as important and interesting as the dominating uncertainty contributions are those that do not contribute to the combined measurement uncertainty. Firstly, there is virtually no height dependency of the irradiation level in the new Laser-DSR setup (see discussion in Section 6.4.1.3), thus, the assigned standard measurement uncertainty $u_{dz} < 0.02\%$ in the entire spectral range. Even more striking is the elimination of undesired spectral components by using spectral filters or a grating or prism monochromator (see Section 6.4.1.1). Consequently, the assigned standard measurement uncertainty $u_{\lambda bl} \approx 0$. Finally, the uncertainty related to a potential spatial non-uniformity of the test cell's EQE $u_{CellInh}$ is virtually zero even for the most extreme consideration discussed in Section 6.4.1.2.

6.4.4 Uncertainty in Short Circuit Current

The Monte Carlo method used for the spectral uncertainty analysis above (see Section 6.4.3) can also be applied for determining the standard uncertainty of a short circuit current ($I_{\rm SC}$) measurement using the new Laser-DSR setup.

As introduced in Section 2.2 the I_{SC} of a solar cell under a certain spectral irradiance distribution $E_{\lambda}(\lambda)$ is given by

$$I_{\rm SC} = \int s(\lambda) E_{\lambda}(\lambda) d\lambda, \qquad (6.24)$$

with absolute spectral responsivity $s(\lambda)$. Setting $E_{\lambda}(\lambda)$ equal to the



Figure 6.13: Histogram of the deviation $\Delta I_{\rm SC}$ between Monte Carlo simulated and undisturbed short circuit current $I_{\rm SC}$ of a GaAs test cell measured with a (a) Si or (b) GaAs reference cell of identical uncertainty $u_{\rm RC}$. The improved spectral match in case (b) results in a minor reduction of the standard measurement uncertainty of $I_{\rm SC}$ from $u_{\rm Isc} \approx 0.33\%$ to $u_{\rm Isc} \approx 0.32\%$.

AM1.5g (λ) reference solar spectral irradiance distribution [23], the $I_{\rm SC}$ under standard test conditions ($I_{\rm SC}^{\rm STC}$) is achieved.

As detailed in the Appendix E a random draw m for the input uncertainties discussed above is used to compute s_m^{TC} for all wavelengths λ by Eq. (6.22). This disturbed $s_m^{\text{TC}}(\lambda)$ is applied to Eq. (6.24) and the resulting short circuit current $I_{\text{SC}}^{\text{TC},m}$ is computed. Comparing $I_{\text{SC}}^{\text{TC},m}$ to the undisturbed (all uncertainties set to zero) I_{SC} yields their relative deviation ΔI_{SC}^m for this specific draw of random input variables. Repeating this procedure 100,000 times for the input uncertainty distributions discussed in Sections 6.4.1 to 6.4.3 gives the PDF of ΔI_{SC} illustrated as histogram in Fig. 6.13a.²³ The standard deviation, thus, standard uncertainty in the I_{SC} measurement of a GaAs cell (using a Si reference cell) is determined to be $u_{\text{Isc}} \approx 0.33\%$.

It was demonstrated in the previous discussion of the spectrally resolved uncertainties (see Section 6.4.3), that application of a spectrally

 $^{^{23}}$ In other words, the Monte Carlo method propagates the input uncertainty distributions through the model and yields an uncertainty distribution of the output variable.

well matching reference cell for SR-measurements substantially reduces uncertainties in some spectral ranges. Therefore, a reference cell of identical spectral features as the GaAs test cell has been assumed yielding a minor improvement of standard uncertainty of short circuit current to $u_{\rm Isc} \approx 0.32\%$ (see Fig. 6.13b). This result clearly confirms the previous statement that the increasing uncertainty for short and long wavelengths is essentially negligible regarding the uncertainty in short circuit current. In fact, the main contribution to $u_{\rm Isc}$ arises from the uncertainties relevant between 400 and 860 nm as approximately 91% of the entire cell's current are generated in that spectral range (from 500 to 860 nm still 78% of the total current are generated). As depicted in Figs. 6.11a and 6.12 and highlighted in Table 6.2 the uncertainty of the reference cell $u_{\rm RC}$ is the main contributor to the combined uncertainty in that spectral range. Thus, the most substantial reduction of $u_{\rm Isc}$ is expected from a reduction of $u_{\rm RC}$.

6.5 Conclusions & Comparison

In the course of this work a new DSR-measurement system based on ultrashort laser pulses has been developed aiming at a significant reduction of measurement uncertainties. In this chapter the new facility was presented and a detailed analysis of its measurement uncertainty was conducted. In this conclusion the main results of this chapter are briefly recapitulated and a short comparison of the new facility to other setups, mainly focusing on measurement uncertainties, is conducted.

6.5.1 General Presentation of the New Setup

Starting from a description of the general concept of the new setup, the radiation sources applied in the tunable ultrashort pulse laser system were introduced and their main properties were discussed (see Section 6.2.1). In this discussion the necessity for spectrally filtering or shaping the ultrashort laser pulses was revealed from the radiation bandwidths and potentially present undesired spectral components. In Section 6.2.3 three different approaches tackling this were introduced: bandpass filters, a

grating monochromator and a prism monochromator, that was specifically developed for the Laser-DSR facility in the course of a bachelor's thesis under supervision of the author of this work [114].

After a brief recapitulation of the necessity of temporal shaping (detailed in Chapter 4) and the developed approach for tackling this issue (detailed in Chapter 5), the optical concept of the new Laser-DSR measurement facility was presented in Section 6.2.4. It was demonstrated by ray-tracing simulations that the optical concept of the new setup enables monochromatic illumination of a 16x16 cm² area with a non-uniformity of less than 3%. In addition, a second optical setup was presented that, by taking advantage of the same optical concept, achieves uniform illumination of a 4x4 cm² area with a non-uniformity of approximately 1%, thereby enhancing irradiance (and measurement signal) by a factor of 13 as compared to the first, larger area setup. First experimental results confirmed the ray-tracing simulations of the smaller area regarding uniformity and indicated the wavelength-independence of the optical concept.

The presentation of the new measurement setup was concluded by a discussion of the optical losses induced by the necessary temporal, spatial and spectral shaping of the radiation and by comparing the available optical power in the measurement plane to setups using conventional light sources (see Section 6.3). It was demonstrated that the new setup provides 10 to 500 times higher monochromatic optical power as a state-of-the-art DSR-setup based on optical filters and even about 100 to 10,000 times higher optical power as a typical grating monochromator DSR-setup.

6.5.2 Measurement Uncertainty of the New Setup

After completion of the setup's presentation, its measurement uncertainties were discussed in detail in Section 6.4. Firstly, the various measurement uncertainties arising from the setup itself were discussed in Section 6.4.1, starting from a discussion of wavelength-related uncertainties (see Section 6.4.1.1), followed by spatial uncertainties (see Section 6.4.1.2) and concluded by the height dependency of the setup (see Section 6.4.1.3). It was shown that the quasi-monchromatic nature of the laser radiation and the applied spectral shaping concepts virtually eliminate any undesired spectral components from the laser beam. Consequently, uncertainties related to those spectral components vanish. Furthermore, the uncertainty related to the height of the test object (longitudinal distance between reference and test object) approaches zero owing to the optical concept presented in Section 6.2.4. After these setup-related uncertainties, further uncertainties were introduced in Section 6.4.2 in order to enable a full modeling of the entire measurement uncertainty of the new setup.

The full uncertainty model was applied in Section 6.4.3 to compute and simulate²⁴ spectrally resolved standard and expanded measurement uncertainties when measuring a $2x2 \text{ cm}^2$ GaAs test cell with a Si reference cell. A detailed discussion of the occurring uncertainties and their relative contributions to the overall uncertainty revealed the major potential for a further reduction of the overall uncertainty: e.g. by reduced reference cell uncertainty and improved lateral test cell positioning. The impact of the reference cell uncertainty was further emphasized by simulating the standard measurement uncertainty of the new setup regarding short circuit current (I_{SC}) measurements. In Section 6.4.4 this uncertainty was determined to be $u_{\rm Isc} \approx 0.33\%$ for a 2x2 cm², being equivalent to an expanded uncertainty in $I_{\rm SC}$ of approximately 0.66%. Although larger cells were not measured yet, the detailed uncertainty analysis in Section 6.4 allows to conclude that only $u_{\rm el}$, $u_{\rm PosSize}$ and $u_{\rm CellInh}$ are sensitive to the test cell size. Taking this into account the standard uncertainty in measuring the $I_{\rm SC}$ of a 15.6x15.6 cm² test cell is estimated to be $u_{\rm Isc} \approx 0.35\%$ resulting in an expanded uncertainty of $U_{\rm Isc} \approx 0.70\%$. Thus, the expanded measurement uncertainty of the reference cell itself, given by $U_{\rm BC} \approx 0.55\%$, is the main contributor to the overall measurement uncertainty of the new setup.

6.5.3 Comparison to State-Of-The-Art Facilities

The exceptionally low measurement uncertainty of $U_{\rm Isc} \approx 0.70\%$ for large solar cells imposes a detailed comparison of the new Laser-DSR setup

 $^{^{24}}$ Using the Monte Carlo method detailed in Appendix E.

to other DSR-facilities. However, as the concepts (and sometimes even the objectives) of these setups typically vary significantly, a true quantitative comparison proves to be inappropriate in most cases. Therefore, the following discussion starts with a qualitative comparison of the new setup to state-of-the-art DSR-measurement facilities used for calibration of solar cells at Fraunhofer ISE CalLab. Afterwards, the new setup will be compared to a much more similar concept developed at PTB Braunschweig [17] based on the, to the best of the author's knowledge, available information.

The currently applied DSR-measurement systems at Fraunhofer ISE CalLab PV Cells are a grating [101] and a filter monochromator [107] setup. Whereas the grating monochromator features free choice of wave-lengths and bandwidths, only a limited number of fixed center wavelengths at given bandwidths is available in the filter monochromator setup. On the other hand, the grating monochromator setup provides several orders of magnitude less optical power so that it is predominantly used for relative SR-measurements, with an irradiation field significantly smaller than the active area of the device under test and for concentrator cells. In contrast, the filter monochromator setup provides enough irradiance for an uniform illumination of large area solar cells, potentially allowing for absolute SR-measurements.

Owing to its significantly higher spectral power (compare Fig. 6.9b) the new Laser-DSR setup allows for a combination of both: full flexibility in choice of wavelengths and uniform illumination of entire solar cells (up to $18 \times 18 \text{ cm}^2$ with the current optical setup). Moreover, the orders of magnitude higher spectral power allows for even lower bandwidths as compared to typical grating monochromator measurements. Another consequence of the increased spectral power is the significant improvement of signal-to-noise ratio allowing for reduced standard deviations and measurement duration. Another advantage related to the speed of the measurement system is given by the dual reference measurement mode that was introduced in Section 6.2.2. As two wavelengths are measured simultaneously, the measurement duration is halved. In combination with the speed enhancement owing to the increased signal level, a reduction of

measurement duration by a factor of 3 or 4 is expected for a comparable number wavelength steps.

Apart from these rather obvious improvements in speed, flexibility and resolution, the measurement uncertainty is significantly reduced as compared to the grating and filter monochromator setup currently being used in the Fraunhofer ISE CalLab PV Cells.²⁵

Regarding the spatial radiation properties the most striking improvement is the (virtual) elimination of any height dependence. It was shown that within ± 20 mm no variation in the averaged irradiance is observed, thereby canceling the related measurement uncertainty. Furthermore, the refined optical concept of the Laser-DSR setup yields an improvement in uniformity of the monochromatic irradiance field from approximately 10% in the filter monochromator setup to less than 3% non-uniformity. Consequently, the uncertainties related to spatial non-uniformity are significantly reduced. A similar effect results from chopping the monochromatic radiation prior to spatially homogenizing it in the fiber device and the rod (see Section 6.2.4). This concept results in a spatially constant phase across the entire measurement plane, making any consideration of spatial phase variations and related measurement uncertainties redundant [119].

Turning the discussion towards spectral uncertainties, it was demonstrated that the new setup allows for neglecting any uncertainty contribution related to undesired spectral components in the monochromatic radiation. Furthermore, the radiation bandwidth is significantly reduced from approximately 10 to 15 nm in the filter monochromator setup to less than 5 nm (optionally, less than 5 nm are available as well). Thus, the Laser-DSR setup achieves bandwidths-related uncertainties similar to those obtained by the grating monochromator setup. However, the uncertainty in center wavelength in the spectral range from 270 to 520 nm is assumed to be slightly higher with the new Laser-DSR setup. As no monochromator is applied in that spectral range, the repeatability of the

²⁵The following discussion of uncertainties related to spatial radiation properties is limited to the filter monochromator setup as the concept of measuring relative SRs, as with the grating monochromator, imposes a significantly deviating discussion of such uncertainties. For the spectral uncertainty discussion both grating and filter monochromator are considered.

laser itself is limiting, resulting in an estimated uncertainty of ± 1 nm. For wavelengths longer than 520 nm the uncertainty in center wavelengths is expected to be as low as in the grating monochromator setup.

In contrast to the very different filter and grating monochromator setup, a DSR-measurement facility very similar to the one presented in this chapter has been developed at the PTB Braunschweig [15, 17] at about the same time as this development at the Fraunhofer ISE took place. Utilizing a tunable ultrashort pulse laser system, significantly enhancing the pulse repetition rate by a fiber device and spectrally shaping the radiation with a grating monochromator, DSR-measuremements are conducted. Major conceptual differences of the setups are as follows: the PTB-setup does not allow for a simultaneous measurement at two wavelengths at a time, a single grating monochromator is used in the PTBsetup and the uniform illumination is achieved by two lenses imaging the monochromator output. Apart from slight deviations caused by application of a different combination of nonlinear optical processes to cover the entire spectral range, both setups provide approximately equal optical power in the measurement plane [17] (even in the dual reference mode of the ISE-setup).

Regarding measurement uncertainties, the PTB-setup achieves an expanded uncertainty of $U_{\rm Isc} = 0.4\%$ for 2x2 cm² cells, which represents the lowest reported uncertainty for DSR-measurements. Assuming the same reference uncertainty and taking into account minor uncertainty contributions being neglected in this chapter so far²⁶ the new Laser-DSR setup achieves $U_{\rm Isc} \approx 0.42\%$ for a 2x2 cm² GaAs cell measured with a silicon based reference. The same consideration assuming a large area

 $^{^{26}}$ As so are: spectral and spatial uncertainties of the bias irradiation (approximately 0.03% standard uncertainty each), instability in cell temperatures and imperfect monitoring principle (combined uncertainty of approximately 0.03%) and interpolation errors in wavelength and current (approximately 0.02% standard uncertainty each). As the reference cell has been dominating the uncertainty analysis in Section 6.4 so far, these contributions were neglected beforehand.

Furthermore, the reference object of the PTB is calibrated in units of A/W instead of A/W · m² as the reference cells considered so far. Applying an aperture, the irradiance (in W/cm²) and, from this, the effective area of the reference object $A^{\rm RC}$ are determined (applied in the correction term $f_{\rm Size}$ in Eq. (6.12)). Consequently, the uncertainty of the aperture itself (approximately 0.02%) needs to be taken into account as well [17].

15.6x15.6 cm² solar cell yields an expanded uncertainty of 0.48%. Thus, it is concluded that the new Laser-DSR facility at Fraunhofer ISE CalLab PV Cells is capable of achieving measurement uncertainties in the range of nowadays most accurate DSR-facilities.

Chapter 7

Supercontinuum Radiation for Solar Cell Characterization

In this chapter the application of spectrally shaped supercontinuum radiation for solar cell characterization is discussed. After motivating the advantages of supercontinuum radiation, an experimental setup for generation and precise spectral shaping of a supercontinuum is presented. Subsequently, a detailed analysis reveals that spectrally shaped supercontinuum radiation outperforms state-of-the-art solar simulators regarding spectral mismatch and its uncertainty. Moreover, a differential short circuit current measurement approach is presented that takes advantage of the precise spectral shaping capabilities while simultaneously overcoming the drawback of insufficient supercontinuum power. Finally, the potential of this new measurement method is emphasized by first measurement results. Parts of this chapter were presented in a previous publication of the author of this work [120].

7.1 Introduction

In the preceding Chapter 6 a new facility for differential spectral responsivity (DSR) measurements was presented, that has been developed in the course of this work. Moreover, in Section 6.4.3 it was demonstrated that the new setup allows for very low uncertainties in measuring the absolute spectral responsivity $s(\lambda)$. Consequently, and as outlined in Section 6.4.4, the short circuit current

$$I_{\rm SC} = \int s(\lambda) E_{\lambda}(\lambda) d\lambda, \qquad (7.1)$$

of the device under test can also be retrieved highly accurately when setting spectral irradiance $E_{\lambda}(\lambda)$ in Eq. (7.1) as a standard solar spectral irradiance distribution (e.g. as AM1.5g). However, the necessity of measuring $s(\lambda)$ with highest possible accuracy generally limits this method's speed regarding $I_{\rm SC}$ measurement. A much faster approach is represented by solar simulators that replace the mathematical integration in Eq. (7.1) by a physical integration using broadband light sources that imitate the considered standard solar spectrum (see also Section 2.2.2).

In order to achieve a best possible spectral match of simulator spectrum E_{Sim} and standard solar spectrum E_{STC} , spectrally filtered xenon or halogen lamps or, alternatively, a variety of light-emitting diodes (LEDs) are applied. However, due to their limited spectral shaping capabilities, a spectral mismatch

$$MM = \frac{\int s_{TC}(\lambda) E_{Sim}(\lambda) d\lambda \int s_{RC}(\lambda) E_{STC}(\lambda) d\lambda}{\int s_{TC}(\lambda) E_{STC}(\lambda) d\lambda \int s_{RC}(\lambda) E_{Sim}(\lambda) d\lambda}$$
(7.2)

remains that corrects for the differences of how a test cell (TC) and a reference cell (RC) evaluate the respective spectra (see Section 2.2.2.1). In addition to the applied correction, MM is subject of an uncertainty $u_{\rm MM}$ that reduces the overall measurement accuracy.

Owing to their much more precise spectral shaping capabilities, supercontinuum lasers have recently attracted interest regarding their applicability as radiation sources in solar simulators [121]. In fact, it was demonstrated by *Dennis* et al. [122] that individually manipulating spectral amplitudes of supercontinuum radiation allows for a virtually perfect imitation of standard solar spectra, thereby making any mismatch correction redundant (MM = 1 for $E_{\text{Sim}} = E_{\text{STC}}$ in Eq. (7.2)). Thus, making supercontinuum radiation sources applicable for solar cell characterization might allow for fast and accurate I_{SC} measurements.

7.2 Differential Supercontinuum Measurement Approach

Although their spectral features demonstrate the potential of outperforming any other solar simulator radiation source, the output power of nowadays commercially available supercontinuum lasers impedes their usage in solar simulators for large area industrial solar cells. In order to overcome this limitation a supercontinuum-based differential measurement approach is presented in this chapter that has been developed in the course of this work [120].

The new approach takes advantage of combining a chopped and low powered, but (nearly) perfectly shaped supercontinuum spectrum with high powered bias irradiation of a much less demanding spectrum. Measuring the differential current response of the device under test, the $I_{\rm SC}$ is directly measured as no spectral mismatch correction has to be applied.¹ In Fig. 7.1a the experimental setup for generation and spectral shaping of supercontinuum radiation is shown that has been developed based on the work presented by *Dennis* et al. in [122].

In the following subsections the individual parts of the experimental approach are explained in more detail.

¹Naturally, this does only hold for a shaped supercontinuum spectrum that perfectly resembles the considered standard solar spectrum. Moreover, the simplification of linear cells is underlying this statement. The case of nonlinear cells is briefly addressed in Section 7.4.



Figure 7.1: (a) Schematic setup for generation and shaping of supercontinuum radiation. The ultrashort pulses originate from the same laser system that is being used for the measurement facility presented in Chapter 6 (compare Fig. 6.1). The supercontinuum is generated inside a photonic crystal fiber (PCF) and spectrally shaped by a spatial amplitude mask and a grating light valve (GLV). (b) Illustration of a single GLV pixel with some voltage applied to the deflected ribbons resulting in an optical path length difference for radiation incident from top (image from [123]).

7.2.1 Generation of Supercontinuum Radiation

Instead of applying a commercially available supercontinuum laser, the developed approach takes advantage of the laser radiation emitted from the measurement facility presented in Chapter 6. Coupling ultrashort laser pulses of approximately 100 fs pulse duration into a photonic crystal fiber (PCF, *FemtoWHITE 800*/NKT Photonics) broadband supercontinuum radiation is generated by highly nonlinear optical effects.² Amplitude and spectral shape of the generated supercontinuum vary with incident pulse intensity and wavelength. For typical input parameters applied in this work, a supercontinuum ranging from 450 to more than 1600 nm is generated. A typical supercontinuum spectrum generated with the setup

 $^{^{2}}$ In Section 3.2.5 a brief description of the physical processes underlying supercontinuum generation is given. For a more profound discussion the interested reader is referred to [29].

is given as dashed blue line in Fig. 7.2a for the spectral range up to 1200 nm, that is most relevant for silicon solar cell measurements.

7.2.2 Spectral Shaping of Supercontinuum Radiation

The supercontinuum radiation generated by the PCF is collected by a microscope objective (not shown in Fig. 7.1a) and spectrally dispersed by a prism made of F2 flint glass. In analogy to the prism monochromator described in Section 6.2.3 and Fig. 6.4, a concave mirror is applied to collimate different spectral components while simultaneously focusing identical ones in the focal plane of the mirror. The spectral shaping itself is conducted in that focal plane by a combination of a Grating Light ValveTM(GLV) from Silicon Light Machines and a spatial amplitude mask.

The GLV is a dynamically adjustable phase grating that consists of an array of aluminum coated silicon nitride ribbons of 2-4 μ m width and 100-300 μ m length. Six of these form a single pixel of approximately 25.5 μ m pitch that is located over a common electrode as shown in Fig. 7.1b [123]. If a voltage is applied to the ribbons they are deflected towards the substrate by electrostatic force. Applying a voltage to every second ribbon induces an optical path length difference between neighboring ribbons resulting in a (partial) diffraction of the incident radiation. Setting the voltage to zero, the ribbons move back to the non-deflected state and the GLV acts as a mirror. By variation of the applied voltage the optical path length difference can be tuned between 0 and approximately 240 nm in 1024 steps allowing for a pixel-wise control of diffraction efficiency. As the setup shown in Fig. 7.1a exhibits a distinct wavelength-to-pixel relation the GLV therefore allows for spectrally resolved amplitude variations of the back-reflected 0th order beam. Slightly tilting the GLV and implementing an additional mirror separates the input radiation from its back-reflected counterpart and completes this fully computer-controlled approach for real-time variations of the shaped supercontinuum output.

Full extinction requires perfectly destructive interference and, thus, fulfillment of the $\lambda/4$ -condition. As a consequence, the full extinction potential of the GLV is limited to wavelengths below approximately 950 nm. Therefore, a specifically designed spatial amplitude mask is additionally


Figure 7.2: (a) Unshaped (dashed blue line) and shaped (solid blue) supercontinuum (SC) spectrum that mimics the AM1.5g standard solar spectrum (black). The red line indicates a 5 nm bandwidth moving average of the AM1.5g standard spectrum. Published in [120]. (b) Comparison of computed spectra combining SC with additional UV radiation to the AM1.5g standard solar spectrum (black lines). UV sources are either frequency-doubled SHG radiation (bottom, blue line, 425 nm center wavelength), a single LED at 425 nm center wavelength (middle, red line) or two LEDs at 365 and 425 nm center wavelengths (top, green line).

inserted close to the GLV to allow for more efficient extinction of wavelengths longer than 950 nm by (rather) fixed losses.

In Fig. 7.2a a shaped supercontinuum (solid blue line) that mimics the AM1.5g (black line) is shown, demonstrating the excellent spectral match of the shaped supercontinuum obtained from the presented setup. It is noteworthy, that this shaped spectrum rather represents a first shaping attempt with the presented setup and that substantial improvements in spectral match are expected to be realized in near future. Especially the development of a refined control of the setup's spectral output, being conducted in the course of X. Wang's master thesis that is supervised by the author of this work [124], might improve spectral shaping capabilities significantly.

7.2.3 Prospects for Tackling the Lack of UV Radiation

In addition to the excellent spectral match of shaped supercontinuum and AM1.5g standard spectrum, the lack of UV radiation below approximately 460 nm is apparent from Fig. 7.2a. As this lack induces a non-vanishing spectral mismatch correction, following prospects for tackling that missing spectral part are being discussed in the next section:

- add frequency-doubled radiation of the incident laser pulse (SHG)
- add one or more LEDs in the UV spectral range
- apply a PCF generating supercontinuum radiation in the entire spectral range

The first two options might be realized by combining the additional radiation from frequency-doubling or LEDs with the shaped supercontinuum radiation by the optical mixing rod applied in the measurement system (see e.g. Figs. 6.6 and 6.7). Moreover, the frequency-doubled radiation is directly available from the SHG-unit of the same measurement facility (compare Fig. 6.1). UV-LEDs are commercially available at several center wavelengths. Spectral shaping of either additional radiation source by the presented shaping setup is unfavorable due to their small bandwidth (SHG, see Fig. 6.2a) or their incoherent nature (LEDs). In Fig. 7.2b computed spectra obtained when adding SHG or LED radiation to the supercontinuum spectrum are shown.

The third option requires application of a new PCF that is capable of covering the entire relevant spectral range by supercontinuum radiation. In fact, researchers have recently demonstrated that they are capable of producing a PCF that exhibits this feature [125]. Assuming application of such a PCF allows for a virtually perfect imitation of a standard solar spectrum and, thus, appears to be most promising.

The potential of either of these approaches regarding spectral mismatch and its uncertainty is assessed in the next section.

7.3 Spectral Performance of Shaped Supercontinuum

7.3.1 Spectral Mismatch and Uncertainty

In order to assess the performance of shaped supercontinuum radiation as compared to state-of-the-art broadband sources applied in solar simulators, the spectral mismatch MM and its assigned uncertainty $u_{\rm MM}$ are evaluated in this section.

As introduced above (see Eq. (7.2)), the MM compensates for deviations of test and reference cell spectral responsivities ($s_{\rm TC}$ and $s_{\rm RC}$) as well as deviations of simulator and standard solar spectrum ($E_{\rm Sim}$ and $E_{\rm STC}$). Thus, measurements with any combination of these four attributes might be corrected by MM to achieve the test cell's $I_{\rm SC}$ under a standard solar spectrum according to

$$I_{\rm SC}^{\rm STC,TC} = \frac{I_{\rm SC}^{\rm Sim,TC}}{\rm MM} \frac{I_{\rm SC}^{\rm STC,RC}}{I_{\rm SC}^{\rm Sim,RC}}.$$
(7.3)

Whereas the MM is merely a correction term that might even vanish (MM = 1), the limited accuracy in measuring $s_{\rm TC}$, $s_{\rm RC}$ and $E_{\rm Sim}$ results in an uncertainty of the spectral mismatch $u_{\rm MM}$ that is non-vanishing in any case. As this uncertainty affects the overall accuracy of measuring $I_{\rm SC}^{\rm STC,TC}$, a comprehensive evaluation of a broadband sources' spectral performance requires consideration of $u_{\rm MM}$.

For assessing $u_{\rm MM}$ the individual uncertainties in $s_{\rm TC}$, $s_{\rm RC}$ and $E_{\rm Sim}$ need to be considered. In this work, the uncertainties in $s_{\rm TC}$ and $s_{\rm RC}$ are taken from [126] where a reference solar cell measured at the PTB³ and test cells measured with a filter monochromator DSR-setup are assumed. In Fig. 7.3 spectral responsivities and expanded uncertainties for respective test and reference solar cells are shown.⁴

³Physikalisch-Technische Bundesanstalt, German National Metrology Institute.

⁴The increasing uncertainty of the test cell measurement for longer wavelength mainly results from the test cell's band edge. Therefore, the wavelength-dependent



Figure 7.3: Spectral responsivity and associated expanded uncertainty of (a) a test solar cell measured with a filter monochromator DSRfacility and (b) a reference solar cell measured at the PTB. Uncertainty data for (a) and (b) are taken from [126].

The uncertainty in E_{Sim} is given by

$$u_{\rm Sim} = \left[\left(\frac{\partial E_{\rm Sim} \left(\lambda \right)}{\partial \lambda} \frac{u_{\rm WL}}{E_{\rm Sim} \left(\lambda \right)} \right)^2 + \left(\frac{\partial^2 E_{\rm Sim} \left(\lambda \right)}{\partial \lambda^2} \frac{u_{\rm bw}^2}{2E_{\rm Sim} \left(\lambda \right)} \right)^2 + u_{\rm NL}^2 + u_{\rm drift}^2 \right]^{1/2}$$
(7.4)

taking into account wavelength uncertainty $u_{\rm WL} = \pm 0.3$ nm, spectral resolution $u_{\rm bw} = \pm 3$ nm, nonlinearity $u_{\rm NL} = 1\%$ and long-term drift $u_{\rm drift} = 1\%$ of the spectrometer applied for measuring $E_{\rm Sim}$. These represent the four most important uncertainty contributions and are based on the spectral irradiance measurement analysis conducted in [16]. Further it is assumed, that the spectrometer is corrected for scattered light and is thermally stable and that the measured irradiance is incident on the detector at angles smaller than 40°. Typical uncertainties $u_{\rm Sim}$ are shown in Fig. 7.4 and demonstrate that sharp spectral features, as present in the

uncertainty given in Fig. 7.3a is contracted to the spectral responsivity range of nonsilicon test solar cells considered in the analysis below.



Figure 7.4: (a) Typical spectral irradiance distributions E_{Sim} of a LED and xenon solar simulator. (b) Uncertainties in spectral irradiance measurement u_{MM} of the simulator spectra shown in (a) according to Eq. (7.4).

wavelength range from 800 to 1000 nm of the xenon spectrum, substantially increase u_{Sim} .

For computation of $u_{\rm MM}$ the Random Walk Monte Carlo method as proposed in [126] is applied. The Random Walks in that method account for the correlation of uncertainties of neighboring wavelengths in $s_{\rm TC}$, $s_{\rm RC}$ and $E_{\rm Sim}$. In total 10,000 independent Random Walks from pseudo-random numbers are processed. They are normalized so that their standard deviations satisfy the given, wavelength-dependent uncertainties of $s_{\rm TC}$, $s_{\rm RC}$ and $E_{\rm Sim}$ (compare Figs. 7.3 and 7.4b) [127]. Finally, and in analogy to Section 6.4.4 and Fig. 6.13, the standard deviation of the resulting 10,000 MM values computed from Eq. (7.2) gives $u_{\rm MM}$.

7.3.2 Capabilities of Shaped Supercontinuum

In a first consideration, MM and $u_{\rm MM}$ are computed for 14 silicon solar test cells measured against another silicon reference cell (for exemplified spectral responsivities see Fig. 7.6a) under following spectral irradiance distributions: a Class C and a Class A xenon solar simulator (classes refer to IEC 60904-9, Ed. 2), a two-source simulator that combines a xenon and a halogen lamp, a LED simulator consisting of 18 different LEDs and the spectrally shaped supercontinuum as shown in Fig. 7.2a. In order to demonstrate the capabilities of shaped supercontinuum radiation regarding its spectral performance, the considered spectral range is firstly restricted to wavelengths that are covered by the currently available supercontinuum radiation ($\lambda > 460$ nm).

The mean values of MM and $u_{\rm MM}$ over all 14 cells are denoted by the single point in Fig. 7.5a, the straight line marks their median and the box illustrates the first and third quantiles. The whiskers denote the minimum and maximum values. The shaped supercontinuum considerably outperforms the state-of-the-art solar simulators. The spectral mismatch MM is much closer to unity and, more importantly, the uncertainty of this mismatch $u_{\rm MM}$ is significantly reduced.

As it was mentioned above, the currently applied shaped supercontinuum represents a rather first attempt of GLV-based spectral shaping. From the results presented by *Dennis* et al. [122] a much more precise spectral match is expected for a refined version of GLV-control and setup. Therefore, a perfectly shaped supercontinuum spectrum has been assumed and added to the simulations shown in Fig. 7.5a ("AM1.5q"). Per definition, the MM equals unity for all combinations of different cells. However, in agreement with a previous statement in this section, $u_{\rm MM}$ is non-vanishing. Considering that the assumed perfect spectrum extinguishes any still clearly present spectral mismatch of the currently available shaped supercontinuum (compare Fig. 7.2a), the reduction of $u_{\rm MM}$ is even surprisingly low. That this result can be attributed to the limited accuracy in measuring $E_{\rm Sim}$, becomes obvious when adding a moving average of the AM1.5g standard solar spectrum to the simulation (in this case with 5 nm bandwidth, shown as red line in Fig. 7.2a, denoted by "ave. AM1.5g" in Fig. 7.5a). As the moving average version exhibits less distinct spectral features (compare discussion of Fig. 7.4), $u_{\rm Sim}$ is reduced resulting in a reduction of $u_{\rm MM}$ that is clearly visible in Fig. 7.5a.

Thus, it might be concluded that even though a perfect spectral shaping is feasible with supercontinuum radiation sources, a less detail-rich spectrum might significantly reduce the overall measurement uncertainties. It has to be mentioned, that this conclusion is strongly related to the



Figure 7.5: Spectral mismatch MM and its uncertainty $u_{\rm MM}$ when measuring 14 silicon solar test cells with a silicon solar reference cell under various spectral irradiance distributions. (a) Limited to the spectral range where supercontinuum radiation is available in the current setup ($\lambda > 460$ nm). (b) Entire relevant spectral range from 300 to 1200 nm.

assumed accuracy in measuring the solar simulator's spectral irradiance distribution $E_{\rm Sim}$. With way more precise measurement tools, $u_{\rm MM}$ of the perfect AM1.5g approaches that one of the moving average version. However, with the simulation approach presented in this section, $u_{\rm MM}$ of the perfect spectrum never succeeds in outperforming its moving average version.



Figure 7.6: (a) Sampling of silicon solar test cells (solid lines) assumed for simulations presented in Fig. 7.5. The solar cell with spectral responsivity shown as dashed black line has been taken as reference cell for the simulations. Published in [120]. (b) Non-silicon (and spectrally filtered silicon) solar cells considered for simulations presented in Fig. 7.7. Again the dashed black line from (a) has been applied as reference solar cell's spectral responsivity. Published in [120].

7.3.3 Full Spectrum Analysis

After the previous demonstration of how a shaped supercontinuum outperforms state-of-the-art solar simulators given that it covers the considered spectral range, the simulation is extended to the entire relevant spectral range from 300 to 1200 nm in this section (see Fig. 7.5b). Whereas there are only minor changes from Fig. 7.5a to Fig. 7.5b for the considered solar simulator spectra, the supercontinuum performance clearly degrades due to the lack of UV radiation in the currently available shaped supercontinuum. The MM spreads significantly and $u_{\rm MM}$ increases substantially, but both are still comparable to other solar simulators. Thus, the currently available supercontinuum radiation might still be applied for $I_{\rm SC}$ measurements of silicon solar cells while another silicon cell is used as reference.

The impact of lacking UV radiation becomes more dramatic when nonsilicon and spectrally filtered silicon solar test cells are added to the simulation (see Fig. 7.6b for the test cells' spectral responsivities). In Fig. 7.7



Figure 7.7: Spectral mismatch MM and its uncertainty $u_{\rm MM}$ for the entity of cells shown in Fig. 7.6. In addition to previous spectral irradiance distributions, those are added that combine supercontinuum radiation with additional UV sources as shown in Fig. 7.2b.

data points corresponding to the box plots are additionally given and highlight that the unfavorably poor spectral match of reference and test cell yields a significant increase in $u_{\rm MM}$ for all spectral irradiance distributions. In fact, the accumulation of data points at the very bottom of each box corresponds to the results shown in Fig. 7.5b when only silicon cells were considered. Moreover, the MM spreads for all spectra, but most pronounced for the shaped supercontinuum due to the lack of UV radiation.⁵

In Section 7.2.3 various approaches were discussed to compensate that lack of UV radiation resulting in the suggested spectra shown in Fig. 7.2b.

⁵The as well dramatic increase in MM for the Class C xenon spectrum results from a spectral filter applied to the xenon lamp that highly reduces the amount of UV radiation.

As shown in Fig. 7.7, applying additional SHG radiation to the supercontinuum indeed significantly improves the MM values, thereby becoming comparable to the other solar simulators. However, this goes along with a dramatic increase of $u_{\rm MM}$ that is attributed to the large gradient of the SHG radiation and the resulting increase of $u_{\rm Sim}$ for measuring that spectrum. A significantly better performance is achieved by replacing the SHG radiation with a LED emitting at the same center wavelength. As the LED exhibits a much broader bandwidth (15 nm as compared to 4 nm for the SHG), $u_{\rm Sim}$ for measuring the supercontinuum plus LED spectrum is only slightly higher as $u_{\rm Sim}$ for measuring the supercontinuum spectrum itself. At the same time, nearly identical MM values are obtained as for adding SHG radiation. Thus, LEDs are clearly favorable over SHG radiation for compensating the lack of UV radiation in the currently available supercontinuum.⁶

However, as stated above, applying a new type of PCF that generates a supercontinuum covering the entire relevant spectral range appears to be most promising. Assuming that case and a refined shaping setup (see discussion above), a virtually perfect AM1.5g spectrum can be achieved. Comparing the results for this perfect AM1.5g replication and a moving average version of the AM1.5g emphasizes again, that shaping a less detail-rich version is favorable. It is not only less complex but also yields lower uncertainties. Moreover, the remaining high uncertainties for the group of poorly matching reference and test cells in "AM1.5g" and "ave. AM1.5g" demonstrate, that spectrally well matching cells are still required for achieving lowest measurement uncertainties in spite of a nearly perfectly shaped spectrum.

⁶Adding a further LED slightly improves the MM further at the cost of an as well slight increase in $u_{\rm MM}$ and an additional component. Although the two LEDs approach might be useful in some cases, the improvement compared to a single LED does not seem to justify the application of a further LED.

7.4 Short Circuit Current Measurements

The spectral performance analysis conducted in Section 7.3 demonstrated that the currently available supercontinuum radiation allows for $I_{\rm SC}$ measurements of silicon solar cells despite the lack of UV radiation.

Thus, in very first measurements with the new experimental approach (presented in Section 7.2), the $I_{\rm SC}$ s of two 4 cm² silicon solar cells have been measured.⁷ The results are shown in Table 7.1 and demonstrate an excellent agreement of $I_{\rm SC}$ s obtained from the new differential supercontinuum approach (SC) to those from the DSR-method, that exhibits lowest uncertainties but is orders of magnitude slower (compare discussion in Section 7.1). Naturally, the $I_{\rm SC}$ measurement with the currently available supercontinuum still requires a spectral mismatch correction due to its lack of UV radiation. Consequently, the spectral responsivity of the device under test $(s_{\rm TC})$ has to be measured as well to achieve MM according to Eq. (7.2). However, due to the twofold appearance of s_{TC} in Eq. (7.2), a relative measurement of $s_{\rm TC}$ suffices, thereby significantly reducing the spectral measurement effort. Furthermore, by application of an appropriate PCF, generating a supercontinuum that covers the entire spectral range, any mismatch correction becomes redundant and $s_{\rm TC}$ is not required anymore for $I_{\rm SC}$ measurements with this new approach.

The cells applied for the measurements presented in Table 7.1 are considered to be linear. Thus, the level of bias irradiation does not vary the $I_{\rm SC}$ response of the test cells. However, it has to be mentioned that the presented measurement approach readily allows for fast and accurate $I_{\rm SC}$ measurements of nonlinear cells as well. In that case several $I_{\rm SC}$ measurements at different bias irradiation levels are required to achieve the $I_{\rm SC}$ under standard test conditions (STC) as described in Section 2.2.2.2.

Furthermore and in analogy to the white-light-response (WLR) method proposed [115], this approach yields that level of bias irradiation that represents STC and that needs to be applied for absolute spectral responsivity

⁷For these measurements the same optical setup as introduced in Fig. 6.7 has been applied. As discussed there, this currently limits the irradiated area to $5x5 \text{ cm}^2$. However, it has to be mentioned that the generated supercontinuum power is sufficiently high to measure I_{SC} s of large area solar cells (up to 18 cm² edge length).

measurements. Thus, the differential supercontinuum approach might be readily implemented in the spectral responsivity measurement routine of the new setup (presented in Chapter 6) for evaluating test cell linearity and appropriate bias irradiation level.

7.5 Conclusions

In this chapter a new measurement approach was presented that takes advantage of the spectral shaping capabilities of supercontinuum radiation. Using the ultrashort laser pulses from the laser system applied in Chapter 6, a supercontinuum was generated by highly nonlinear optical effects in a PCF. Afterwards the supercontinuum was spectrally shaped to resemble a desired standard solar spectrum by real-time amplitude variations using a GLV and spatial amplitude masks.

In order to overcome the general problem of low output power from supercontinuum sources, a differential approach was presented for fast and accurate $I_{\rm SC}$ measurements. Illuminating a device under test by steady and high powered bias irradiation in addition to chopped supercontinuum radiation, allows for retrieving the differential current response of the test cell to the virtually perfect supercontinuum spectrum. First experimental results demonstrated the excellent accuracy of this new approach.

Cell	MM	$u_{ m MM}/\%$	$I_{ m SC}/{ m mA}$		Dev /%
			\mathbf{SC}	DSR	Dev./ /0
Si A	1.021	0.16	143.98	144.06	0.05
Si B	1.004	0.12	129.17	129.28	0.08

The general capabilities of supercontinuum radiation were demonstrated by a detailed analysis of spectral mismatch and its uncertainty.

Table 7.1: $I_{\rm SC}$ s measured with differential supercontinuum approach (SC) and compared to the $I_{\rm SC}$ s from the differential spectral responsivity (DSR) method demonstrating excellent agreement. Moreover, the spectral mismatches (MM) and associated uncertainties ($u_{\rm MM}$) are given.

The shaped supercontinuum is capable of outperforming any state-ofthe-art solar simulator regarding spectral properties. The analysis also revealed two major conclusions for the further development of supercontinuum solar simulators: although a virtually perfect replication of a standard solar spectrum can be achieved, firstly, a less detail-rich version of a standard spectrum is favorable and, secondly, spectrally well matching reference cells are still required for achieving lowest possible measurement uncertainties.

In future development steps of this approach, a new PCF should be applied that generates a supercontinuum covering the entire relevant spectral range, thereby making any spectral mismatch corrections redundant. In the meantime, applying an additional UV-LED covering parts of the currently missing UV spectrum appears to be most promising regarding spectral mismatch and its uncertainty.

Chapter 8

EQE-Measurement of CPV Modules

In this chapter a novel experimental approach for a direct measurement of the external quantum efficiency (EQE) of concentrator photovoltaic (CPV) modules will be presented that has been developed in the course of this work.¹ Firstly, the state-ofthe-art in EQE determination of CPV modules and challenges involved in a direct measurement of this EQE will be outlined. Afterwards it will be discussed, which special requirements an experimental setup for such a direct measurement needs to meet and how these are achieved in the presented approach. Finally, a validation of this novel experimental approach will be given by comparing measured short circuit current density and optical efficiency of a CPV mono module with independent indoor and outdoor measurements as well as simulations. Parts of the results of this chapter were published in [128].

¹The work presented in this chapter has been accomplished in collaboration with Christoph Rapp and Thomas Mißbach from the CPV-calibration group at Fraunhofer ISE.

8.1 Challenges and State-Of-The-Art

The external quantum efficiency (EQE) is an important measurand for characterization and calibration of photovoltaic (PV) devices (see also section Section 2.2.2.2). Firstly, it gives spectrally resolved insight into the electro-optical performance of a device and, secondly, it enables spectral mismatch corrections as well as highly precise short circuit current density determination for any spectral irradiance E_{λ} via

$$J_{\rm SC} = \int \text{EQE}(\lambda) E_{\lambda}(\lambda) \frac{q\lambda}{hc_0} d\lambda.$$
(8.1)

Especially for multi-junction cells a precise $J_{\rm SC}$ determination of the individual sub-cells is of tremendous importance, since imperfections in spectral mismatch correction might result in current-limiting artifacts that bias the electrical characterization of multi-junction devices. As these cells are typically applied in highly concentrating PV (CPV) modules,² that focus the sun's irradiation onto a smaller cell area, knowledge of the individual sub-cell $J_{\rm SC}$ and, thus, of the CPV module EQE (EQE_{mod}) is essential.

However, the various approaches for generation of quasi-monochromatic radiation vital for EQE-measurements are limited to EQE-measurements of PV cells³ or non-concentrator (flat) PV modules [17, 53, 101, 105, 107, 110]. When it comes to CPV modules, above measurement approaches cannot be applied without any further angular radiation shaping. This results from the fact that the concentrator optics in CPV modules are specifically designed for the angular properties of the solar radiation. Consequently, any deviations in the angular properties of the radiation from EQE-measurement facilities to the sun's angular properties result in erroneous concentrator and, thus, current generation performance. Therefore, any EQE-measurement approach for CPV modules requires a

²Throughout this chapter *module* denotes the combination of a single solar cell and any optical component (e.g. glasses, lenses or mirrors) and it does not necessarily include the consideration of interconnection of multiple cells inside a module.

³Naturally, the term *cells* includes non-concentrator and concentrator cells.

tight control of the angular radiation characteristics (in addition to its spectral and spatial properties).

As there is no measurement facility available yet for a direct EQEmeasurement of CPV modules, EQE_{mod} is typically computed from measured cell-EQEs and simulated optical efficiencies η_{opt} of the concentrator optics according to

$$\mathrm{EQE}_{\mathrm{mod}}^{\mathrm{comp}}\left(\lambda\right) = \mathrm{EQE}_{\mathrm{cell}}^{\mathrm{meas}}\left(\lambda\right)\eta_{\mathrm{opt}}\left(\lambda\right). \tag{8.2}$$

The spectral optical efficiency $\eta_{\text{opt}}(\lambda)$ of the concentrator optics generally depends on various parameters like e.g. geometry, absorption and refractive index, that are partially temperature-dependent. For siliconeon-glass Fresnel lens concentrators *Hornung* et al. developed a FEMbased numerical model for calculation of $\eta_{\text{opt}}(\lambda)$ that they successfully validated by experimental results [129]. These results were combined with solar irradiation spectra and ambient temperature-conditions to estimate the temperature-impact on energy generation in CPV modules [130]. *Steiner* et al. [131] applied both, $\eta_{\text{opt}}(\lambda)$ and standard spectral irradiance $E_{\lambda}(\lambda) \doteq \text{AM1.5d}$ (according to ASTM G173-03 [132]), to compute individual sub-cell J_{SC} s of multi-junction cells according to

$$J_{\rm SC} = \int {\rm EQE}_{\rm cell}^{\rm meas}(\lambda) \,\eta_{\rm opt}(\lambda) \, E_{\rm AM1.5d}(\lambda) \, \frac{q\lambda}{hc_0} {\rm d}\lambda. \tag{8.3}$$

With these sub-cell J_{SC} s they simulated the current-voltage characteristics of a CPV module taking into account the cell interconnections and predicted the energy yield over a period of one year with a maximum deviation of 3%.

Although the above outlined approach has been applied successfully, direct experimental access to EQE_{mod} might significantly reduce the effort as an individual consideration of $\eta_{\text{opt}}(\lambda)$ would become redundant. Therefore, an experimental approach for direct measurement of the EQE of a CPV module has been developed in the course of this work [128], that takes advantage of the new laser-based measurement facility presented in Chapter 6. In the subsequent sections this approach will be detailed and results will be presented that demonstrate its robustness, reproducibility and precision.

8.2 Experimental Approach

8.2.1 Spectral, Spatial and Angular Requirements

Spectral and Spatial Requirements

Regarding the spectral and spatial properties of the radiation that will be applied for EQE-measurements of CPV modules, identical requirements as for cell-EQE or non-CPV module measurements hold. A spectrally narrow radiation is favorable as the spectral resolution of the setup scales with radiation bandwidth. Also spatial invariance needs to be achieved, so that either the device under test (DUT) or the intensity distribution of the radiation itself need to be spatially homogeneous. In the latter case the radiation field might outshine the DUT, which is then tested in its entity so that non-uniform features of the DUT itself are negligible. In the former case the DUT needs to be larger than the radiation field, but homogeneous, so that the field can be non-uniform in turn. Any deviations from these demands lead to additional corrections of the measured EQE that intrinsically incorporate measurement uncertainties (see also Section 6.4.1.2).

Angular Requirements

In contrast to the spectral and spatial requirements that hold for both non-CPV and CPV measurements, the angular properties of the radiation field are solely crucial for CPV applications. This rises from the concentrator optics used in CPV modules that are specifically designed for the angular characteristics of the sun, commonly called *sunshape*. Any deviations of the angular radiation properties from the sunshape result in a different imaging performance of the concentrator optics and, thus, a potentially different current generation of the DUT. Therefore, the monochromatic



Figure 8.1: Various sunshapes as published by [133–135]. The DLR mean profile [134] is used as reference profile for the angular characteristics of the radiation subsequently.

radiation used for the EQE-measurements of CPV modules needs to imitate the solar angular distribution.

In a most straightforward geometrical consideration, that takes into account the diameter of the sun disk $(d_{\text{Sun}} = 1.391684 \cdot 10^9 \text{ m})$ and the averaged distance from sun to earth $(d_{\text{Sun-Earth}} = 1.496 \cdot 10^{11} \text{ m})$, the angular sun radius

$$\theta_{\rm Sun} = \arctan\left(d_{\rm Sun}/2d_{\rm Sun-Earth}\right) = 4.65\,{\rm mrad}$$
(8.4)

is achieved. This value has also been experimentally confirmed by *Puliaev* et al. in 2000 [133]. However, the angular intensity distribution across the sun disk is not constant due to the sun's radial temperature profile [134]. In addition to that, multiple scattering processes in the earth's atmosphere smear the disk profile so that it is surrounded by an aureole, called the *circumsolar region*. Therefore, numerous sunshape measurements were conducted in the past.⁴ However, as the circumsolar ratio (CSR), the ratio of circumsolar intensity relative to the entire sun's intensity, varies with climate, season, location, day of time and wavelength [137], no distinct terrestrial sunshape can be identified. Instead, mean average sunshapes for various CSR conditions were published by e.g. *Neumann* et al. in

 $^{^{4}}$ A detailed discussion of the different measurements and approaches is beyond the scope of this work. The interested reader is referred to *Buie* et al. [136].

2002 [134]. Based on a statistical analysis of their data Neumann et al. also published a single mean profile, subsequently called DLR mean profile, that serves for an annual statistical analysis of concentrator optics performance. In Fig. 8.1 the DLR mean profile, sunshapes for various CSR conditions [134] and a standard solar scan as published by *Rabl* and *Bendt* in 1982 [135] are shown to illustrate the variety of sunshapes. The vertical line marks the solar disk radius [133].

As the DLR mean profile represents a statistical annual average of the sunshape, this profile is used as a reference profile for the angular shaping of the monochromatic radiation discussed subsequently.

8.2.2 Setup Description

In order to obtain the previously described spectral, spatial and angular radiation properties required for EQE-measurements of CPV modules, the ultrashort pulse laser system of the setup presented in Section 6.2 has been equipped with several components for spatio-temporal and angular shaping of the radiation. As apparent from the schematic setup shown in Fig. 8.2, no monochromator is applied in this experimental approach so that the bandwidth of the quasi-monochromatic radiation stays in the natural range of the ultrashort pulses (see Fig. 6.2a), which is sufficiently narrow for a demonstration of functionality of this experimental approach. However, for an advanced stage of this approach, monochromators could be easily implemented.

Spatio-Temporal Shaping of Ultrashort Laser Pulses

As it was discussed in Chapter 4 the usage of ultrashort laser pulses in characterization of solar cells might result in undesired current-generation from two-photon absorption. Especially in concentrator applications, as present in this experimental approach, care has to be taken that the ultrashort pulses are temporally shaped so that no nonlinear effects appear. Therefore, the ultrashort pulses are converted into continuous radiation by a prototype of the fiber-based pulse-to-cw-converter that was detailed in Chapter 5. The prototype version has been designed for the direct coupling of laser radiation with Gaussian intensity distribution (see Fig. 8.3).

8.2 Experimental Approach



Figure 8.2: Schematical representation of the experimental approach for EQE-measurement of CPV modules. Published in [128].



Figure 8.3: Schematical representation of the monolithic pulse-to-cw converter prototype consisting of 19 fibers with distinct lengths L_1 to L_7 tapered and spliced to a long multimode fiber. The insets illustrate the evolution of the temporal pulse shapes while the radiation propagates through the fiber device, the close-ups show the proximal end of the device and the bundle to multimode splice.



Figure 8.4: Angular shaping of radiation: (a) Schematic representation of the angular shaping of an extended source by an optical element. (b) Angular shaping of pulse-to-cw converter output with off-axis parabolic mirror (OPM) as applied in the experimental approach. Fresnel lens and solar cell illustrate a typical CPV module.

The individual lengths of the 19 fibers are carefully adjusted according to their spatial position at the converter's proximal end so that each ultrashort pulse is divided into seven temporally equidistant pulses of nearly identical amplitude (please note the length denotation L_1 to L_7 in the fiber cross section in Fig. 8.3). At their distal end the 19 fibers are tapered and spliced to a 100 m long multimode fiber with 800 μ m core diameter that temporally broadens the individually pulses till they temporally overlap at the multimode fiber's distal end.⁵ Additionally, the spatial intensity distribution is homogenized inside the long multimode fiber by mode coupling processes. Thereby a nearly top-hat intensity distribution is achieved at the output of the pulse-to-cw converter (compare Fig. 5.11 in Section 5.4.1), that is crucial for the angular shaping discussed subsequently.

Angular Shaping of Radiation

In order to convert the radiation emitted by any extended source into a desired angular distribution the straightforward approach sketched in Fig. 8.4a can be applied. Placing the extended source at the front focal

⁵Based on the idea, design and specifications developed in the course of this work, the converter has been manufactured at the Fraunhofer Institute for Applied Optics and Precision Engineering (IOF) in Jena, Germany.



Figure 8.5: Analysis of angular properties of radiation field: (a) CMOS-camera image in the focal plane of a lens placed at the target position of the experimental setup. (b) Angular distribution of the setup achieved from radially integrated back-transformation of the spatial information shown in (a) into angular information. Published in [128].

plane of an optical element, illustrated by the thin lens in Fig. 8.4a, converts the spatial property of the extended source into an angular property according to $d\theta \approx -dx/f$ with dx denoting the distance to the optical axis of the system (in paraxial approximation). Thus, all rays emitted from the point $dx = d_{\text{Source}}/2$ will propagate at an angle of $\theta = -d_{\text{Source}}/2f$ to the target plane. For an extended source with homogeneous lateral intensity distribution (as in case of the nearly top-hat fiber output described above), the angular distribution is as well a top-hat distribution. Other angular profiles could be created by an appropriate variation of the lateral intensity distribution of the extended radiation source.

In Fig. 8.4b the adaption of this angular shaping technique for the EQE-measurement setup is schematically shown. The distal end of the pulse-to-cw converter serves as extended light source and an 30°-off-axis parabolic mirror (OPM) is applied as achromatic optical element with focal length f = 76.2 mm. Focal length of OPM and core diameter of multimode fiber yield a theoretical outer angle of $\theta_{\text{max}} = 5.25 \text{ mrad}$ that is slightly larger than the angular sun radius $\theta_{\text{Sun}} = 4.65 \text{ mrad}$.

For an experimental analysis of the angular distribution created by



Figure 8.6: Measured spatial intensity distribution after the $25x25 \text{ mm}^2$ aperture. The Gaussian intensity distribution results from the output characteristics of the pulse-to-cw converter. Published in [128].

the combination of pulse-to-cw converter and OPM, a lens of known focal length f = 125 mm has been placed at the target position (where the CPV module will be located in the measurements). In analogy to the previous discussion the lens transforms the angular into a spatial distribution detected by a CMOS-camera. From the imaged intensity distribution (see Fig. 8.5a) the angular properties of the radiation field have been deduced by the paraxial relation $\Delta r \approx f \cdot \Delta \theta$ and radial integration. The result of this analysis is shown in Fig. 8.5b and demonstrates the excellent match of measured angular distribution of the experimental setup and DLR mean profile.

Spatial Shaping of Radiation

Although the fiber end facet is close to a top-hat intensity distribution each surface-point of the fiber still emits the radiation Gaussian-like within a numerical aperture given by the fiber properties. As a consequence, the fiber output has a Gaussian intensity distribution in the farfield or at the position of the OPM, respectively. Therefore, a $25x25 \text{ mm}^2$ spatial aperture is placed in the beam path in order to transmit the rather homogeneous central part of the Gaussian distribution (see Fig. 8.6). The remaining non-uniformity of this $25x25 \text{ mm}^2$ field, which is in the range of 70%, is not critical, as the radiation field is smaller than the DUT that is assumed to be spatially invariant. It is noteworthy, that a smaller aperture would significantly improve the non-uniformity to e.g. lower than 10% for a 10x10 mm² aperture.

Peripheral Devices and Reference Measurement

The EQE-measurements are conducted with the DSR-method [14]. Thus, the setup (Fig. 8.2) is completed by a chopper wheel, lock-in amplifiers, transimpedance amplifiers and a halogen lamp for bias irradiation. These components are identical to those applied in the new DSR-facility presented in Chapter 6 and are given in Section 6.2.5. Furthermore, a beam splitter is implemented that routes approximately 8% of the radiation onto a silicon monitor cell for tracking intensity fluctuations of the monochromatic radiation during the measurement. For a reference measurement the DUT is replaced by a silicon cell of well-known spectral characteristics.

8.3 Validation of Measurement Approach

8.3.1 Device Under Test: CPV Mono Module

For a demonstration of applicability of this novel measurement approach that provides direct experimental access to the EQE of CPV modules, three so called FLATCONTM-type [138] CPV mono modules⁶ have been used, that have well-known characteristics owing to long-term outdoor measurements. Each mono module consists of a Fresnel lens and an entirely processed lattice matched three-junction PV cell that has only the $Ga_{0.50}In_{0.50}P$ top, the $Ga_{0.99}In_{0.01}As$ middle or the Ge bottom junction electrically activated. With these so-called isotype cells current-limiting artifacts, that might heavily affect short circuit current measurements, are avoided and each junction can be characterized individually.

 $^{^6{\}rm The}$ word mono denotes that there are no cell interconnections, even though multiple cells and lenses are assembled into one physical module.

8.3.2 Measurement Results

In Fig. 8.7a the cell and mono module EQEs for an electrically active top (blue circles), middle (green squares) and bottom cell (red triangles) are plotted. The cell EQEs (open symbols) have been taken from grating monochromator measurements of identically processed cells of the same wafer [101]. The mono module EQEs (filled symbols) have been measured with the experimental approach discussed in this chapter.

The mono module EQEs imitate the general trend of the cell EQEs quite well and demonstrate robustness, as the standard deviations, that are included as error bars, are smaller than the symbol for wavelengths longer than 380 nm ($< 0.5\%_{\rm rel}$). 380 nm wavelength marks the lower wavelength limit as the intrinsic absorption of the prototype pulse-tocw-converter increases dramatically for shorter wavelengths. This is also apparent from the substantial increase in standard deviation caused by a very low measurement signal at 380 nm. The longer wavelength limit is set to 1040 nm as the oscillation features of the cells, induced by the top-cell anti-reflection coating and thin-film interference, could not have been resolved without the grating monochromator that has not yet been implemented at the time these measurements have been conducted (compare Fig. 6.1). Moreover, the measurement approach demonstrates a very good reproducibility that is apparent from the different color tones in the middle cell module EQE, each representing a measurement on a different day.

The middle cell is of especial importance here as its EQE is entirely covered by the presently available spectral range. This allows a short circuit current density $(J_{\rm SC})$ computation with the module EQE according to Eq. (8.1) with $E_{\lambda}(\lambda) = AM1.5d$ as standard spectrum for direct irradiance scaled to 1000 W/m^2 [132]. In Table 8.1 the $J_{\rm SC}$ from the presented EQE-approach is given and compared to $J_{\rm SC}$ s deduced from indoor simulator and outdoor measurements. It is noteworthy that no temperature corrections have been applied to the optics. Taking the temperature related uncertainties into account the low deviations of the $J_{\rm SC}$ values can be regarded as a validation of the experimental approach.



Figure 8.7: (a) Cell and mono module EQEs of the CPV module under test. Cell EQEs are known from grating monochromator measurements of identically processed cells from the same wafer [101]; the module EQEs have been measured with the setup presented here. Published in [128]. (b) Measured optical efficiency of CPV module for top and middle cell versus simulated optical efficiency of a same type Fresnel lens at 25°C digitized from [131].

Moreover, there is an obvious drop in the EQE from cell to CPV module level, that is expected because of reflection, absorption and scattering losses of the concentrator optics. This drop is equivalent to the previously introduced optical efficiency $\eta_{\text{opt}} = \text{EQE}_{\text{module}}/\text{EQE}_{\text{cell}}$ (see e.g. Eq. (8.2)). In Fig. 8.7b the optical efficiency from the presented measurements is compared to a simulation from *Steiner* et al. [131] for a same

Method	EQE-Approach	Indoor	Outdoor
$J_{ m SC}[{ m mA/cm}^2]$	11.6	11.5	12.5
Deviation to EQE-Approach $[\%]$	_	0.9	7.2

Table 8.1: Integrated short circuit current density $J_{\rm SC}$ of presented EQE-approach compared to an indoor simulator and an outdoor measurement (both scaled to $1000 \,\mathrm{W/m^2}$).

type Fresnel lens at 25°C lens temperature (which is close to the temperature conditions for the presented experimental data). The bottom cell as well as EQE values lower than 50% are omitted in this graph, as they are strongly affected by spectral uncertainties and bandwidth deviations from cell to module measurement. The measurement result shows a good agreement to the simulation data, especially the characteristic drop at short wavelengths, that is caused by absorption and chromatic aberration of the silicon-on-glass Fresnel lens, is imitated very well. The overall conformity of the middle cell measurement is less pronounced than for the top cell measurement and a slight wavelength-dependent drift of the data is apparent. This might be due to misalignment, but needs further investigation.

The presented results demonstrated that the experimental approach can be applied successfully for a direct EQE-measurement of CPV modules. In a further development of this experimental approach the spectral range should be extended and the angular distribution should be optimized by custom-made fiber tips and/or OPMs to improve the angular match shown in Fig. 8.5b. Finally, the approach should be applied to true multi-junction cells with adapted bias irradiation to saturate the sub-cells that are not being measured.

Chapter 9

Summary & Outlook

Highly accurate electrical characterization of photovoltaic devices is of tremendous importance for assessing advances in solar cell technology and for reducing uncertainties in solar cell production lines. As the main contribution to the overall measurement uncertainty in production lines results from the short circuit current $(I_{\rm SC})$ of the applied reference cell, a reduction of the $I_{\rm SC}$ uncertainty is of major interest. Nowadays lowest reported $I_{\rm SC}$ uncertainties are obtained from differential spectral responsivity (DSR) measurements. A further reduction of these uncertainties is (often) impeded by the limited spectral power provided by conventional DSR-facilities.

In this work a DSR-setup was developed that overcomes this problem by taking advantage of the high spectral output power provided by laser radiation. In order to enable coverage of the entire spectral range, ultrashort laser pulses and several nonlinear optical components were applied for this purpose. The work resulted in realization of a measurement facility that reduces the present $I_{\rm SC}$ uncertainty for large area solar cells at Fraunhofer ISE CalLab by at least $25\%_{\rm rel}$.

Subsequently, main achievements of this work are briefly summarized:

(1) A theoretical model addressing the interaction of ultrashort laser pulses and solar cells under short circuit conditions was developed. The model was validated by an experimental method enabling temporal energy redistribution within an ultrashort pulse train by ring cavities. A semi-analytical approach for solving the model facilitated the comparison of solar cell $I_{\rm SC}$ s generated under pulsed versus continuous illumination. This comprehensive study revealed the fundamental conclusion that temporal shaping of ultrashort pulses is advisable prior to their usage for highly accurate solar cell characterization.

(Chapter 4, partially published in [33])

- (2) An innovative monolithic fiber-based device was developed for conversion of an ultrashort pulse train into continuous radiation in an efficient, robust and reproducible manner. Furthermore, a new method for Estimation of Fiber Properties from Impulse Responses (EFPIR-method) was established. With EFPIR attenuation and mode coupling properties of step-index multimode fibers can be retrieved from impulse responses. This method represents the first such characterization approach that does not require any farfield output patterns of the fiber under test. It allows for prediction of impulse and frequency responses as well as bandwidths and coupling lengths of step-index multimode fibers. (Chapter 5, partially published in [68])
- (3) A new DSR-facility was developed based on a wavelength-tunable ultrashort pulse laser system. A prism monochromator was designed and constructed under supervision of the author of this work for reducing the rather broad bandwidths of the ultrashort pulses. The fiber devices mentioned in (2) are applied for temporal shaping of the pulses. An optical setup was designed that allows for highly efficient illumination of large area solar cells with less than 3% irradiance non-uniformity. A detailed uncertainty analysis of the new measurement system was conducted. It revealed a remarkable relative improvement of at least $25\%_{\rm rel}$ in $I_{\rm SC}$ uncertainty compared to currently applied systems at Fraunhofer ISE CalLab. (Chapter 6, partially published in [53])

9 Summary & Outlook

- (4) A supercontinuum-based method was developed that enables fast and accurate I_{SC} measurements. The supercontinuum was generated by coupling ultrashort pulses emitted from the applied laser system into a photonic crystal fiber. Real-time spectral shaping of the supercontinuum was conducted by a specifically designed optical setup using a spatial mask and a dynamically adjustable phase grating. The supercontinuum was shaped to imitate a standard solar spectrum and was capable of outperforming any state-of-the-art solar simulator regarding the spectral mismatch and its uncertainty. First experiments demonstrated speed and accuracy of this very promising new characterization method. (Chapter 7, partially published in [120])
- (5) The new DSR-facility and a prototype of the fiber device for temporal shaping were applied for external quantum efficiency (EQE) measurements of concentrator photovoltaic (CPV) mono modules.¹ These measurements represent the worldwide first EQE measurements of such a concentrator module, thereby enabling a new method for the electro-optical performance characterization of CPV modules and demonstrating the potential of the new DSR-setup beyond typical DSR-measurement applications. (Chapter 8, partially published in [128])

Subsequently, the results of this work are presented in more detail:

In **Chapter 4** the interaction of ultrashort laser pulses and solar cells under short circuit conditions was studied in a comprehensive theoretical and experimental analysis.

The increased spectral power of ultrashort pulses as compared to conventionally applied white light sources appears to make ultrashort pulse lasers a very promising tool for highly accurate solar cell characterization. However, any deviation in the short circuit current $(I_{\rm SC})$ generated by

¹This work was conducted in collaboration with C. Rapp and T. Mißbach both working with the CPV-calibration group at Fraunhofer ISE.

ultrashort pulses as compared to continuous radiation might impede their usage in such measurements.

In order to assess these potential deviations a theoretical model describing the interaction of ultrashort pulses and solar cells was developed. The most general form of the model takes into account possible nonlinearities arising from injection dependence of minority carrier lifetimes, saturation of carrier generation and two-photon absorption (TPA). Detailed discussions and implementation of a quasi-steady-state approximation resulted in substantial simplifications of the model with TPA remaining the only significant nonlinear contribution for most cases.

Appearance and significance of TPA in excess carrier generation was proven experimentally. For finding this experimental evidence, a sophisticated optical setup was devised that enables convenient temporal pulse energy redistribution within a pulse train by two ring resonators with adjustable cavity lengths. In such way, the temporal shape of an incident ultrashort pulse train was varied with negligible variations in average power, direction and focal spot size. Consequently, the observed changes in $I_{\rm SC}$ with varying pulse shapes could be attributed to nonlinear carrier generation by TPA.

For validating the theoretical model based on the experimental findings, the model was solved for the generated I_{SC} . Implementing an extremely high front surface recombination in the model imitated the strong charge carrier extraction in short circuit conditions. This strongly confined recombination feature requires fine spatial and temporal discretization, representing a crucial bottleneck in fully numerical approaches. This is overcome by adapting a semi-analytical approach using a Green's function to solve the theoretical model.

Finally, the validated model was applied for the most prominent, but experimentally hardly feasible, comparison of $I_{\rm SC}$ s generated from ultrashort pulses and continuous illumination. The analysis revealed that certain circumstances yield a significant contribution of TPA to the $I_{\rm SC}$. This represents the first theoretical and experimental evidence-based finding, that allows for recommending temporal shaping of the ultrashort laser pulses prior to their usage in highly accurate solar cell characterization facilities.

In **Chapter 5** the temporal shaping requirement revealed in Chapter 4 was tackled by converting ultrashort pulses into continuous wave (cw) radiation.

For this purpose a monolithic fiber-based device was developed that takes advantage of modal dispersion in multimode fibers and optical path length differences. Firstly, the pulse repetition rate is enhanced by homogeneous illumination of a bundle consisting of 19 optical fibers of distinct different lengths. Secondly, the generated sub-pulses are re-combined in a single multimode fiber and temporally stretched by modal dispersion so that they overlap with their next neighbors. Homogenization of spatial intensity distribution by mode coupling is exploited for homogeneous fiber bundle illumination and serves robustness and reproducibility of pulseto-cw-conversion. Moreover, the homogeneous output of the fiber device was used for generation of monochromatic radiation with sun-like angular properties (as discussed in Chapter 8).

During the work on that fiber device, a new method for Estimation of Fiber Properties from Impulse Responses (EFPIR-method) was proposed. With impulse responses from two different fiber lengths and initial radiation characteristics as input parameters, the EFPIR-method reveals mode coupling and attenuation properties of step-index multimode fibers by means of an iterative downhill simplex algorithm. The fiber properties obtained from the EFPIR-method can be applied for prediction of impulse and frequency responses as well as bandwidths and coupling lengths. The EFPIR-method represents the first such characterization method being purely based on impulse responses and makes spatial output patterns for fiber characterization redundant.

In Chapter 6 the new differential spectral responsivity (DSR) setup, that was developed in the course of this work, was described in detail.

An initial recapitulation of the ultrashort pulse laser system properties emphasized the necessity of applying spectral shaping components for bandwidth reduction and elimination of undesired spectral components (as e.g. remaining fundamental radiation after frequency doubling or tripling). Combining bandpass filters, a prism and a grating monochromator was identified as ideal solution regarding optical efficiency and speed, as it allows for simultaneous measurement at two different quasimonochromatic wavelengths. Moreover, prism and grating monochromator can be optimized for their specific spectral ranges. Whereas a broad variety of grating monochromators is commercially available, no such solutions for prism monochromators have been known. Therefore, a prism monochromator was developed and constructed under supervision of the author of this work exhibiting an excellent wavelength repeatability of less than 0.13 nm.

A crucial requirement for highly accurate DSR-measurements is uniform illumination of the device under test. Ray-tracing simulations were conducted to design a novel optical system that images the homogeneous intensity distribution of a mixing rod's exit facet in the measurement plane. An appropriate choice of optical components resulted in illumination of a 16x16 cm² area with less than 3% non-uniformity. A downscaled (optional) second version yields illumination of a 4x4 cm² area with 13 times higher irradiance and approximately 1% non-uniformity (experimentally confirmed). Moreover, spatial phase variations of the chopped monochromatic radiation are eliminated by the new optical configuration, thereby canceling a significant uncertainty contribution present in other DSR-setups.

An output power analysis demonstrated that the new setup outperforms state-of-the-art filter- or grating-monochromator based DSR-setups by a wavelength-dependent factor of 10 to 500 (filter) or 100 to 10,000 (grating), respectively. Furthermore, the achieved output power is very comparable to a similar DSR-setup that was developed at PTB Braunschweig at about the same time as this development took place.

Chapter 6 was concluded by a detailed measurement uncertainty analysis starting from a discussion of the individual uncertainty contributions. Applying a Monte Carlo method for uncertainty propagation, spectrally resolved and integrated combined measurement uncertainties were determined. The analysis revealed an expanded² $I_{\rm SC}$ uncertainty of 0.66% for 2x2 cm² and 0.70% for 15.6x15.6 cm² solar cells, which is a relative improvement of at least 25%_{rel} compared to state-of-the-art measurement setups at Fraunhofer ISE CalLab. The uncertainty analysis further identified the uncertainty of the reference solar cell as major contribution. Using a calibrated photodiode instead of a solar cell as reference object, the expanded uncertainty in $I_{\rm SC}$ can be reduced down to 0.42% (for 2x2 cm² solar cells) and 0.48% (for 15.6x15.6 cm² solar cells). This represents a relative improvement of approximately 50%_{rel} compared to the applied state-of-the-art measurement setups and is comparable to the best yet reported uncertainties for primary solar cell calibration.

In Chapter 7 a fast and accurate I_{SC} measurement approach based on spectrally shaped supercontinuum radiation was presented.

Coupling ultrashort laser pulses emitted by the laser system applied in Chapter 6 into a photonic crystal fiber (PCF) the high pulse peak power and dispersion characteristics of the PCF result in generation of a broadband supercontinuum. The coherence properties of the supercontinuum allow for precise spectral shaping enabling a (virtually) perfect imitation of any standard solar spectrum. In this work, the shaping was accomplished by a prism and a grating light valve³ (GLV). The prism dispersed the spectral components that were subsequently real-time modified in their individual amplitudes by the GLV.⁴ First shaping attempts showed an excellent spectral match of shaped supercontinuum and AM1.5g standard solar spectrum.

Furthermore, a detailed Random Walk Monte Carlo analysis regarding the spectral mismatch and its uncertainty was performed. It revealed that spectrally shaped supercontinuum radiation outperforms any stateof-the-art solar simulator. Although being capable of perfectly imitating

²Expanded uncertainty refers to a coverage probability of 95.45% ($k_{cov} = 2$).

³A grating light value is a dynamically adjustable phase grating that diffracts a controllable amount of incident radiation into higher diffraction orders.

 $^{^4\}mathrm{An}$ additional spatial amplitude mask compensates for reduced extinction ratios of the GLV above 950 nm.

a standard solar spectrum, the analysis further demonstrated that resembling a smoother version of such a spectrum (as e.g. a moving average) results in a reduction of mismatch uncertainties. This finding is of major importance for the further development of solar simulators based on supercontinuum radiation sources. Also it was demonstrated that spectrally well matching reference cells are still required for highly accurate measurements in spite of virtually perfect spectral shaping capabilities.

The shaped supercontinuum developed in this work was further applied for $I_{\rm SC}$ measurements. In order to overcome the current problem of limited supercontinuum output power impeding one sun illumination of large industrial solar cells, a differential measurement approach was proposed in this work. Illuminating a solar cell with high powered bias irradiation of less demanding spectral properties in addition to chopped and low powered, but nearly perfectly shaped supercontinuum radiation facilitates fast and accurate differential $I_{\rm SC}$ measurements. First $I_{\rm SC}$ measurements of linear silicon solar cells deviated by less than 0.1% from their respective calibrated value demonstrating accuracy of this new measurements approach.

In Chapter 8 external quantum efficiency (EQE) measurements of concentrator photovoltaic (CPV) mono modules were presented that were conducted with the developed laser-based DSR-facility (see Chapter 6).⁵ These represent the worldwide first EQE measurements of a CPV module and demonstrate the application potential of the new setup beyond standard measurement schemes.

Taking advantage of spatial homogenization in a prototype of the fiberbased pulse-to-cw-converter (see Chapter 5), monochromatic radiation with sun-like angular properties was generated. Measurements of this angular distribution demonstrated excellent agreement to the sun's properties. Thus, the crucial requirement for measuring CPV modules, that utilize concentrator optics specifically designed and optimized for the sun's angular distribution, was fulfilled making EQE and $I_{\rm SC}$ measurements of such modules feasible. The performed measurements showed a good

⁵This work was conducted in collaboration with C. Rapp and T. Mißbach both working with the CPV-calibration group at Fraunhofer ISE.

agreement to independent measurements and simulations regarding $I_{\rm SC}$ and optical module efficiency. Thereby applicability of the developed measurement approach was demonstrated that enables a new way for characterization of electro-optical CPV module properties.

Outlook

Subsequently, pending work and investigations related to this thesis are outlined.

Within the next months, the new DSR-facility presented in Chapter 6 will be completed by implementing some yet missing or not installed components (grating monochromator, final version of pulse-to-cw-converter, optics for large area illumination). Afterwards, the developed measurement software⁶ will be completed followed by intense testing of software and setup. This initialization phase is followed by several months of parallel and comparative DSR and $I_{\rm SC}$ measurements that will give detailed insight into the actual performance characteristics of the new setup. Based on this experiences, the measurement uncertainty analysis presented in Chapter 6 is going to be validated or adjusted in minor contributions. Finally, setup and method will be implemented into the accredited part of the calibration lab and released for solar cell calibrations (aiming) at unprecedented accuracy.

Apart from the remaining steps for full integration of the new setup into the calibration routine, the promising project of supercontinuum based $I_{\rm SC}$ measurements will be pursued (see Chapter 7). Current ideas are focusing on implementation of a new PCF capable of generating a supercontinuum that covers the entire relevant spectral range. Also, it is aimed for developing a refined software for GLV-control that exhibits much better spectral shaping capabilities as the currently applied one. After these setup improvements, extensive $I_{\rm SC}$ measurements of various solar cell types (silicon, non-silicon, linear, nonlinear) will be conducted in a comparative manner to asses the potential of the developed measurement approach. Owing to the first promising results, it is expected that

⁶The software development has not been discussed within the thesis.
the planned further development of this approach makes it competitive to state-of-the-art methods regarding measurement uncertainties. Given that this ambitious goal is reached, a further highly accurate $I_{\rm SC}$ measurement method is obtained being beneficial for the calibration process at Fraunhofer ISE CalLab.

Furthermore, a refined optical setup for concentrator cells and modules is under development. Implementation of this optical setup will enhance application capabilities of the new laser-based DSR-facility and will conveniently enable measurements like those presented in Chapter 8.

Deutsche Zusammenfassung und Ausblick

Hochpräzise elektrische Solarzellen-Messungen sind von wesentlicher Bedeutung für die Bewertung technologischer Fortschritte und die Minimierung von Produktionsunsicherheiten in der Solarzellen-Herstellung. Vor allem die Messunsicherheit des kalibrierten Kurzschlussstroms ($I_{\rm SC}$) der verwendeten Referenz-Solarzelle ist dabei von besonderem Interesse, da ein signifikanter Anteil der Gesamtmessunsicherheit in Produktionslinien auf diese zurückzuführen ist. Die geringsten Messunsicherheiten im $I_{\rm SC}$ werden heutzutage durch differentielle Messungen der spektralen Empfindlichkeit (DSR, aus dem Englischen: *Differential Spectral Responsivity*) einer Solarzelle erzielt. Eine weitere Verringerung der Messunsicherheiten bei DSR-Messungen wird (häufig) durch die vorhandene spektrale Leistung in typischen DSR-Messplätzen limitiert.

In dieser Arbeit wurde ein DSR-Messplatz entwickelt, der durch die Verwendung monochromatischer Laserstrahlung über deutlich höhere spektrale Leistungen verfügt. Um die notwendige spektrale Durchstimmbarkeit des Messplatzes zu gewährleisten, wurde dafür ein Ultrakurzpuls-Laser in Kombination mit weiteren nichtlinearen optischen Komponenten eingesetzt. Der entwickelte Messplatz verringert die Unsicherheiten in $I_{\rm SC}$ Messungen großer Industrie-Solarzellen um mindestens 25%_{rel} relativ zu bisher verwendeten Messsystemen im Fraunhofer ISE CalLab.

Nachfolgend werden die Hauptergebnisse dieser Arbeit kurz zusammengefasst:

- (1) Es wurde ein theoretisches Model entwickelt, dass die Wechselwirkung zwischen ultrakurzen Pulsen und Solarzellen unter Kurzschlussbedingungen beschreibt. Das Modell wurde durch ein Experiment validiert, welches die zeitliche Umverteilung von Pulsenergien innerhalb eines Zuges ultrakurzer Pulse ermöglichte. Ein halb-analytischer Ansatz wurde eingesetzt, um das theoretische Modell hinsichtlich der generierten $I_{\rm SC}$ s zu lösen und Vergleiche zwischen gepulster und kontinuierlicher Anregung zu ziehen. Diese Untersuchungen resultierten in der wichtigen Schlussfolgerung, dass eine zeitliche Umformung der ultrakurzen Pulse empfehlenswert ist, wenn diese ohne Einfluss nichtlinearer Effekte für hochpräzise Solarzellen-Charakterisierungen eingesetzt werden sollen. (Kapitel 4, teilweise publiziert in [33])
- (2) Um aufeinanderfolgende ultrakurze Laserpulse auf effiziente und reproduzierbare Weise in zeitlich kontinuierliche Strahlung zu konvertieren, wurde ein innovatives monolithisches Faserbauteil entwickelt. Außerdem wurde eine Methode zur Charakterisierung von Stufen-Index Multimode-Fasern erarbeitet, die die Abschätzung ihrer Modenkopplungs- und Dämpfungseigenschaften anhand von Impulsantworten ermöglicht (EFPIR-Methode, aus dem Englischen: *Estimation of Fiber Properties from Impulse Responses*). EFPIR ist die erste derartige Methode die allein auf der Verwendung von Impulsantworten basiert und keinerlei Fernfeld-Messungen der von den Fasern emittierten Strahlungsfelder erfordert. Mittels EFPIR können zudem Impuls- und Frequenzantworten sowie Bandbreiteneigenschaften und Kopplungslängen der untersuchten Fasern bestimmt werden.

(Kapitel 5, teilweise publiziert in [68])

(3) Basierend auf einem spektral durchstimmbaren Ultrakurzpuls-Lasersystem wurde ein neues DSR-Messsystem entwickelt. Im Zuge dessen wurde unter der Betreuung des Autors dieser Arbeit auch ein Prismenmonochromator entworfen und gebaut, der zur Reduzierung der spektralen Bandbreiten der ultrakurzen Pulse eingesetzt wird. Die in (2) erwähnten Faserbauteile werden zur Konvertierung der zeitlichen Pulseigenschaften eingesetzt. Des Weiteren wurde ein optisches System zur effizienten Beleuchtung von Industrie-Solarzellen mit weniger als 3% Strahlungsfeld-Inhomogenität entworfen. Schließlich wurde eine detaillierte Messunsicherheitsanalyse des neuen Messplatzes durchgeführt, die eine bemerkenswerte relative Verbesserung der $I_{\rm SC}$ -Messunsicherheit um mindestens $25\%_{\rm rel}$ verglichen mit den bisher verwendeten Messsystemen des Fraunhofer ISE CalLab gezeigt hat. (Kapitel 6, teilweise publiziert in [53])

(4) Es wurde eine Superkontinuum-basierte Methode zur schnellen und präzisen $I_{\rm SC}$ Messung entwickelt. Durch Einkopplung der vom verwendeten Lasersystem emittierten ultrakurzen Laserpulse in eine photonische Kristallfaser (PCF, aus dem Englischen: Photonic Crystal Fiber) wurde ein breitbandiges Superkontinuum erzeugt. Mittels räumlicher Amplitudenmasken und einem dynamisch modulierbaren Phasengitter (GLV, aus dem Englischen: Grating Light Valve) wurde ein optischer Aufbau entwickelt, der eine Echtzeit-Modifikation des Superkontinuum-Spektrums ermöglicht. Das von der PCF emittierte Superkontinuum wurde auf diese Art derart spektral modifiziert, dass es ein Standardsonnenspektrum nachbildet. Es konnte gezeigt werden, dass ein derartig erzeugtes Spektrum eine deutlich verbesserte spektrale Fehlanpassung und geringere Unsicherheiten aufweist als üblicherweise verwendete Sonnensimulatoren. Durch erste Experimente konnten Geschwindigkeit und Genauigkeit dieser vielversprechenden neuen I_{SC} Messmethode demonstriert werden.

(Kapitel 7, teilweise veröffentlicht in [120])

(5) Mit dem neu entwickelten DSR-Messplatz und einem Prototypen des in (2) erwähnten Faserbauteils wurden externe Quanteneffizienzen (EQE) von Konzentrator-Photovoltaik(CPV)⁷-Mono-Modulen

⁷CPV, aus dem Englischen: *Concentrator Photovoltaic*.

gemessen.⁸ Diese Messungen sind die weltweit ersten EQE-Messungen solcher CPV-Module und ermöglichen eine neuartige Charakterisierung elektro-optischer Eigenschaften von CPV-Modulen. Außerdem demonstrieren sie das Potential des neuen DSR-Messplatzes hinsichtlich möglicher Anwendungen, die weit über die typischen DSR-Messverfahren hinausgehen.

(Kapitel 8, teilweise veröffentlicht in [128])

Nachfolgend werden die Ergebnisse dieser Arbeit detaillierter zusammengefasst:

In **Kapitel 4** wurde die Wechselwirkung zwischen ultrakurzen Pulsen und Solarzellen in Kurzschlussbedingungen in einer umfangreichen theoretischen und experimentellen Arbeit untersucht.

Die erhöhte spektrale Leistung ultrakurzer Pulse im Vergleich zu üblicherweise eingesetzten Weißlichtquellen erscheint vielversprechend hinsichtlich ihrer Anwendung für hochpräzise Solarzellen-Messungen. Allerdings muss dafür gewährleistet sein, dass der unter Verwendung ultrakurzer Laserpulse erzeugte Kurzschlussstrom ($I_{\rm SC}$) nicht von dem unter kontinuierlicher Beleuchtung generierten abweicht.

Um die potentiellen Abweichungen bewerten zu können, wurde ein theoretisches Modell entwickelt, welches die Wechselwirkung zwischen ultrakurzen Laserpulsen und Solarzellen beschreibt. Das Modell berücksichtigt Nichtlinearitäten, die durch injektions-abhängige Ladungsträgerlebensdauern, durch Sättigung in der Generation von Überschussladungsträgern oder durch Zwei-Photonen-Absorption (TPA, aus dem Englischen: *Two-Photon Absorption*) entstehen können. Detaillierte Diskussionen und die Einführung einer quasi-statischen-Näherung resultierten in einer signifikanten Vereinfachung des Modells, welches für die meisten relevanten Fälle gilt und lediglich die TPA als nichtlinearen Term beinhaltet.

Die Relevanz der TPA bei der Ladungsträgergeneration mit ultrakurzen Pulsen konnte experimentell nachgewiesen werden. Dazu wurde

 $^{^8 \}rm Diese$ Arbeiten wurden in Zusammenarbeit mit C. Rapp und T. Mißbach aus der CPV-Kalibriergruppe des Fraunhofer ISE durchgeführt.

ein experimenteller Aufbau entwickelt, welcher durch Verwendung zweier in ihrer Länge justierbarer Ringresonatoren die Umverteilung von Pulsenergien innerhalb eines Zuges ultrakuzer Pulse ermöglicht. Auf diese Weise konnte die zeitliche Form der ultrakurzen Pulse variiert werden, ohne dabei die durchschnittliche Leistung, die Richtung oder die geometrischen Strahlungseigenschaften signifikant zu beeinflussen. Dadurch konnten die mit den zeitlichen Pulsformänderungen einhergehenden Variationen im gemessenen $I_{\rm SC}$ auf TPA zurückgeführt werden.

Um das theoretische Modell anhand dieser experimentellen Ergebnisse validieren zu können, wurde das Modell hinsichtlich des generierten $I_{\rm SC}$ gelöst. Die unter Kurzschlussbedingungen herrschende starke Extraktion der Überschussladungsträger wurde durch eine sehr hohe Oberflächenrekombinationsgeschwindigkeit simuliert, welche sehr feine räumliche und zeitliche Diskretisierungen erfordert. Da diese Diskretisierungen erheblichen Rechenaufwand in voll-numerischen Lösungsansätzen bedeuten, wurde ein halb-analytischer Ansatz unter Verwendung einer Greenschen Funktion für die Lösung des theoretischen Modells angewendet.

Abschließend wurde das derart validierte Modell für den bedeutendsten, jedoch experimentell schwer zugänglichen Vergleich generierter $I_{\rm SC}$ s unter gepulster und kontinuierlicher Bestrahlung angewendet. Durch diesen Vergleich konnte gezeigt werden, dass gewisse experimentelle Bedingungen zu einem signifikanten Beitrag der TPA zum generierten $I_{\rm SC}$ führen. Dies ermöglichte erstmalig die theoretisch und experimentell validierte Schlussfolgerung, dass zeitliche Pulsformungen empfehlenswert sind, um ultrakurze Pulse für die hochpräzise Vermessung von Solarzellen einzusetzen.

In **Kapitel 5** wurde die in Kapitel 4 dargelegte Notwendigkeit zeitlicher Pulsformungen durch die Konvertierung der ultrakurzen Pulse in zeitlich kontinuierliche Strahlung (cw., aus dem Englischen: *continuous wave*) gelöst.

Es wurde ein monolithisches, Faser-basiertes Bauteil entwickelt, welches Modendispersion in Multimode-Fasern sowie optische Weglängenunterschiede zur Puls-zu-cw-Konvertierung nutzt. In diesem Bauteil wird zunächst die Pulsfolgefrequenz der ultrakurzen Pulse durch homogene Beleuchtung eines aus 19 Einzelfasern unterschiedlicher Länge bestehenden Bündels erhöht. Anschließend werden die so erzeugten Teil-Pulse in einer einzelnen Multimode-Faser wiedervereint und durch Modendispersion derart verbreitert, dass sich benachbarte Pulse zeitlich überlappen. Die homogene Beleuchtung des Faserbündels wird in robuster und reproduzierbarer Weise durch Modenkopplung in einer Multimode-Faser realisiert. Diese Modenkopplung erzeugt ebenso eine homogene Intensitätsverteilung am Ausgang des Faserbauteils, was für die in Kapitel 8 vorgestellte Erzeugung monochromatischer Strahlung mit sonnenähnlichen Winkeleigenschaften ausgenutzt wird.

Während der Arbeiten am Puls-zu-cw-Konverter wurde des Weiteren eine neue Methode zur Abschätzung von Fasereigenschaften anhand ihrer Impulsantworten (EFPIR, aus dem Englischen: *Estimation of Fiber Properties from Impulse Responses*) entwickelt. Mit Impulsantworten bei zwei unterschiedlichen Faserlängen und den Eigenschaften der eingekoppelten Strahlung als Eingangsparameter, werden die Modenkopplungsund Dämpfungseigenschaften von Stufen-Index Multimode-Fasern mittels Downhill-Simplex-Verfahren bestimmt. Die ermittelten Fasereigenschaften eignen sich zur Vorhersage von Impuls- und Frequenzantworten sowie zur Bestimmung von Bandbreiten und Kopplungslängen der untersuchten Stufen-Index Multimode-Fasern. Die EFPIR-Methode ist das erste derartige Verfahren, welches ausschließlich auf Impulsantworten basiert und keinerlei räumliche Fernfeld-Messungen der von der Faser emittierten Strahlung benötigt.

In **Kapitel 6** wurde das im Rahmen dieser Arbeit entwickelte neue System zur Messung der differentiellen spektralen Empflindlichkeit (DSR, aus dem Englischen: *Differential Spectral Responsivity*) von Solarzellen vorgestellt.

Eine einleitende Zusammenfassung der Eigenschaften des verwendeten Ultrakurzpuls-Lasersystems machte dabei die Notwendigkeit deutlich, Elemente zur spektralen Anpassung einzusetzen, um spektrale Bandbreiten zu reduzieren und unerwünschte spektrale Anteile aus der Laserstrahlung zu eliminieren (welche z.B. durch verbleibende Fundamentalstrahlung nach Frequenzverdopplung oder -verdreifachung verursacht werden). Hinsichtlich optischer Effizienz und Geschwindigkeit des Gesamtsystems wurde die Kombination von Bandpassfiltern, einem Prismen- und einem Gittermonochromator als optimale Lösung erachtet, da die Verwendung zweier Monochromatoren die gleichzeitige Messung bei zwei unterschiedlichen Wellenlängen erlaubt. Zudem können Prismen- und Gittermonochromator für den jeweils verwendeten Spektralbereich optimiert werden. Da im Gegensatz zu dem breiten Angebot an kommerziellen Gittermonochromatoren keine vergleichbaren Prismenmonochromatoren bekannt waren, wurde unter Anleitung des Autors dieser Arbeit ein Prismenmonochromator entwickelt und gebaut, der eine sehr gute Wellenlängenreproduzierbarkeit von weniger als 0.13 nm aufweist.

Von entscheidender Bedeutung für hochpräzise DSR-Messungen ist die homogene Beleuchtung der Solarzellen. Mittels strahlenoptischer Simulationen wurde ein neues optisches System entworfen, welches die homogene Intensitätsverteilung am Ausgang eines optischen Mischers auf die Messebene abbildet. Durch geeignete Wahl der optischen Komponenten kann dadurch eine Inhomogenität kleiner als 3% über einer Fläche von 16x16 cm² erreicht werden. Eine verkleinerte Version des Aufbaus wurde ebenfalls ausgelegt, welche eine 4x4 cm² große Fläche mit ca. 1% Inhomogenität und 13-fach höherer Strahlungsintensität beleuchtet (experimentell validiert). Des Weiteren werden durch den entworfenen optischen Aufbau räumliche Phasenvariationen des frequenz-modulierten Lichts eliminiert, die in vorherigen Messplätzen zu beträchtlichen Messunsicherheitsbeiträgen führten.

Eine Untersuchung der optischen Ausgangsleistung des neuen DSR-Systems konnte zeigen, dass es (wellenlängenabhängig) die 10- bis 500fache Ausgangsleistung eines aktuell verwendeten DSR-Messplatzes basierend auf Weißlichtquellen und optischen Filtern liefert. Gegenüber typischer Gittermonochromator-basierter DSR-Systeme wird sogar die 100- bis 10.000-fache Ausgangsleistung erreicht. Im Vergleich zum Laserbasierten DSR-Messplatz der PTB Braunschweig, der ungefähr zur gleichen Zeit entwickelt wurde wie der im Rahmen dieser Arbeit entwickelte Messplatz, werden sehr ähnliche Ausgangsleistungen erzielt.

Kapitel 6 wurde mit einer detaillierten Messunsicherheitsanalyse des neuen Systems abgeschlossen. Dabei wurden zunächst die individuellen Beiträge zur Messunsicherheit detailliert beschrieben und abgeschätzt, bevor diese mittels einer Monte Carlo Analyse durch die Messung propagiert wurden, um spektral aufgelöste sowie spektral integrierte $(I_{\rm SC})$ kombinierte Messunsicherheiten zu bestimmen. Es konnte gezeigt werden, dass durch das neue Messsystem erweiterte Unsicherheiten⁹ im $I_{\rm SC}$ in Höhe von 0.66% für 2x2 cm² und 0.70% für 15.6x15.6 cm² Zellen erreicht werden. Dies stellt eine relative Verbesserung um mindestens $25\%_{\rm rel}$ im Vergleich zu bisher im Fraunhofer ISE CalLab verwendeten Messplätzen dar. Des Weiteren zeigte die Analyse, dass der größte Messunsicherheitsbeitrag durch die verwendete Referenz-Solarzelle verursacht wird. Würde stattdessen eine kalibrierte Photodiode als Referenz-Objekt eingesetzt werden, könnte die Unsicherheit im $I_{\rm SC}$ auf 0.42% (für 2x2 cm²) Solarzellen) und 0.48% (für 15.6x15.6 cm² Solarzellen) reduziert wer-Dies würde eine relative Verbesserung um ca. $50\%_{\rm rel}$ gegenüber den. bisher im CalLab verwendeter Messmethoden bedeuten und wäre vergleichbar zu den besten bisher veröffentlichten Messunsicherheiten für Primärkalibrierungen von Solarzellen.

In **Kapitel 7** wurde die Entwicklung eines schnellen und präzisen I_{SC} Messverfahrens basierend auf der Verwendung von Superkontinuumstrahlung vorgestellt.

Das dafür verwendete Superkontinuum wurde durch Einkoppeln der ultrakurzen Laserpulse des in Kapitel 6 verwendeten Lasersystems in eine photonische Kristallfaser (PCF, aus dem Englischen: *Photonic Crystal Fiber*) erzeugt. Die hohen Pulsspitzenleistungen und die Dispersionseigenschaften der PCF führen zu hoch-nichtlinearen Effekten über vergleichsweise lange Distanzen, sodass die quasi-monochromatische

⁹Erweitert bedeutet hier ein Vertrauensintervall in Höhe von 95.45% ($k_{cov} = 2$).

Eingangsstrahlung in sehr breitbandiges weißes Laserlicht konvertiert wird. Die Kohärenzeigenschaften des Superkontinuums erlauben sehr feine Manipulationen der spektralen Strahlungseigenschaften, sodass eine (nahezu) perfekte Kopie gewünschter Standardspektren erzeugt werden kann. Im Rahmen dieser Arbeit wurde für diese spektrale Manipulation ein optischer Aufbau realisiert, der die Strahlung zunächst durch ein Prisma dispergiert und die Amplituden der individuellen spektralen Anteile anschließend durch ein dynamisch modulierbares Phasengitter (GLV, aus dem Englischen: *Grating Light Valve*) in Echtzeit anpasst.¹⁰ In ersten Versuchen zur spektralen Anpassung konnte eine exzellente Übereinstimmung des angepassten Superkontinuums zum AM1.5g Standardspektrum gezeigt werden.

Um die Vorteile dieser spektral angepassten Strahlungsquelle bewerten zu können, wurde eine Random Walk Monte Carlo Analyse hinsichtlich der spektralen Fehlanpassung und ihrer Messunsicherheit durchgeführt. Es konnte gezeigt werden, dass das angepasste Superkontinuum signifikant bessere spektrale Eigenschaften zeigt als üblicherweise verwendete Sonnensimulatoren. Zudem wurde gezeigt, dass, obwohl die Superkontinuumsquelle dies ermöglichen würde, eine perfekte Nachbildung eines Standardspektrums durchaus höhere Messunsicherheiten bedingen kann als die Nachbildung einer weniger detail-reichen, gemittelten Version des Standardspektrums. Diese Schlussfolgerung ist von entscheidender Bedeutung für die weitere Entwicklung Superkontinuum-basierter Sonnensimulatoren. Des Weiteren wurde gezeigt, dass spektral gut angepasste Referenzzellen trotz perfekt angepasster Superkontinuumstrahlung notwendig sind, um möglichst geringe Messunsicherheiten zu erzielen.

Der in dieser Arbeit entwickelte experimentelle Aufbau wurde zudem für erste I_{SC} Messungen eingesetzt. Da die aktuellen Ausgangsleistungen von Superkontinuum-Strahlungsquellen zu gering sind für die Beleuchtung großflächiger Industrie-Solarzellen mit der notwendigen Intensität von einer Sonne, wurde ein differentielles Messverfahren angewendet. Das spektral nahezu perfekt angepasste Superkontinuum geringer Intensität

 $^{^{10}}$ Zusätzlich zum GLV wird eine statische Amplitudenmaske eingesetzt, welche die limitierte Amplituden
extinktion des GLV für Wellenlängen größer als 950 nm kompensiert.

wurde frequenz-moduliert und mit deutlich intensiverer Bias-Bestrahlung weniger anspruchsvoller spektraler Eigenschaften kombiniert. Durch Messung der differentiellen Stromantwort der Test-Solarzelle konnte dann der $I_{\rm SC}$ dieser Solarzelle auf schnelle und präzise Art und Weise bestimmt werden. Die ersten Messungen an linearen Silizium-Solarzellen zeigten bemerkenswert geringe Abweichungen von kleiner als 0.1% im $I_{\rm SC}$ gegenüber ihren mittels der DSR-Methode kalibrierten Stromwerten.

In Kapitel 8 wurden Messungen externer Quanteneffizienzen (EQE) von Konzentrator-Photovoltaik(CPV)¹¹-Mono-Modulen vorgestellt, die mit dem neu entwickelten Laser-DSR-Messplatz durchgeführt wurden.¹² Diese Messungen sind die weltweit ersten EQE-Messungen an CPV-Modulen und demonstrieren das Anwendungspotential des neuen Messplatzes, welches weit über die üblichen DSR-Messungen hinausgeht.

Zur Durchführung der Messungen wurde die homogene Intensitätsverteilung am Ausgang eines Prototypen des in Kapitel 5 entwickelten Pulse-zu-cw-Konverters genutzt, um monochromatische Strahlung mit sonnenähnlichen Winkeleigenschaften zu erzeugen. Die Überprüfung der erzeugten Winkelverteilung durch experimentelle Messungen zeigte eine exzellente Übereinstimmung mit den Sonneneigenschaften. Dadurch wurde das entscheidende Kriterium für die Messung von CPV-Modulen erfüllt, in denen Optiken verwendet werden, die speziell für diese Winkelverteilungen ausgelegt und optimiert sind. Die durchgeführten Messungen zeigten eine sehr gute Übereinstimmung mit unabhängigen Messungen des $I_{\rm SC}$ und Simulationen der optischen Moduleffizienz und validierten damit diese neue Messmethode zur Charakterisierung elektro-optischer Eigenschaften von CPV-Modulen.

Ausblick

Nachfolgend werden noch ausstehende und weiterführende Entwicklungen

¹¹CPV, aus dem Englischen: Concentrator Photovoltaic.

¹²Diese Arbeiten wurden in Zusammenarbeit mit C. Rapp und T. Mißbach aus der CPV-Kalibriergruppe des Fraunhofer ISE durchgeführt.

kurz zusammengefasst, die in direktem Zusammenhang mit den Ergebnissen dieser Arbeit stehen.

In den nächsten Monaten wird der im Rahmen dieser Arbeit entwickelte DSR-Messplatz durch den Einbau diverser noch fehlender bzw. noch nicht installierter Komponenten vervollständigt (Gittermonochromator, finale Versionen der Puls-zu-cw-Konverter, Optiken für die Messung großflächiger Industrie-Solarzellen). Anschließend wird die entwickelte Software zur Steuerung des Aufbaus und zur Durchführung der Messungen¹³ vervollständigt, um System und Software intensiv zu testen. In den auf diese Initialisierungsphase folgenden Monaten werden vergleichende DSR und $I_{\rm SC}$ Messungen durchgeführt, die detaillierte Auskunft über die tatsächliche Leistung des Messplatzes geben werden. Basierend auf diesen Erkenntnissen wird die in Kapitel 6 vorgestellte Messunsicherheitsanalyse validiert oder geringfügig angepasst. Abschließend werden Messsystem und -verfahren in den akkreditierten Bereich des Fraunhofer ISE CalLab PV Cells eingebunden, um Solarzellen-Kalibrierungen mit (wie zu erwarten) deutlich reduzierten Messunsicherheiten zu ermöglichen.

Neben diesen verbleibenden Arbeiten zur vollständigen Integration des Systems in den Kalibrierablauf des Fraunhofer ISE CalLab, soll das vielversprechende Projekt zur Superkontinuum-basierten $I_{\rm SC}$ Messung (siehe Kapitel 7) fortgesetzt werden. Aktuell wird dabei die Verwendung einer verbesserten PCF in Betracht gezogen, die das gesamte relevante Spektrum mit Superkontinuumstrahlung abdecken kann. Außerdem soll der experimentelle Aufbau durch optimierte Software signifikant verbessert werden, um eine noch bessere spektrale Anpassung zu ermöglichen. Anschließend sollen umfangreiche $I_{\rm SC}$ Messungen vergleichender Art durchgeführt werden, um die Möglichkeiten dieser neuen Messmethode analysieren und bewerten zu können. Dank der vielversprechenden ersten Ergebnisse ist zu erwarten, dass dieses neue $I_{\rm SC}$ Messverfahren wettbewerbsfähig zu anderen Verfahren wird und demzufolge die Möglichkeiten und das Portfolio des Fraunhofer ISE CalLab signifikant erweitert.

¹³Die Software-Entwicklung wurde in der vorliegenden Arbeit nicht beschrieben.

Des Weiteren wird an der Entwicklung eines zusätzlichen optischen Systems gearbeitet, das die Charakterisierung von Konzentrator-Zellen und -Modulen ermöglicht. Dieses soll in den Laser-DSR-Messplatz eingebaut werden und wird die Anwendungsmöglichkeiten des Systems deutlich erweitern, wie z.B. bereits in Kapitel 8 dieser Arbeit gezeigt.

Appendix A

Anharmonic Oscillator Solution

In Section 3.2.1 the anharmonic oscillator is introduced that allows for a description of nonlinear optical effects in the interaction of an electric field and a bound electron. As their is no general solution of Eq. (3.3) known for nonlinear restoring forces F_{restore} a perturbation theory is applied. Closely following the derivation shown in [28, 22-25] the anharmonic oscillator equation is solved for the lowest two correction-terms of a noncentrosymmetric potential energy function with $F_{\text{restore}} = -m \left(\omega_0^2 r + a_2 r^2\right)$ (where a_2 describes the strength of nonlinearity) in this appendix. As an example, these solutions of electron oscillation will be used to demonstrate how to retrieve the linear susceptibility and the second harmonic susceptibility.

The anharmonic oscillator equation with above given F_{restore} reads

$$\frac{\mathrm{d}^{2}\tilde{r}}{\mathrm{d}t^{2}} + 2\gamma \frac{\mathrm{d}\tilde{r}}{\mathrm{d}t} + \omega_{0}^{2}\tilde{r} + a_{2}\tilde{r}^{2} = -\frac{q}{m}\tilde{E}\left(t\right).$$
(A.1)

Applying a procedure similar to the Rayleigh-Schröder perturbation theory the applied electric field $\tilde{E}(t)$ is changed to $\kappa \tilde{E}(t)$ with κ denoting the strength of perturbation. With this and the power series expansion

$$\tilde{r} = \kappa \tilde{r}^{(1)} + \kappa^2 \tilde{r}^{(2)} + \kappa^3 \tilde{r}^{(3)} + \dots$$
(A.2)

Eq. (A.1) is solved so that each power of κ satisfies Eq. (A.1) individually.

With this the equations

$$\frac{\mathrm{d}^{2}\tilde{r}^{(1)}}{\mathrm{d}t^{2}} + 2\gamma \frac{\mathrm{d}\tilde{r}^{(1)}}{\mathrm{d}t} + \omega_{0}^{2}\tilde{r}^{(1)} = -\frac{q}{m}\tilde{E}(t)$$
(A.3a)

$$\frac{\mathrm{d}^2 \tilde{r}^{(2)}}{\mathrm{d}t^2} + 2\gamma \frac{\mathrm{d}\tilde{r}^{(2)}}{\mathrm{d}t} + \omega_0^2 \tilde{r}^{(2)} + a_2 \left(\tilde{r}^{(1)}\right)^2 = 0 \tag{A.3b}$$

$$\frac{\mathrm{d}^2 \tilde{r}^{(3)}}{\mathrm{d}t^2} + 2\gamma \frac{\mathrm{d}\tilde{r}^{(3)}}{\mathrm{d}t} + \omega_0^2 \tilde{r}^{(3)} + 2a_2 \tilde{r}^{(1)} \tilde{r}^{(2)} = 0 \tag{A.3c}$$

are obtained (when stopping the expansion after the third term).

With $\tilde{E}(t) = E_1 \exp(-i\omega_1 t) + E_2 \exp(-i\omega_2 t) + c.c.$ (with *c.c.* denoting the complex conjugate) Eq. (A.3a) is solved by

$$\tilde{r}^{(1)}(t) = r^{(1)}(\omega_1) \exp(-i\omega_1 t) + r^{(1)}(\omega_2) \exp(-i\omega_2 t) + c.c.$$
(A.4)

with amplitudes

$$r^{(1)}(\omega_j) = -\frac{q}{m} \frac{E_j}{\omega_0^2 - 2i\omega_j\gamma - \omega_j^2},\tag{A.5}$$

which is identical to the solution of the linear oscillator (compare Eq. (3.4)).

Squaring Eq. (A.4) (as necessary for applying it to Eq. (A.3b)) yields the frequencies $\pm 2\omega_1, \pm 2\omega_2, \pm (\omega_1 + \omega_2), \pm (\omega_1 - \omega_2)$ and 0. Consequently, Eq. (A.3b) needs to be solved for all frequencies that are appearing. For $2\omega_1$, for example,

$$\frac{\mathrm{d}^{2}\tilde{r}^{(2)}}{\mathrm{d}t^{2}} + 2\gamma \frac{\mathrm{d}\tilde{r}^{(2)}}{\mathrm{d}t} + \omega_{0}^{2}\tilde{r}^{(2)} = \frac{-a_{2}q^{2}E_{1}^{2}\exp\left(-i2\omega_{1}t\right)}{m^{2}\left(\omega_{0}^{2} - 2i\omega_{1}\gamma - \omega_{1}^{2}\right)^{2}}$$
(A.6)

needs to be solved. Applying $\tilde{r}^{(2)}(t) = r^{(2)}(2\omega_1) \exp(-i2\omega_1 t)$ to Eq. (A.6) gives

$$r^{(2)}(2\omega_1) = \frac{-a_2 q^2 E_1^2}{m^2 D(2\omega_1) D^2(\omega_1)}$$
(A.7)

with $D(\omega_j) = \omega_0^2 - 2i\omega_j\gamma - \omega_j^2$. In an analogous manner the remaining frequency contributions and also Eq. (A.3c) can be solved.

Appendix A: Anharmonic Oscillator Solution

For obtaining the susceptibilities it is taken advantage of the relation $P(\omega_j) = -Ner(\omega_j)$ and the power series expansion of the polarization density (see Eq. (3.2)) that gives for the two considered frequencies¹ or orders, respectively

$$P^{(1)}(\omega_1) = \varepsilon_0 \chi^{(1)}(\omega_1) E(\omega_1), \qquad (A.8a)$$

$$P^{(2)}(2\omega_1) = \varepsilon_0 \chi^{(2)}(2\omega_1; \omega_1, \omega_1) E^2(\omega_1).$$
 (A.8b)

Solving for the susceptibilities the presented approach gives

$$\chi^{(1)}(\omega_1) = \frac{Nq^2}{\varepsilon_0 m D(\omega_1)} \tag{A.9a}$$

$$\chi^{(2)}(2\omega_1, \omega_1, \omega_1) = \frac{Nq^3a_2}{\varepsilon_0 m^2 D(2\omega_1) D^2(\omega_1)}.$$
 (A.9b)

¹Although technically not necessary, the notation $\chi^{(2)}(2\omega_1;\omega_1,\omega_1)$ is often used to emphasize which frequency, written as first argument - here $2\omega_1$ -, is generated from which initial frequencies, later arguments - here two times ω_1 .

Appendix B

Nonlinear Schrödinger Equation

The nonlinear Schrödinger equation (NLS) is used in Section 3.2.5 to explain the existence of solitons. In this section the NLS is derived along the lines of [28, pp. 377-383] incorporating slight modifications to the variables in order to come to an appearance of the NLS as shown in Eq. (3.19), Section 3.2.5.

In analogy to Eq. (3.15) given in Section 3.2.3 the wave equation can be written as

$$\frac{\partial^2 \tilde{E}(z,t)}{\partial z^2} - \frac{1}{\varepsilon_0 c_0^2} \frac{\partial^2 \tilde{D}(z,t)}{\partial t^2} = 0.$$
(B.1)

with $\tilde{D} = \varepsilon_0 \tilde{E} + \tilde{P}$. Introducing Fourier amplitudes $E(z, \omega)$ and $D(z, \omega)$, that are connected via $D(z, \omega) = \varepsilon(\omega) E(z, \omega)$, and $k^2(\omega) = \varepsilon_r(\omega) \frac{\omega^2}{c_0^2}$ yields

$$\frac{\partial^2 E(z,\omega)}{\partial z^2} + k^2(\omega) E(z,\omega) = 0.$$
 (B.2)

With $E(z,\omega) \approx A(z,\omega-\omega_0) \exp(ik_0 z)$ and the slowly varying amplitude approximation $\left(\frac{\partial^2 A}{\partial z^2} \ll \frac{\partial A}{\partial z}\right)$ Eq. (B.2) becomes

$$2ik_0\frac{\partial A}{\partial z} + \left(k^2 - k_0^2\right)A = 0 \tag{B.3}$$

that can be reduced to

$$\frac{\partial A}{\partial z} - i \left(k - k_0\right) A = 0 \tag{B.4}$$

when taking into account that k^2 differs only slightly from k_0^2 in practice. As briefly introduced in Section 3.2.5, the propagation constant k can be expanded into a Taylor series reading

$$k = k_0 + \Delta k_{\rm NL} + k_1 \left(\omega - \omega_0\right) + \frac{1}{2} k_2 \left(\omega - \omega_0\right)^2$$
(B.5)

with $\Delta k_{\rm NL} = n_2 I \omega_0/c_0$ denoting the contribution of the nonlinear refractive index introduced in Section 3.2.2.2, $k_1 = 1/v_g(\omega_0)$ (v_g denotes the group velocity) and k_2 (a measure for group velocity dispersion (GVD)) as defined in Eq. (3.21). With this Taylor expansion of k the wave equation becomes

$$\frac{\partial A}{\partial z} - i\Delta k_{\rm NL}A - ik_1\left(\omega - \omega_0\right)A - \frac{1}{2}ik_2\left(\omega - \omega_0\right)^2A = 0, \qquad (B.6)$$

which reads after transformation into the temporal domain

$$\frac{\partial \tilde{A}}{\partial z} + k_1 \frac{\partial \tilde{A}}{\partial t} + \frac{1}{2} k_2 \frac{\partial^2 \tilde{A}}{\partial t^2} - i\Delta k_{\rm NL} \tilde{A} = 0.$$
(B.7)

Introducing a co-moving frame $\tau_{\rm co} = t - z/v_g = t - k_1 z$ (with $\tilde{A}(z,t) = \tilde{A}_s(z,\tau_{\rm co})$) and $I = 2n_0\varepsilon_0c_0|\tilde{A}_s|^2$ the wave equation reads

$$\frac{\partial \tilde{A}_s}{\partial z} + \frac{1}{2}ik_2\frac{\partial^2 \tilde{A}_s}{\partial \tau_{\rm co}^2} = i\gamma_0|\tilde{A}_s|^2\tilde{A}_s \tag{B.8}$$

with $\gamma_0 = 2n_0\varepsilon_0 n_2\omega_0$. From Eq. (B.8), that is of the form of a NLS, the impacts of GVD and self-phase modulation (SPM) (compare Section 3.2.5) are quite apparent. The GVD term on the left hand side describes pulse broadening by k_2 , whereas the right hand side shows the pulse broadening impact of SPM (hidden as n_2 in the γ_0 variable). Eq. (B.8) also shows that formation of a temporal soliton (SPM balanced by GVD or

vice versa) is possible if the pulse takes the form

$$\tilde{A}_s(z,\tau) = A_s^0 \operatorname{sech}\left(\tau_{\rm co}/\tau_0\right) e^{i\kappa z} \tag{B.9}$$

with $\kappa = -k_2/2\tau_0^2$ and

$$|A_s^0|^2 = -\frac{k_2}{\gamma_0 \tau_0^2}.$$
 (B.10)

Thus, not only that k_2 and γ_0 need to have opposite signs, also the pulse duration τ_0 and amplitude A_s^0 have to match to create the fundamental soliton in above equations. If the amplitude exceeds the level for formation of a fundamental soliton, higher-order solitons might be created. For a mathematical description of these higher orders a reformulation of Eq. (B.8) is convenient, introducing the variable normalizations $\tilde{a}_s = \tilde{A}_s/|A_s^0|, \zeta = z/L_D = z|k_2|/\tau_0^2$ and $t' = \tau_{co}/\tau_0$ [139, p. 147]

$$i\frac{\partial\tilde{a}_s}{\partial\zeta} = \frac{\mathrm{sign}\,(k_2)}{2}\frac{\partial^2\tilde{a}_s}{\partial t'^2} - N_{\mathrm{sol}}^2|\tilde{a}_s|^2\tilde{a}_s,\tag{B.11}$$

with $N_{\rm sol}^2 = L_D \gamma_0 |A_s^0|^2$ denoting the soliton order number (compare Eq. (3.19)).

Appendix C

On the Negligence of Light Trapping

On first sight, light trapping might be an important contribution to the excess charge carrier generation being applied for the analysis presented in Chapter 4 (as e.g. given in Eq. (4.3) or subsequently incorporated in the continuity equations Eqs. (4.8a) and (4.15)). In this appendix it will be outlined why light trapping can be neglected when ratios of short circuit current densities ($J_{\rm SC}$) or excess carrier densities (Δn) are considered.

In 1993 Basore published a useful approach to model light trapping effects based on geometrical considerations [140]. He introduced a factor $f_{\rm abs}$ that describes the fraction of incident light absorbed in the cell, thereby contributing to the $J_{\rm SC}$. Neglecting the front surface reflection, the factor is given by

$$f_{\rm abs} = (1 - T_1) + R_{\rm b1}T_1(1 - T_2) + \frac{R_{\rm b1}R_{\rm f1}T_1T_2(1 - T_{\rm n})(1 + R_{\rm bn}T_{\rm n})}{1 - R_{\rm b1}R_{\rm fn}T_{\rm n}^2},$$
(C.1)

with ray propagation angles θ_i , back-surface reflections $R_{b,i}$, front-surface reflections $R_{f,i}$ and transmitted power T_i with *i* denoting the ray path number 1, 2 or n after (i - 1) reflections. As light trapping is only relevant for slowly absorbed wavelengths penetrating the device's back-surface and being trapped, the transmission terms T_i can be linearized by

$$T_{\rm i} = \exp\left(-\frac{\alpha W}{\cos\theta_{\rm i}}\right) \approx 1 - \frac{\alpha W}{\cos\theta_{\rm i}},$$
 (C.2)

with absorption coefficient α and thickness W ($\alpha W \ll 1$). Thus, $f_{\rm abs}$ can be reduced to

$$f_{\rm abs} \approx \alpha W \left(\frac{1}{\cos \theta_1} + \frac{R_{\rm b1}}{\cos \theta_2} + \frac{R_{\rm b1} R_{\rm f1} \left(1 + R_{\rm bn} \right)}{1 - R_{\rm bn} R_{\rm fn}} \right) = \alpha W R', \quad (C.3)$$

with R' as additional contribution to the overall photon absorption by light trapping, thereby being equivalent to an increase of radiation intensity $I \rightarrow IR'$ in e.g. Eq. (4.14). Thus, the excess carrier generation term can be written as

$$G = \left(\alpha_{\rm bb} + \beta \frac{IR'}{2}\right) \frac{IR'}{h\nu}.$$
 (C.4)

If the ratios of G for different intensities I_A and I_B are considered,

$$\frac{G_{\rm A}}{G_{\rm B}} = \frac{2\alpha_{\rm bb}I_{\rm A} + \beta I_{\rm A}^2 R'}{2\alpha_{\rm bb}I_{\rm B} + \beta I_{\rm B}^2 R'} \tag{C.5}$$

results and following cases might be distinguished regarding the relevance of R':

• $(R' \to 1)$: • $(R' \to \infty)$ or $(\beta I^2 \gg 2\alpha_{bb}I)$: • $\frac{G_A}{G_B} \approx \frac{2\alpha_{bb}I_A + \beta I_A^2}{2\alpha_{bb}I_B + \beta I_B^2}$ • $(R' \to \infty)$ or $(\beta I^2 \gg 2\alpha_{bb}I)$: $\frac{G_A}{G_B} \approx \frac{\beta I_A^2 R'}{\beta I_B^2 R'} = \frac{\beta I_A^2}{\beta I_B^2}$ • $(\beta I^2 \ll 2\alpha_{bb}I)$: $\frac{G_A}{G_B} \approx \frac{2\alpha_{bb}I_A}{2\alpha_{bb}I_B}$

The above cases demonstrate that as long as linear $(\beta I^2 \ll 2\alpha_{\rm bb}I)$ or nonlinear absorption $(\beta I^2 \gg 2\alpha_{\rm bb}I)$ are dominating the generation term or in cases of no $(R' \to 1)$ or perfect trapping $(R' \to \infty)$, the generation ratios are not affected by light trapping effects. For all other cases, light trapping cannot be readily neglected and does, in general, enhance the impact of nonlinear absorption. Appendix C: On the Negligence of Light Trapping

For judging the relative contributions of linear and nonlinear generation their ratio is taken according to

$$\frac{G_{\rm NL}}{G_{\rm L}} = \frac{\beta I^2}{2\alpha_{\rm bb}I} \tag{C.6}$$

and plotted in Fig. C.1 for various average illumination intensities $I_{\rm av}$. The material parameters for intrinsic silicon (Si) are taken from *Bristow* et al. [55] and *Green* [61]. For a straightforward estimation of peak intensity,



Figure C.1: Ratio of nonlinear (βI^2) and linear $(2\alpha_{\rm bb}I)$ carrier generation in silicon (Si) versus normalized photon energy (with $E_{\rm g} = 1.12 \text{ eV}$) at various average illumination intensities $I_{\rm av}$. The grayish region denotes ranges with similar contributions of linear and nonlinear absorption.

vital for a sound estimation of nonlinear absorption, Gaussian pulses of 100 fs FWHM pulse duration ($\tau_{\rm FWHM}$) emitted at 80 MHz pulse repetition rate ($f_{\rm rep}$) are converted into a rectangular shape with equivalent pulse duration $\tau_{1/e^2} \approx 2 \cdot 0.85 \cdot \tau_{\rm FWHM}$ and peak intensity

$$I_{\rm peak} = \frac{I_{\rm av}}{f_{\rm rep}\tau_{1/e^2}}.$$
 (C.7)

The gray shaded region denotes the range of the $\beta I^2/2\alpha I$ ratio in the vicinity of unity, pointing out the spectral ranges for which neither linear

nor nonlinear absorption can be neglected. Although the spectra transit the grayish region, the range itself is comparably small (around 100 nm). Thus, given that a significantly broader spectral region is considered in the experiments or other spectral ranges than those in the grayish region are chosen, light trapping can be neglected in a first approximation. This approximation holds for the work presented in Chapter 4, as broad wavelength spectra rather than specific wavelengths requiring consideration of light trapping are investigated.

Appendix D

Green's Function Approach for Solving the Continuity Equation

A continuity equation that takes a form

$$\frac{\partial \Delta n\left(z,t\right)}{\partial t} = D_{\rm a} \frac{\partial^2 \Delta n\left(z,t\right)}{\partial z^2} + G\left(z,t\right) - \frac{\Delta n\left(z,t\right)}{\tau} \tag{D.1}$$

as given in Eq. (4.17) can be solved by Green's functions of the Heat Equation. In this appendix the Green's function approach will be briefly sketched along the lines of [20, pp. 56-60] based on a solution provided in [141].

With $\Delta n' = \exp(t/\tau) \Delta n$, Eq. (D.1) can be transformed into a Heat Equation

$$\frac{\partial \Delta n'(z,t)}{\partial t} - D_{\rm a} \frac{\partial^2 \Delta n'(z,t)}{\partial z^2} = G(z,t) \exp(t/\tau).$$
 (D.2)

With boundary conditions¹

$$D_{\rm a} \frac{\partial \Delta n'}{\partial z} \bigg|_{z=0} = S_0 \Delta n'(z=0) \tag{D.3a}$$

$$D_{\rm a} \frac{\partial \Delta n'}{\partial z} \Big|_{z=W} = -S_W \Delta n'(z=W),$$
 (D.3b)

describing the recombination at front (z = 0) and back surface (z = W)of a semiconductor with thickness W and surface recombination velocities S_0 and S_W , the excess carrier density for an initial value $\Delta n'(z, t = 0) = 0$ is given by

$$\Delta n\left(z,t\right) = \int_{0}^{t} \mathrm{d}t' \int_{0}^{W} \mathrm{d}\zeta \Gamma\left(z,\zeta,t-t'\right) G\left(\zeta,t'\right) \exp\left(-\frac{t-t'}{\tau}\right), \quad (\mathrm{D.4})$$

with carrier generation term $G(\zeta, t')$ (as e.g. given by Eq. (4.3) or any subsequent simplifications) and Green's function

$$\Gamma(z,\zeta,t) = \sum_{n=1}^{\infty} \frac{y_n(z)y_n(\zeta)}{\|y_n\|^2} \exp\left(-\lambda_n^2 D_{\mathbf{a}}t\right).$$
(D.5)

The exponential function in Eq. (D.5) represents the decay term of the *n*-th mode of the Green's function with lifetime $1/(\lambda_n^2 D_a)$, where λ_n is given by the positive solutions of the transcendental equation²

$$\frac{\tan\left(\lambda_n W\right)}{\lambda_n} = \frac{D_{\rm a}\left(S_0 + S_W\right)}{D_{\rm a}^2 \lambda_n^2 - S_0 S_W}.$$
(D.6)

As the considerations in Chapter 4 are restricted to short circuit conditions, thus, $S_0 \to \infty$ and $S_W \to 0$ for front junction solar cells, λ_n can be approximated by

$$\lambda_n \approx (2n-1) \,\frac{\pi}{2W}.\tag{D.7}$$

¹These boundary conditions describe excess charge carrier flow through the interfaces, being used for simulation of current extraction under short circuit conditions.

²As the lifetime reduces significantly for higher order modes n of the Green's function, a restriction of the sum in Eq. (D.5) to low order modes might be a valid and useful approximation to increase speed of numerical approaches.

which supersedes the transcendental Eq. (D.6) and simplifies computational approaches. Finally, the remaining parameters of Eq. (D.4) are given by

$$y_n(z) = \cos \lambda_n z + \frac{S_0}{D_a \lambda_n} \sin \lambda_n z$$
 (D.8)

and

$$\|y_n\|^2 = \frac{S_W}{2D_a\lambda_n^2} \frac{D_a^2\lambda_n^2 + S_0^2}{D_a^2\lambda_n^2 + S_W^2} + \frac{S_0}{2D_a\lambda_n^2} + \frac{W}{2} \left(1 + \left(\frac{S_0}{D_a\lambda_n}\right)^2\right). \quad (D.9)$$

The Green's function approach solves the bottleneck of a finite element approach associated with the strong surface recombination, simulating short circuit conditions, by expressing the diffusion process analytically. Further, only times t' need to be considered that effectively introduce an inhomogeneity, represented as $G(z,t) \exp(t/\tau)$ in Eq. (D.2). In case of ultrashort laser pulses, that are followed by a comparably long time (duty cycle ~10⁻⁵) of virtually zero signal (thus, with virtually G = 0), the computational effort is significantly reduced as compared to finite element approaches. Such properties of a Green's function approach suggest its superiority for a computational analysis of the interaction between ultrashort laser pulses and solar cells compared to a finite element approach.

Appendix E

Monte Carlo Method for Measurement Uncertainty

When discussing measurement uncertainties, one major topic arises from the question of how the uncertainty of an input value affects the uncertainty of the measurement outcome. Or, in other words, how is any input uncertainty propagated through the system.

In the framework of GUM ("Guide to the expression of uncertainty in measurement") [117] the measurement is modeled and standard uncertainties u_i are attributed to the input variables from their assumed or measured probability density functions (PDFs). From these standard uncertainties (and their correlations) the combined standard measurement uncertainty u_{tot} of the output variable is determined by the law of propagation of uncertainty [117, p. 19, 5.2.1]. Finally, the expanded uncertainty U_{tot} for a the desired coverage probability is calculated from the effective degree of freedom computed by the Welch-Satterthwaite formula [117, p. 73, G.4.1] and the corresponding coverage factor. For the GUM framework being valid, several requirements need to be fulfilled as e.g. that higher-order nonlinear contributions can be modeled by normal PDFs or that the PDF of the output is approximated by a normal or scaled and shifted t-distribution.

An alternative approach for propagating uncertainties through a measurement model is provided by the Monte Carlo (MC) method, that is going to be outlined in this appendix along the lines of [118]. In contrast to the GUM framework, the MC method is valid for a much broader class of problems, however, it does not yield *exact* uncertainties as provided by GUM in its range of validity.

The workflow of the MC method is schematically shown in Fig. E.1. Firstly, for each uncertainty u_i , M random numbers are generated that resemble the PDF of the input variable. As this is done for all i = 1, 2, ..., kuncertainty contributions, M times k random numbers are generated in total. If there is no wavelength-correlation for a given uncertainty,¹ a new set of random numbers is generated and applied to the next wavelength. In contrast, if there is a wavelength-correlation² the same set of random numbers is applied to the next wavelength. Applying this generation of random numbers for the different wavelengths λ_n (denoted by different colors in Fig. E.1), according to previously defined relations (see e.g. Section 6.4.1.1 for typical spectral dependencies of uncertainties), yields a 3D-matrix of k times M times N elements.

Once the random matrix has been generated, the measurement model $y_{m,n} = f(x_i)$ can be applied column-wise to retrieve the output values $y_{m,n}^3$ (bottom reddish block in Fig. E.1). So e.g. $y_{1,1}$, the output value for wavelength λ_1 and Monte Carlo draw m = 1, is achieved by solving the model equation $y_{1,1} = f(x_{1,1,1}, x_{2,1,1}, \dots, x_{k,1,1})$ (left column in blue box). Doing this likewise for the remaining Monte Carlo draws at wavelength λ_1 yields the output values $y_{2,1}$ to $y_{M,1}$ (left to right in blue box). The entity of output values $y_{1,1}$ to $y_{M,1}$, denoted by $y(\lambda_1)$, yields the PDF of $y(\lambda_1)$ as e.g. represented by the histogram shown in Fig. E.2a for $\lambda_n = 600$ nm.

¹An intuitive example for this case is given by the wavelength repeatability discussed in Section 6.4.1.1. When the center wavelength λ_0 is changed, any *actual* wavelength within $\lambda = \lambda_0 \pm \Delta \lambda_{01}$ is achieved with a random deviation $\lambda - \lambda_0$ that is not correlated to the random deviations at other center wavelengths.

²An intuitive example for this case is e.g. the uncertainty in positioning the test cell u_{pos} discussed in Section 6.4.1.2. As the test cell is not moved when changing the center wavelength and, additionally, the irradiation distribution is assumed to be invariant with respect to wavelengths, the uncertainties in positioning u_{pos} are correlated for all wavelengths.

³Please note that the index *i* is dropped owing to the dimensional reduction going along with the computation of $y_{m,n}$. This is done again when dropping the wavelength or Monte Carlo draw dimension as visualized in Fig. E.1.



Figure E.1: Schematic illustration of Monte Carlo method for determination of both spectrally resolved standard measurement uncertainties $u_{\text{tot}}(\lambda)$ and standard uncertainty in short circuit current (I_{sc}) measurement u_{Isc} from input uncertainties u_i and probability density functions (PDFs).

The corresponding standard uncertainty $u_{\text{tot}}(\lambda_1)$ can be computed from the standard deviation of $y(\lambda_1)$ according to

$$u_{\text{tot}}(\lambda_1) = \sqrt{\frac{1}{M-1} \sum_{m=1}^{M} (y_{m,1} - \bar{y}_1)^2}$$
(E.1)

with average of the output value

$$\bar{y}_1 = \frac{1}{M} \sum_{m=1}^M y_{m,1}.$$
(E.2)

In the same manner standard uncertainties u_{tot} for all wavelengths λ_n can be retrieved (very right side in Fig. E.1).

In the front-to-back direction in Fig. E.1, the entity of output values



Figure E.2: Graphical representation of Monte Carlo method outcome (data corresponds to the results presented in Section 6.4.3). (a) Histogram of $y(\lambda_n)$ for M = 100,000 and $\lambda_n = 600$ nm. (b) Normalized cumulative distribution function (CDF) obtained from the histogram shown in (a). Dashed lines indicate a symmetric coverage probability of 95.45% for determination of the expanded uncertainty $U_{\text{tot}} (\lambda_n = 600 \text{ nm}) = 0.63\%$ (see also Table 6.1).

 $y_{1,1}$ to $y_{1,N}$, denoted by $y_1(\lambda)$,⁴ can be used to compute the short circuit current $I_{\rm SC}$ associated with Monte Carlo draw m = 1 according to

$$I_{\rm SC,1} = \int y_1(\lambda) E_\lambda(\lambda) d\lambda.$$
 (E.3)

Likewise this is done for all draws m = 1, 2, ..., M and a PDF for I_{SC} is obtained yielding a standard uncertainty u_{Isc} by using equivalents of Eqs. (E.1) and (E.2).

The expanded uncertainties $U_{\text{tot}}(\lambda)$ and U_{Isc} for a given coverage probability are determined from cumulative distribution functions (CDFs). The CDFs are obtained from sorting the output quantities into a strictly increasing order (so either $y_{1,n}$ to $y_{M,n}$ for $u_{\text{tot}}(\lambda_n)$ or $I_{\text{SC},1}$ to $I_{\text{SC},M}$ for u_{Isc}) and subsequent cumulative addition of the counts from low to high output values. In Fig. E.2b the CDF for $y(\lambda_1)$ for $\lambda_n = 600$ nm is shown. The dashed blue lines indicate a symmetric coverage probability

⁴Not to be confused with $y(\lambda_1)$.



Figure E.3: (a) Correlation coefficient matrix of output quantites $y(\lambda_n)$ over wavelength λ_n obtained from Monte Carlo method. (b) Convergence of Monte Carlo method versus draw number M exemplified for standard uncertainty of short circuit current u_{Isc} . The red line indicates the expected value $u_{\text{Isc}} = 0.327\%$. The error bars are standard deviations after 50 repetitions of Monte Carlo method with M draws.

of 95.45%. As the example shown in Fig. E.2a can be approximated by a normal distribution, the expanded uncertainty U_{tot} shown in Fig. E.2b is approximately twice the standard deviation u_{tot} of the histogram plot in this case.

For the sake of completeness the correlation coefficients resulting from the Monte Carlo method are discussed as well. For the two input or output quantities $\zeta_{i,j}$ the correlation coefficient is given by

$$r_{\rm corr}\left(\zeta_{i},\zeta_{j}\right) = \frac{\sum_{m=1}^{M} \left(\zeta_{i,m} - \bar{\zeta}_{i}\right) \left(\zeta_{j,m} - \bar{\zeta}_{j}\right)}{\sqrt{\sum_{m=1}^{M} \left(\zeta_{i,m} - \bar{\zeta}_{i}\right)^{2} \sum_{m=1}^{M} \left(\zeta_{j,m} - \bar{\zeta}_{j}\right)^{2}}}$$
(E.4)

with average value $\bar{\zeta}_{i,j}$ (computed in analogy to Eq. (E.2)). As the correlation coefficients are a measure for the mutual dependence of two quantities, a correlation coefficient matrix as shown in Fig. E.3a provides useful information on the measurement itself. E.g. the mutual positive correlation of the output quantities $y(\lambda_n)$ in the spectral range from 520 to 860 nm emphasizes that any variation of the output at e.g. 600 nm is most likely accompanied by a likewise variation at all other positively correlated wavelengths.

Finally, the outcome of the Monte Carlo simulation in dependence of the draw number M is shown in Fig. E.3b. The error bars are standard deviations of the $u_{\rm Isc}$ mean value after 50 repetitions of a Monte Carlo simulation with M draws. With increasing draw number the standard deviation reduces significantly and the expected value of $u_{\rm Isc} = 0.327\%$ (red line, corresponding to the results presented in Section 6.4.4) is obtained. After 100,000 draws the *relative* standard deviation is $< 0.3\%_{\rm rel}$ (equivalent to < 0.001% absolute). For the results presented in Sections 6.4.3 and 6.4.4 M = 100,000 is applied if not stated differently.

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List of Publications

Peer-reviewed Journal Papers

- M. Mundus, J. Hohl-Ebinger, and W. Warta, Estimation of Angledependent Mode-coupling and Attenuation in Step-index Plastic Optical Fibers from Impulse Responses, Optics Express 21(14), pp. 17077 - 17088, 2013.
- M. Mundus, J. A. Giesecke, P. Fischer, J. Hohl-Ebinger, and W. Warta, *Interaction of Ultrashort Laser Pulses and Silicon Solar Cells under Short Circuit Conditions*, Journal of Applied Physics 117(8), pp. 085702-1 085702-7, 2015.

Conference Papers

- M. Mundus, J. Hohl-Ebinger, and W. Warta, Abschätzung winkelabhängiger Modenkopplung und Dämpfung in optischen Stufen-Index-Fasern anhand von Impulsantworten, in: Erstes Deutsches POF Symposium 2013, Aachen, Germany, pp. 109 - 114, 2013. ISBN: 978-3-944586-51-9.
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- M. Mundus, C. Rapp, T. Mißbach, G. Siefer, J. Hohl-Ebinger, A. W. Bett, and W. Warta, *Quantum Efficiency Measurement of*

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• M. Mundus, M. Kumar Dasa, X. Wang, J. Hohl-Ebinger, and W. Warta, Spectrally Shaped Supercontinuum for Advanced Solar Cell Characterization, in: Proceedings of the 31st European Photovoltaic Solar Energy Conference, Hamburg, Germany, pp. 514 - 519, 2015.

Oral Presentations

- M. Mundus, J. Hohl-Ebinger, and W. Warta, *Abschätzung winke*labhängiger Modenkopplung und Dämpfung in optischen Stufen-Index-Fasern anhand von Impulsantworten, Erstes Deutsches POF Symposium 2013, Aachen, Germany, 2013.
- M. Mundus, C. Rapp, T. Mißbach, G. Siefer, J. Hohl-Ebinger, A. W. Bett, and W. Warta, *Quantum Efficiency Measurement of Concentrator Photovoltaic Modules*, 42nd IEEE Photovoltaic Specialist Conference, New Orleans, Louisiana, USA, 2015.
- M. Mundus, M. Kumar Dasa, X. Wang, J. Hohl-Ebinger, and W. Warta, *Spectrally Shaped Supercontinuum for Advanced Solar Cell Characterization*, 31st European Photovoltaic Solar Energy Conference, Hamburg, Germany, 2015.
- M. Mundus, Ultrakurze Laserpulse zur Charakterisierung und Kalibrierung von Solarzellen, Colloquium Optical Technologies, University of Applied Sciences Münster, Steinfurt, Germany, 2015.

Thesis Supervisions

• D. Lill, Automatisierter Prismenmonochromator für ein durchstimmbares Ultrakurzpuls-Lasersystem, Bachelor thesis, Albert-Ludwigs-Universität Freiburg, Freiburg, Germany, 2014.

- F. A. R. Peter, Strahlungsfeld-Modellierung von Dauerlicht-Sonnensimulatoren mittels Ray-Tracing zur Evaluierung messplatzspezifischer Messunsicherheiten, Diploma thesis, Albert-Ludwigs-Universität Freiburg, Freiburg, Germany, 2014.
- X. Wang, An Optical Setup based on Spectrally Shapeable Supercontinuum Radiation for Diverse Applications, Master thesis, Karlsruhe Institute of Technology (KIT), Karlsruhe, Germany, 2016.

Publications not related to this thesis

- M. Mundus, Frequenzverdopplung eines cw-Faserlasers in Kaliumtitanylphosphat (KTP), Bachelor thesis, University of Applied Sciences Münster, Münster, Germany, 2009.
- M. Mundus, J. Gröninger, and K. Dickmann, Frequency Doubling (SHG) of a 200 W cw Fiber Laser in Kaliumtitanylphosphat (KTP), in: Proceedings of ICALEO 2009, Orlando, Florida, USA, pp. 1624-1628, 2009. ISBN: 978-0-912035-59-8.
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- S. K. Reichmuth, C. E. Garza, D. Vahle, M. de Boer, M. Mundus, G. Siefer, A. W. Bett, and H. Helmers, *Transient I-V Measurement Set-Up of Photovoltaic Laser Power Converters under Monochro-matic Irradiance*, in: 32nd European Photovoltaic Solar Energy Conference, Munich, Germany, 2016.

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The ability to provide highly accurate performance evaluations of photovoltaic devices has never been more important given the recent, and anticipated, progress in photovoltaics. The lowest possible measurement uncertainties are required for reliably assessing technological advances and reducing investment uncertainty. As the further reduction of these uncertainties within conventional solar cell measurements is often hindered by the measurement setups themselves, innovative approaches in the development of new measurement facilities are vital.

This thesis addresses such demand by applying ultrashort laser pulses for highly accurate solar cell characterization. Based on a detailed investigation of pulse-solar cell interaction, a setup for spectral responsivity measurements is developed. This cutting-edge measurement setup substantially outperforms current state-of-the-art facilities in terms of measurement accuracy. Furthermore, a novel measurement approach is presented that takes advantage of spectrally shaped supercontinuum radiation. Imitating standard solar spectra with the shaped supercontinuum radiation promises a quicker and more accurate measurement of the solar cell's short circuit current than is presently possible using conventional methods.

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