Compositionally tuned Nd:$(Y_xLu_{1-x})_3Ga_5O_{12}$-laser at 935 nm for H_2O-dial systems

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ABSTRACT

For future satellite based water vapor DIAL systems efficient and rugged sources preferably around 935 nm are required. Especially for the WALES system (Water Vapour Lidar Experiment in Space) four wavelengths between 935.561 nm and 935.906 nm (vac.) have to be addressed. A promising candidate for the direct generation within this spectral range is a simple diode pumped setup based on compositionally tuned neodymium-doped mixed garnet crystals. Within the scope of this work, novel Nd:$(Y_xLu_{1-x})_3Ga_5O_{12}$-crystals (Nd:YLuGG) with different compositions ($0 \leq x \leq 1$) were investigated. Beside the characterization of some relevant crystal properties laser experiments in quasi-continuous operation, Q-switched operation and in single-longitudinal-mode operation were performed. By the R_2-Z_5-transition wavelengths between 935.3 nm and 936.6 nm (vac.) can be addressed with different compositions x. At a repetition rate of 100 Hz nearly 6 mJ were extracted in longitudinal multimode around 935.7 nm (vac.) from a Nd:$(Y_{0.58}Lu_{0.42})_3Ga_5O_{12}$-crystal. The cavity was injection seeded and stabilized with the ramp-and-fire-method to obtain single frequency radiation. At 935.7 nm more than 4.7 mJ were generated. The laser could be tuned over a range of about $\pm$ 0.22 nm in single-longitudinal-mode operation.

Keywords: Nd:YGG, Nd:YLuGG, lidar, dial, mixed garnet, water vapor, single frequency, injection seeding

1. INTRODUCTION AND MOTIVATION

Water vapor is one of the most relevant greenhouse gases but the less understood at the same time. In the field of atmospheric research data of three dimensional global water vapor distributions with high lateral resolution is needed to answer questions like global warming, the Earth's radiation budget and the hydrological circle. This can be provided by the well established LIDAR (light detection and ranging) and DIAL (differential absorption lidar) technology in spaceborne systems. Airborne water vapor DIAL-systems have been applied successfully over the last two decades [1] [2]. Today applied OPO, Raman and Ti:Sapphire technology does not accomplish the efficiency requirements of spaceborne instruments. Furthermore, the complexity of their sequential setup – usually comprising of a DPSSL as a pump laser and frequency converter steps – offers relatively low reliability and lifetime.

The requirements of the laser transmitter for spaceborne H_2O DIAL were studied within the scope of the assessment report of the WALES (Water Vapour Lidar Experiment in Space) mission [3]. The main laser requirements are listed in Table 1. Generally, in a DIAL system laser pulses of at least two different wavelengths (on-line and off-line) are required. The on-line pulse is spectrally matched to a high absorption line of the molecule to be detected while the off-line pulse is matched to a low absorption line for reference. For the spaceborne H_2O-DIAL instrument three on-line wavelengths (λ_1, λ_2, λ_3) that correspond to different absorption cross sections and the off-line wavelength λ_4 were proposed. Each one is especially adapted to a restricted altitude range of the atmosphere to cover the whole troposphere. Consequently, all of these wavelengths have to be addressed by the DIAL transmitter sequentially. Furthermore, single frequency operation with high spectral purities is required for these measurements.

We already demonstrated a Nd:YGG-based MOPA-system comprising a single-frequency oscillator and an InnoSlab-based amplifier stage that was successfully used for water-vapor profile measurements from the ground using the absorption lines at 935.4491 nm and 935.2241 nm [4]. Details about the laser transmitter are given in [5]. In single frequency operation this laser source could be tuned over a range about 0.45 nm (935.10 nm-935.55 nm). However, the required four WALES wavelengths could not be addressed with this transmitter. Spectral fine tuning of the emission spectrum within a very restricted range can be achieved by compositional tuning of the laser host material in mixed garnet crystals. For example Walsh et al. investigated neodymium-doped $Y_3(Ga_xAl_{1-x})_5O_{12}$ ($0 \leq x \leq 1$) (Nd:YGAG) as active medium, that allows for a generation between 935.3 nm (x=1) and 938.5 nm (x=0) [6]. Within the scope of this

work we firstly investigated Nd:$(Y_xLu_{1-x})_3Ga_5O_{12}$-crystals with different compositions ($0 \leq x \leq 1$) in order to address the wavelength range that is required for the WALES instrument.

This article is structured as follows. At the beginning some relevant crystal properties like the wavelength of the R_2-Z_5-transition, the change of refraction index with the temperature and the fluorescence lifetime of the new material Nd:YLuGG are presented for different compositions x. Subsequently, the results of laser experiments with Nd:YLuGG as active material in free-running Q-switched operation (QSW), in free-running quasi-continuous-wave operation (QCW) and in pulsed single-longitudinal-mode operation (SLM) are demonstrated.

Table 1: Requirements for the laser transmitter for the WALES instrument [3].

Parameter	Value
Emission wavelength (vac.)	$\lambda_1 = 935.685$ nm
	$\lambda_2 = 935.561$ nm
	$\lambda_3 = 935.906$ nm
	$\lambda_4 = 935.852$ nm
Pulse energy	≥ 72 mJ
Repetition frequency per wavelength	≥ 25 Hz
Frequency stability	< 60 MHz
Laser linewidth	< 160 MHz
Laser spectral purity (percentage within 1 GHz)	> 99.9 %
Raw data altitude resolution	< 50 m (limits the maximum pulse duration: ~200 ns [7])
Beam propagation factor M^2	< 3

2. CRYSTAL PROPERTIES OF ND:YLUGG

Spectroscopic data of the fluorescence light emitted from the R_2-Z_5-transition in pure Nd:$Y_3Ga_5O_{12}$ and Nd:$Lu_3Ga_5O_{12}$ – crystals that were published in [8] suggest that all WALES-wavelengths can be addressed in certain Nd:$(Y_xLu_{1-x})_3Ga_5O_{12}$-crystals with an adapted composition x. To study the crystal properties versus its composition 5 different crystals with a Lutetium proportion (1-x)= 0.00, 0.30, 0.42, 0.50, 1.00 were analyzed. Beside the center wavelength of the R_2-Z_5-transition, the change of refraction index with the temperature and the fluorescence lifetime were measured.

2.1 Wavelength of the R_2-Z_5-Transition

For spectral fluorescence measurements rod-shaped samples with a diameter of 3 mm and a length of 10 mm were used. The samples were pumped with a fiber coupled diode laser emitting at 806 nm. All experiments were done in normal lab environment at about 300 K temperature. To ensure high absolute spectral accuracy the spectrometer was calibrated with a Krypton-Argon lamp to vacuum wavelength. As the wavelengths of the laser transmitter have to be adapted to the absorption lines in water vapor a high absolute spectral accuracy is indispensable.

To determine the center wavelength of the R_2-Z_5 transition, a Lorentz function was fitted to the appropriate emission peak in the measured spectrum. The center wavelengths were derived from the peak center of the Lorentz fit (see Figure 1). The wavelength of the R_2-Z_5-transition increases approximately linearly with the lutetium proportion. The gain maximum of the Nd:$(Y_{0.58}Lu_{0.42})_3Ga_5O_{12}$-crystal is spectrally located between λ_1 and λ_4 and is therefore a promising candidate to address the four required WALES wavelengths.
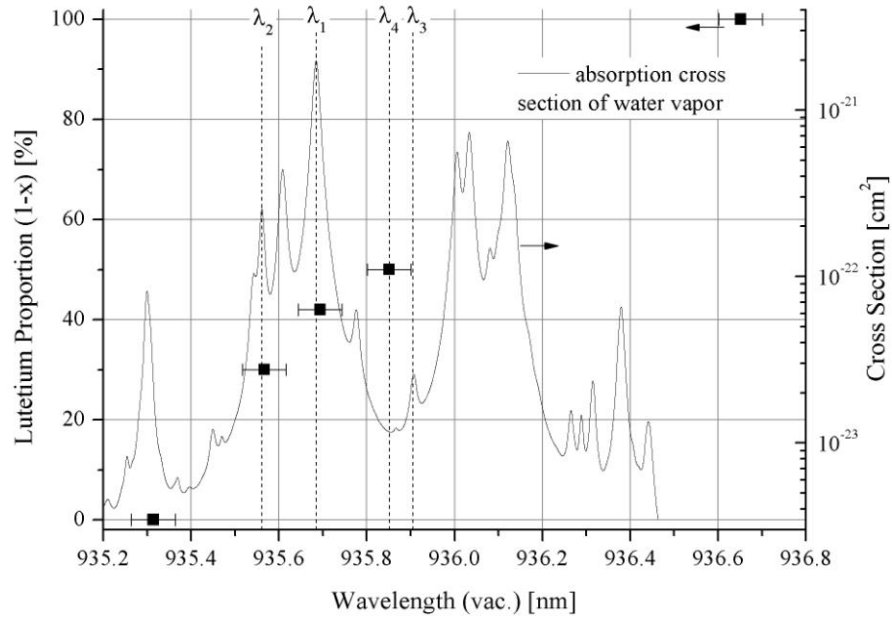
Figure 1: Central wavelengths of the R_2-Z_5 transition for different crystal compositions. These wavelengths were determined by a Lorentz-fit of the appropriate fluorescence spectrum. Overlaid are the water-vapor absorption cross section (from HITRAN 2009 [9]) and the proposed WALES wavelengths λ_1 to λ_4. The wavelength of the R_2-Z_5 transition increases approximately linearly in a range of 935.3 nm and 936.6 nm (vac.) as with the lutetium proportion. In this area the four required WALES wavelengths λ_1 to λ_4 are covered.

2.2 dn/dT-measurements

Generally, the strength of the thermal lens is an important parameter that has to be considered in laser design. The main contribution to the thermal lens is usually given by the change of the refractive index with temperature dn/dT. While this parameter is well known for common laser host materials like YAG no values are available for the YLuGG-host material with different compositions. Therefore, this parameter was measured within the scope of this work. The appropriate experimental setup is shown in Figure 2.

Prisms with an apex angle A of about 30° between the two polished surfaces were contacted with the top and bottom surfaces with indium to a copper heat sink which could be heated. Beside five Nd:YLuGG samples a Nd:YAG sample was analyzed for reference, too.
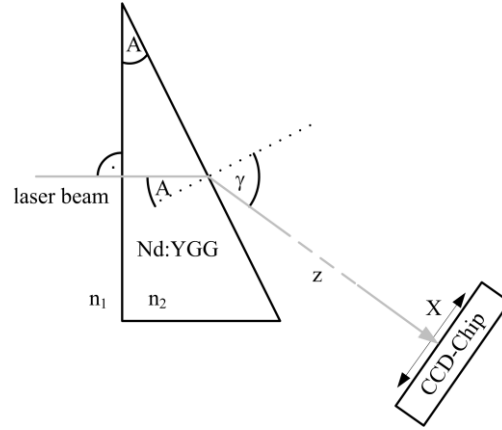
Figure 2: Experimental setup for dn/dT measurements. With a CCD camera the variation of refraction angle was measured. For more details see [5].
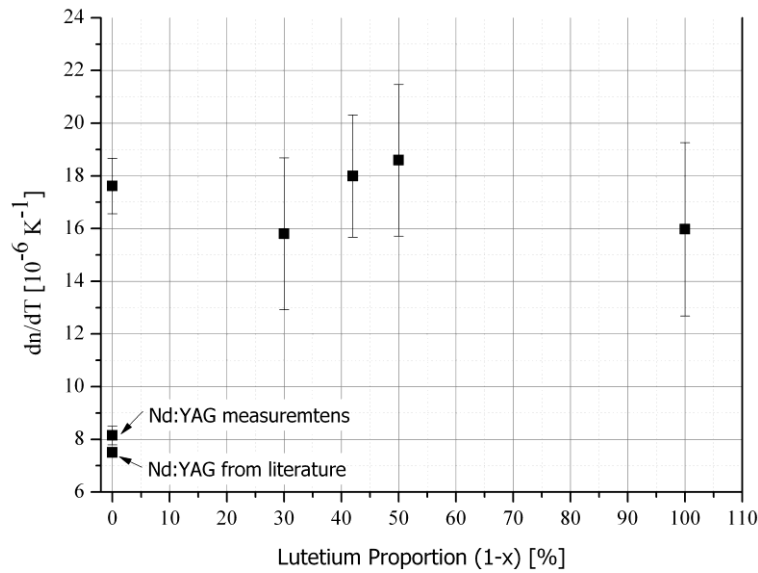

Figure 3: Measurements of the change of the refractive index with temperature for a temperature range of 22°C to 130°C of Nd:YLuGG-crystals with different compositions x as well as for Nd:YAG. The latter crystal was measured for reference. The measured value of Nd:YAG coincides well with a value that was published in [10].

A laser beam with a wavelength of 935 nm was applied perpendicularly to the entrance surface of the prisms. The beam was refracted by the exit surface of the prisms at an angle γ as function of the crystal temperature. The refraction is caused by the change of refraction index from the crystal material n_2 to the surrounding air n_1. With a CCD camera, the beam position (centroid of the intensity distribution) x was measured for different crystal temperatures between 22°C and 130°C. With the translation of the beam position with the temperature dx/dT, the refraction index of the samples at room temperature, the distance between the CCD camera and prism z and the apex angle A of the prism, the change of the refractive index with the temperature dn/dT was determined (see

Figure 3). A detailed description of the calculation of dn/dT that was applied here together with measurements of Nd:YGG are shown in [5].

For all Nd:YLuGG-compositions, the determined values are approximately a factor of two greater than the value of Nd:YAG and are in a range of $15.8 \cdot 10^{-6}$ K^{-1} to $18.6 \cdot 10^{-6}$ K^{-1}. Considering a thermal conductivity of 9 W/m/K that was

measured for YGG [11] this leads to thermal lenses that are about a factor of 4 stronger than in Nd:YAG (see [5]). The reference measurements with Nd:YAG is in the range of the corresponding published value. The short thermal lenses in Nd:YLuGG have to be considered in cavity design.

2.2 Fluorescence Lifetime

The lifetime of the upper laser level was measured. From this, the storage efficiency can be calculated for certain pump durations. In order to determine the lifetime, the temporal behavior of the fluorescence light was detected by a photo diode and was analyzed with an oscilloscope. To determine the fluorescence lifetime, the fluorescence decay was fitted exponentially. The pump duration in all measurements was 200 μs at a repetition rate of 100 Hz. In order to avoid ASE (amplified spontaneous emission)-effects the fluorescence light is to be measured by the lowest possible pumping power. ASE usually leads to an additional depletion of the pump level and therefore to shorter decay times. For this purpose, the lifetime was measured for different pump powers in a low power regime where sufficient fluorescence signal could be obtained. The data was fitted linearly and extrapolated to a pump power of 0 W. Beside the Nd:YLuGG-samples mentioned above, samples from Nd:YGG boule material with different neodymium-doping levels were measured as well (see Figure 4).

Fluorescence lifetimes in a range of 221 μs to 273 μs were measured. Accordingly, for pump duration of 200 μs the storage efficiency is 65% to 71%. No evident correlation between the fluorescence lifetime and the lutetium proportion could be found. Furthermore, no relationship is apparent between the neodymium-doping and the fluorescence lifetime in Nd:YGG. Different crystal qualities could be one possible explanation for this.
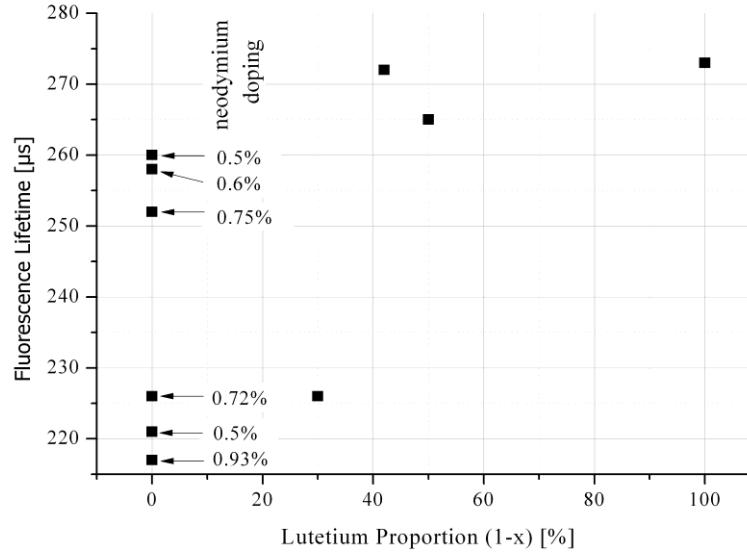

Figure 4: Fluorescence lifetime as function of lutetium proportion. A relationship between the fluorescence lifetime and the lutetium proportion is not apparent. Furthermore, the fluorescence lifetime does not depend on the neodymium-doping.

3. LASER EXPERIMENTS

Laser experiments in QCW (free-running quasi-continuous-wave operation), QSW (free-running Q-switched operation) and SLM (single-longitudinal-mode operation) were accomplished. The appropriate energetic, temporal and spectral beam properties were characterized.

3.1 Setup

In **Fehler! Verweisquelle konnte nicht gefunden werden.** Figure 5 the setup of the resonator is depicted. Here, rod-shaped laser crystals with a diameter of 3 mm and a length of 12 mm were used. For the spectrally adapted Nd:$(Y_{0.58}Lu_{0.42})_3Ga_5O_{12}$ – crystal also longer crystals with a length of 16 mm were used in order to optimize the output

energy. Both optical surfaces were polished plan parallel and had an antireflex coating for the laser wavelength at 935 nm and the pump wavelength at 806 nm. The neodymium-doping level of the samples was in a range of 0.49 at% to 0.90 at% (percentage of the yttrium ion substituted be neodymium-ion).

The crystal rods were pumped from both sides with fiber-coupled diode bars emitting at 806 nm. In all experiments the pump duration was 200 µs at a repetition rate of 100 Hz. For the cavity a planar-convex mirror configuration with a length of 250 mm was used. For Q-switching a configuration comprising a pockels cell and a thin-film polarizer (TFP) was implemented. The same resonator and pump setup was used for all crystals. The mirror tilt and the position of the focus of the pump beam were optimized for each crystal in order to obtain the maximal pulse energy.

To realize single frequency operation, the cavity was seeded with a DFB-diode laser at about 5 mW power and was stabilized by ramp and fire method. The wavelength of the seed-laser could be shifted via temperature tuning. In addition, two quarter-wave plates between the pumping mirrors and the laser crystal were added in order to achieve twisted mode operation. This prevents spatial hole burning caused by standing waves that leads to a reduction of the extraction efficiency. For more details see [5].
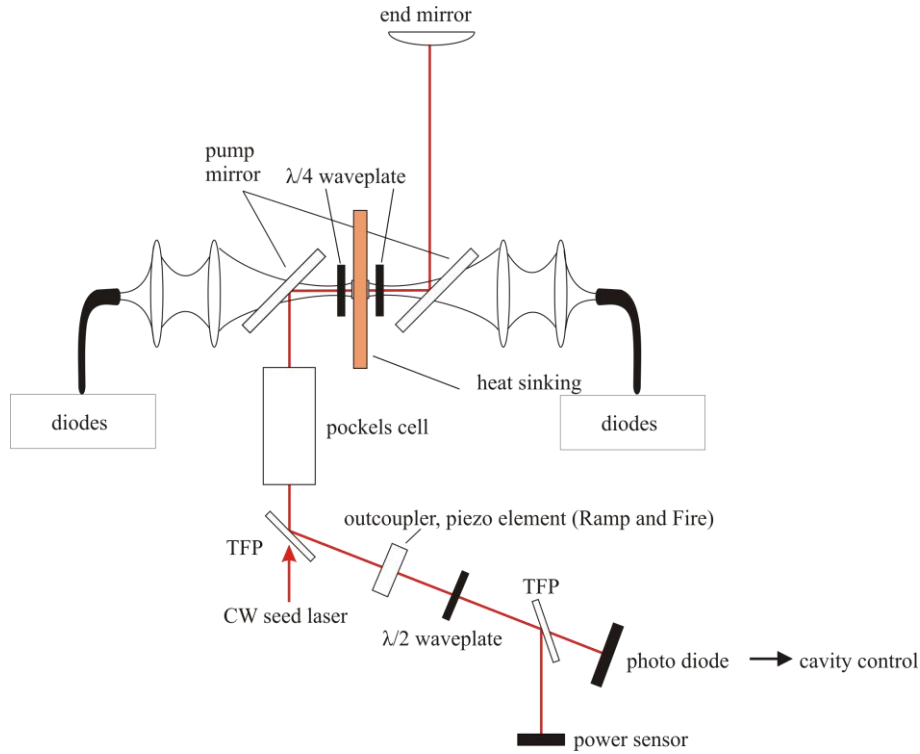

Figure 5: Schematic setup of the laser oscillator. In order to realize single frequency operation the resonator was seeded and stabilized. Twisted mode operation was realized by two quarter-wave plates.

3.2 Performance

The pulse energy in QSW-operation was measured with a thermal power meter. Pulse energies between 3.5 mJ and 5.8 mJ were obtained at pump energies between 55 mJ and 60 mJ (see Figure 6). This corresponds to an efficiency (extracted energy to pump energy) of 6% to 10%. Apart from the Nd:$(Y_{0.50}Lu_{0.50})_3Ga_5O_{12}$ – crystal, the pulse energies which have been generated in 12 mm long crystals show an increasing trend with the lutetium proportion. As this behavior is not understood, this will have to be investigated in more detail taking into account more parameters like the absorption efficiency, emission cross sections and crystal quality. With the Nd:$(Y_{0.58}Lu_{0.42})_3Ga_5O_{12}$ – crystal at a length of 16 mm, the highest energy and optical to optical efficiency of 5.8 mJ and 10%, respectively, was achieved. An excellent beam quality with a beam propagation factor of better than 1.06 was achieved in a Nd:YGG-based oscillator over a wide energy range. For details see [5]. As equal mirror and pump configurations are used together with similar

dn/dT-values it is supposed that similar beam qualities are reached for all compositions. This will have to be proven experimentally.

The emission spectrum was measured in QCW operation (see Figure 7). These measurements confirm that the wavelengths that are required for the WALES transmitter are completely covered by the Nd:YLuGG-crystals. Furthermore, the spectra of the laser based on the Nd:$(Y_{0.58}Lu_{0.42})_3Ga_5O_{12}$-crystals show an intensity dip at the strongest absorption line λ_1 which can be explained by absorption in the surrounding atmospheric water vapor. This gives a further hint that this crystal is perfectly matched to the absorption spectrum.

The temporal behavior of the laser pulses in QSW-operation was measured for different pulse energies with different crystal compositions. Pulse durations below 100 ns (fwhm) were achieved for pulse energies above 3 mJ (see

Figure 8). This oscillator easily fulfills the temporal requirements for WALES.
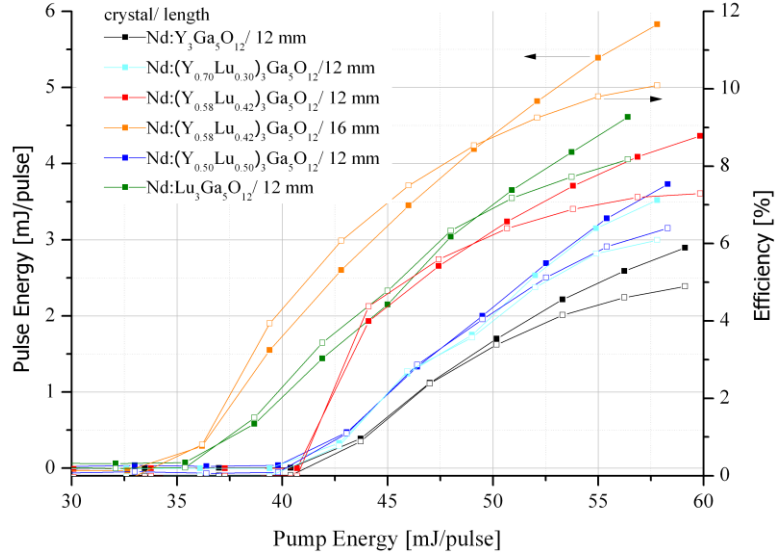

Figure 6: Measured pulse energies and efficiencies as function of the pump energy in free-running Q-switched operation.
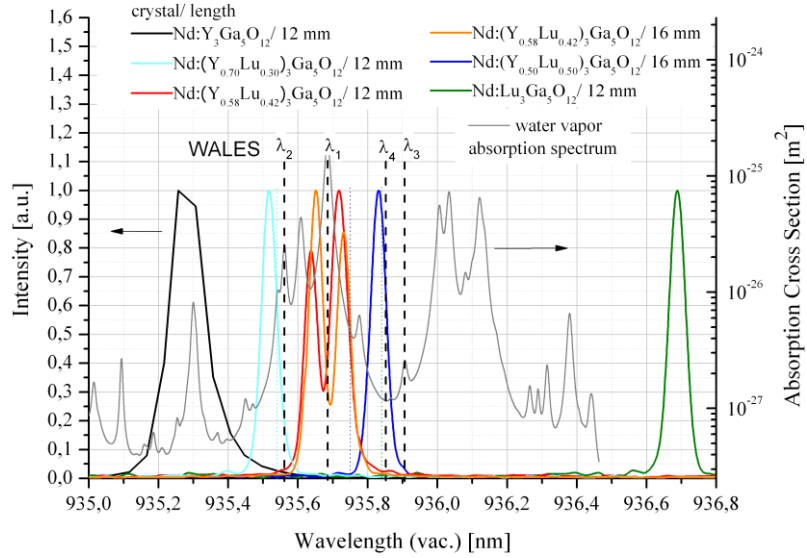
Figure 7: Spectrum of the laser in free-running QCW-operation for different crystal compositions. Overlaid is the absorption spectrum of water vapor [9] and the required WALES wavelengths [3].
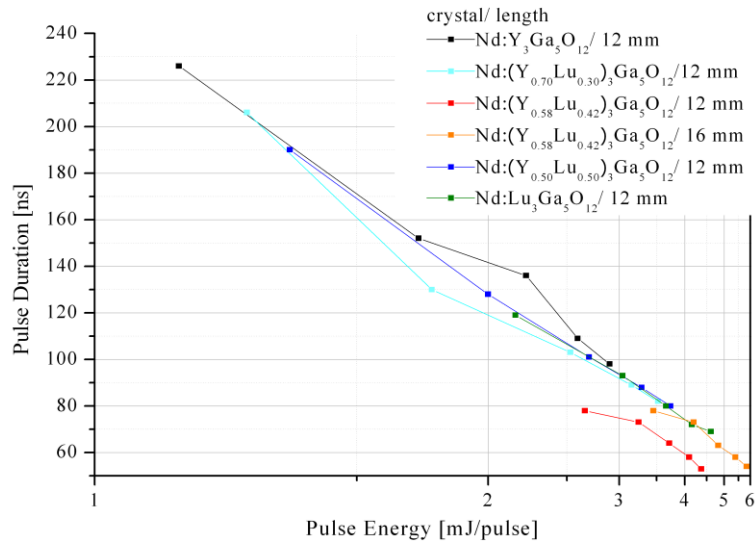

Figure 8: Measured pulse durations (fwhm) in free-running Q-switched operation as function of the pulse energy for different laser crystals.

The spectrally adapted and energetically optimized Nd:$(Y_{0.58}Lu_{0.42})_3$Ga$_5$O$_{12}$-crystal with a length of 16 mm was used to carry out experiments in single-longitudinal-mode operation (SLM). At 935.7 nm (vac.) pulse energy of 4.7 mJ at an extraction efficiency of 8% was obtained. The seed-laser could spectrally be tuned in a range of 935.4 nm to 936.0 nm resulting in temporally and energetically stable pulse generation. Seed wavelength out of this range led to an unstable behavior. This has already been observed in the Nd:YGG-based oscillator (see [4]). Even if stable SLM-operation could be shown, it is not evident whether the spectral purity requirements are fulfilled in whole spectral range. If the oscillator

is seeded in the flanks of the gain profile modes in the vicinity of the gain maximum might reduce the purity. This will have to be investigation in more detail.

4. SUMMARY AND OUTLOOK

Novel mixed garnet crystals with the stoichiometrical relation Nd:$(Y_xLu_{1-x})_3Ga_5O_{12}$ with different compositions ($0 \leq x \leq 1$) were investigated the first time with the scope to use them as active material in a laser transmitter which fulfills the stringent requirements of a spaceborne H_2O-DIAL-system. After nearly all requirements could be demonstrated in a Nd:YGG-based MOPA-system [5] and after first successful water vapor measurements with this system at 935.4491 nm and 935.2241 nm (vac.) [4] the main objective of this work was the generation of the wavelengths between 935.561 nm and 935.906 nm (vac.) that were proposed for WALES. These wavelengths could not be addressed by the Nd:YGG-MOPA.

The fluorescence spectrum was measured spectrally with high absolute accuracy in order to determine the wavelength shift of the gain peak over the crystal composition x. A spectral range of 935.3 nm to 936.6 nm (vac.) could be addressed which covers all WALES-wavelengths. The change of the refraction index with temperature and the fluorescence lifetime were measured for different compositions. As no significant change of the values with the compositions could be found it is supposed that the design of the Nd:YGG-laser [5] can easily be adapted to the new mixed garnets.

In first laser experiments even higher performance could be shown for the mixed garnets with respect to Nd:YGG. In Q-switched multi-mode operation 5.8 mJ could be generated at 100 Hz repetition rate in a 16 mm long Nd:$(Y_{0.58}Lu_{0.42})_3Ga_5O_{12}$-crystal which corresponds to an efficiency of 10% (pump light to laser light). This crystal composition is spectrally matched to the high absorption line in water vapor that was proposed for WALES. In single-longitudinal-mode operation 4.7 mJ which corresponds to an efficiency of 8% were generated. In this case two quarter-wave plates were added in order to achieve twisted mode operation. Energetically and temporally stable pulse generation for seed-wavelengths in a range of 935.4 nm and 936.0 nm (vac.) could be obtained. It is supposed that the beam quality for all compositions is close to the values that were obtained in a Nd:YGG-oscillator. Here, beam propagation factors of better than 1.06 were demonstrated [5].

Together with the successful demonstration of the Nd:YGG-based MOPA that was tested in water vapor measurements, these results show much promise that all the requirements of a laser transmitter for WALES can be fulfilled with a laser source based on Nd:$(Y_xLu_{1-x})_3Ga_5O_{12}$. As these novel crystals show higher performance in first laser experiments than the Nd:YGG-material it is supposed that efficient energy scaling can be done in InnoSlab-based amplifier stages (see [5]) to the required pulse energy level of 72 mJ.

5. ACKNOWLEDGEMENTS

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