The First Glued Tandem Solar Cell Using a ZnO Based Adhesive

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***Abstract*—By adjusting the process flow of a newly developed transparent conductive adhesive (TCA), the first mechanically stable interconnection of a glued III-V on Si tandem solar cell was established utilizing a ZnO-based adhesive. The measured V_{OC} of 1691 mV is a first proof of concept for the developed TCA. The functionality of the two sub-cells in the tandem device was proven by external quantum efficiency (EQE) measurements. The reflectance at the bond interface of such a glued tandem solar cell remains the limiting factor. In order to reduce the optical losses, the effect of a textured silicon surface was investigated in test structures, which indeed showed reduced reflectance. Furthermore, this textured test structure showed an improved connecting resistivity as low as 83 mΩcm².**

Keywords—tandem solar cell, III-V on Si, TCA

I. Introduction

Multi-junction solar cells allow for the highest photo conversion efficiencies of all solar cells. Currently up to 39.2% using III-V compound semiconductor materials alone [1] were reported and 34.1% for a combination of silicon with III-V materials in two-terminal configuration. The used bonding technique (direct wafer bond) for the combination of the III-V material with the silicon bottom cell is very demanding regarding surface preparation [2]. Both surfaces of the sub-cells have to be polished (chemically and mechanically) and absolutely particle-free. These challenges can be avoided by using a transparent and conductive adhesive (TCA) instead of the direct wafer bonding. Implementing a particle-based TCA (Ag coated particles) has already shown a very promising efficiency of 26.4% (in two-terminal configuration) [3]. In this publication we will show the progress of a newly developed particle-free TCA, which does not need the addition of conductive particles but becomes conductive itself upon thermal annealing at 300°C [4]. The adhesive is a dissolved zinc precursor which upon heating decomposes to ZnO. This developed TCA already demonstrated good optical properties (absorption below 2% for an 1-2µm thick layer for wavelengths between 600 and 1200 nm) and a low connecting resistivity (1 Ωcm^2) in glass-glass and Si-Si test structures, respectively [4]. It was also found, that the reflectance at the TCA bond layer significantly affects the performance of the tandem device [5]. In order to reduce the optical losses, this publication will present results on the bonding of a textured silicon surface to a planar silicon surface and investigate the optical and the electrical effect. The developed TCA was for the first time successfully applied to III-V/Si tandem solar cells. The resulting bond and first characterization results of the tandem device are presented.

II. Experimental

In the following section, the experimental details for different bonded test structures and their characterization will be explained. The bond interfaces of all sample structures are sputtered with an indium tin oxide (ITO) contact layer (100 nm), as this significantly reduces the connecting resistivity down to 0.12 Ωcm² (to be published in [6]). The composition of the sputter target is 95/5wt% In_2O_3/SnO_2. The TCA was deposited using a self-built spray coater. The adhesive layer consists of zinc acetylacetonate, dissolved in methanol. In order to improve the conductivity, indium is added as a dopant in the form of $InCl_3$. During the deposition of the adhesive layer, the substrate is placed on a hotplate, which is set to 125°C. After the coating, the two substrates are immediately joined. In order to achieve an electrically conductive bond, the TCA needs to thermally decompose in order to form ZnO:In [7]. If not stated otherwise, the thermal annealing of the TCA is done by placing the sample in a hot

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press applying pressures of up to 2.3 N/cm² and a maximum temperature of 300°C.

A. *Optical impact of textured silicon*

The optical benefit of a textured (random pyramid) silicon surface regarding reflectance at the bond interface was experimentally analyzed in a Si-Si test structure where one of the Si substrates was planar and one was textured. Reference samples were bonded using two planar Si substrates. A sketch of the test structures is shown in Figure 1 (a, b). These test structures are suitable since silicon and GaAs have very similar refractive indices at wavelengths above 1.2 µm, which will therefore lead to a similar optical behavior [5].

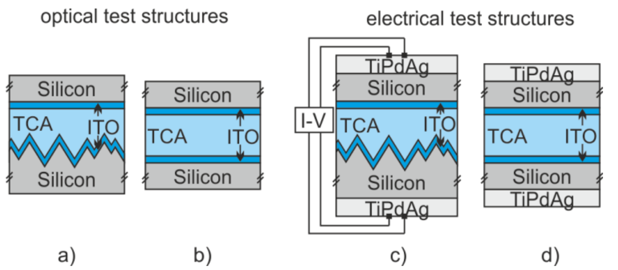

Figure 1: Sketch of the used test structures. Optical test structures are shown in a, b. Electrical test structures are shown in c, d.

The morphology of the bond layer was analyzed using scanning electron microscopy (SEM). The reflectance of the sample was measured using a PerkinElmer Lambda950 UV-Vis spectrometer.

B. *Electrical impact of textured silicon*

The electrical impact of a textured silicon surface on the connecting resistivity was investigated by bonding two sets of Si-Si samples: one with planar surfaces and one with one of the bond interfaces having a random pyramid texture (further denoted as „planar – textured bond"). In order to characterize the samples after bonding, the samples were metallized on both sides by evaporating TiPdAg and cut into 5x5 mm² squares. A sketch of the two sample structures is shown in Figure 1 (c,d). The samples were analyzed for their resistance by measuring their I-V characteristics using a Keithley 2450 SourceMeter.

C. *Bonding of the first tandem solar cell*

The first bonded tandem solar cell consists of a p-type silicon bottom wafer (1 Ωcm^2, 4" FZ) including n- and p-type tunnel oxide passivated contacts (TOPCon, [8]) on the front and back side, respectively. The aluminum gallium arsenide (nominally $Al_{0.21}Ga_{0.79}As$) top-cell is epitaxially grown on a GaAs substrate. The bond interfaces in the tandem solar cell were also coated with 100 nm sputtered ITO as an electrical contact layer. A simplified sketch of the glued cell structure is shown in Figure 2.

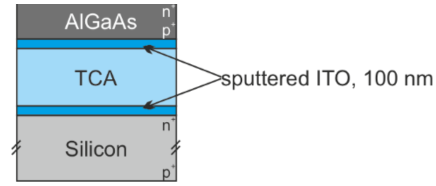

Figure 2: Simplified sketch of the glued tandem structure.

However, the developed bonding process flow as described in [4] did not result in mechanically stable bonds. Only by implementing an initial pressing at a low temperature (125°C, 0.33 N/cm²), a mechanically stable interconnection of a III-V/Si tandem solar cell (both semiconductor surfaces at the bond planar) was realized. After the initial bonding, the GaAs substrate on which the III-V solar cell was grown on, was removed by etching and a simplified metal grid structure was realized by electroplating, allowing for a first characterization of the glued device's with external quantum efficiency (EQE) measurements. The homogeneity of the bond layer was analyzed by scanning acoustic microscopy (SAM) using a PVA TePla WINSAM 8 equipped with a 100 MHz transducer. A second glued tandem cell was prepared in order to test whether the sample could withstand a second hot pressing at higher temperatures after removal of the GaAs substrate. In order to characterize the possible delamination, a transmission photograph was taken while illuminating the sample from the back with a 940nm LED. Areas where the top cell delaminates appear bright due to less absorption, allowing for a characterization of the delaminated area.

III. Results

A. *Optical impact of textured silicon*

The impact of a textured surface on the structure of the TCA bond layer was investigated by SEM and compared to a bond of a planar-planar reference. The obtained images are shown in Figure 3. The SEM characterization shows that the developed TCA covers the texture of the bottom Si substrate without substantial voids. The average bond thickness appears to be in the same range as for the reference sample (≈ 1-2 µm). The measured reflectance spectra of the two samples are shown in Figure 4.

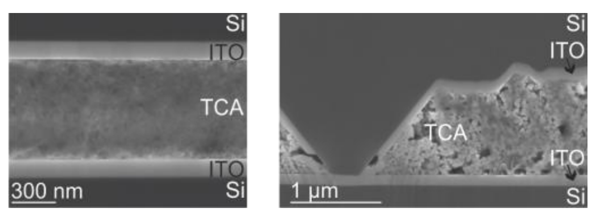

Figure 3: SEM images of the bond layer in a planar-planar reference structure and a planar-textured test structure.

The reflectance above 1200 nm (where silicon is transparent) and especially the difference in reflectance of planar and textured sample can be analyzed in order to estimate the effect of a textured silicon surface on the optics of the bond interface within the relevant wavelength range (730-1200 nm).

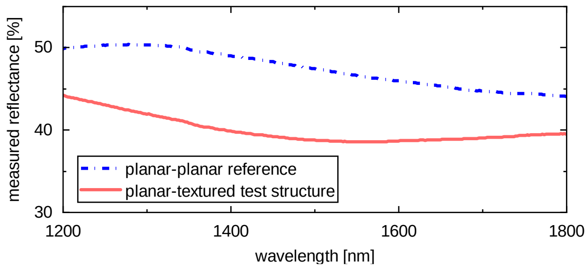

Figure 4: Measured reflectance of the reference sample and the planar-textured test structure.

The measured reflectance shows that the texturing of the bottom silicon surface did indeed reduce the reflectance by 5-10 % absolute in samples bonded with the developed ZnO-based TCA. The same reduction of reflectance at the bond interface can be assumed within the relevant wavelength range of 730 – 1200 nm.

B. Electrical impact of textured silicon

The cut squares from the bonded textured samples were measured for their I-V characteristics by sweeping the current from -20 mA/cm^2 to 20 mA/cm² and measuring the voltage drop across the sample. The I-V curves of the sample are presented in Figure 5 and compared to those of a planar-planar test structure already reported in Ref. [6].

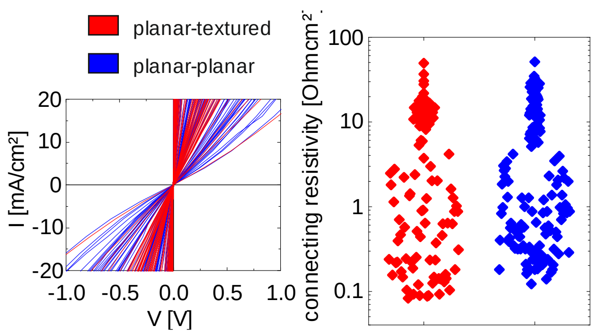

Figure 5: Left: the measured I-V characteristics of the bonded samples. Right: Obtained connecting resistivity.

Almost all samples showed ohmic behavior and therefore allowed a linear fit. The obtained connecting resistivity is plotted in Figure 5 (right). Other contributions to the connecting resistivity (metallization, specific resistivity of wafer) should be negligible. The spreading of the measured connecting resistivity can be attributed to inhomogeneities in the bond layer. Areas that show good bonding also show the lowest connecting resistivity [5]. The lowest measured connecting resistivity of the planar-planar sample was 120 mΩcm² [6]. The planar-textured test structure showed a new lowest connecting resistivity of 83 mΩcm², indicating that the texturing might also improve the connecting resistivity due to the larger contact area. For both test structures, the average connecting resistivity within a 20x20 mm^2 square (that showed homogeneous bonding in the SAM scan) was calculated. The planar sample showed an average connecting resistivity within the chosen area of 0.50 Ωcm^2 while the planar-textured sample showed an average of 0.16 Ωcm^2.

C. Bonding of the first tandem solar cell

Attempts to transfer the developed bonding process [4] directly to III-V/Si samples failed, probably due to the difference in thermal expansion of the two substrates at 300°C. Only by lowering the bonding temperature to 125°C, a first mechanically stable interconnection was achieved. The connecting resistivity of this bond will probably be higher than the connecting resistivity of the silicon-silicon test structures which were bonded at 300°C, since the thermal decomposition into ZnO:In is not complete at 125°C [7]. In Figure 6 the four main decomposition reactions of the used Zn precursor and the temperatures at which they occur are shown.

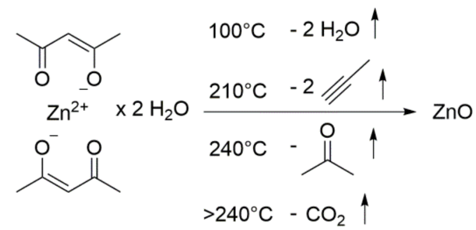

Figure 6: Thermal decomposition of the used Zn precursor into ZnO, adapted from [7].

After the initial pressing, the samples still show signs of thermally induced strain as they are not flat but bowed. A SAM scan of the first bonded solar cell is shown in Figure 7. In the SAM scan all areas that are bonded well appear dark while trapped gas or other cavities appear bright. The change in brightness from the center to the edge of the sample can be an artefact of the samples curvature. After removal of the GaAs substrate, the samples were flat again. A photo of the bonded tandem solar cell after the removal of the GaAs substrate is also shown in Figure 7.

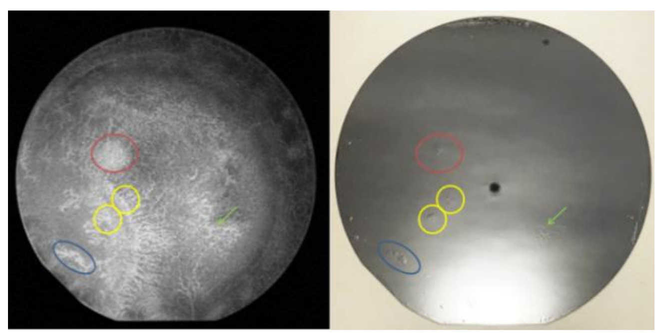

Figure 7: Left: SAM scan of the first glued tandem solar cell using the ZnO-based TCA. Right: Photo of the same tandem solar cell after the removal of the GaAs substrate. Positions of irregularities are highlighted; the dark spot in the center of the photo is the reflection of the camera lens.

The SAM scan of the bonded sample shows some cavities within the bond. However, after removal of the substrate, the thin AlGaAs top-cell remained stable over the whole wafer area, showing only very few irregularities which are highlighted in the photo in Figure 7. By comparing their positions with the SAM scan (same positions highlighted), it can be seen that the irregularities correlate to brighter spots in the SAM scan. The dark spot in the center of the photograph is the reflection of the camera lens.

The bonded sample was metallized by electrochemical plating of a simplified grid structure and characterized by measuring the I-V characteristics. The resulting V_{OC} of 1691 mV is a first promising proof of concept for the developed TCA interconnection concept on cell level. To prove the functionality of both sub-cells, an EQE measurement was performed. The resulting curve is presented in Figure 8.

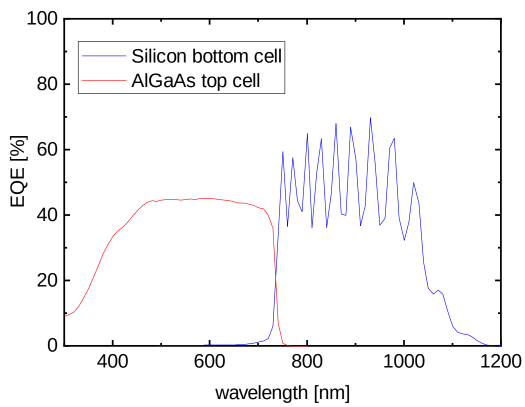

Figure 8: EQE of the glued AlGaAs/Si tandem solar cell (frontside without anti-reflection coating)..

The EQE shows, that both sub-cells are functional, which again is a first proof of concept for the developed TCA. It should be noted, that the sample was not coated with an anti-reflection coating. The resulting high reflectivity of the surface leads to the overall moderate level of the EQE. The oscillations in the bottom cell EQE are caused by interference effects at the planar bond interfaces. Due to the lowered temperature in the initial pressing step, we expect a higher connecting resistivity for the first glued tandem solar cell compared to the electrical test structures (hot-pressed at 300°C).

In order to test whether the thin III-V top-cell is stable during a second hot pressing (up to 300°C), which is necessary for a low connecting resistivity, a second bonded tandem solar cell was prepared. After the initial bonding (125°C) and the etching of the GaAs substrate, the sample was placed inside the hot press and heated up to 300° while applying up to 2.3 N/cm². In order to investigate, whether this process leads to delamination of the top cell, a transmission IR photo was taken before and after the second hot pressing, which can be seen in Figure 9. Delaminated areas appear brighter.

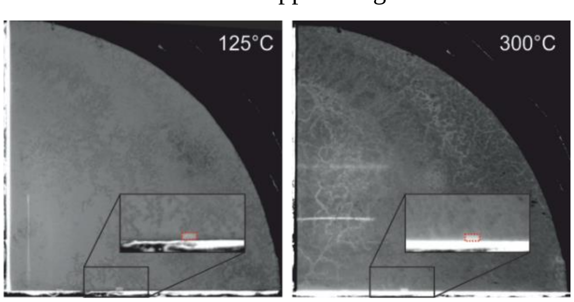

Figure 9: IR Photos of a glued tandem solar cell before (left) and after (right) the second hot pressing step. The inset shows a small delaminated area (indicated by a red frame), which was already present before the second hot pressing.

The IR photo shows, that the thin III-V top cell remained stable during the second hot pressing without additional delamination. This should allow the fabrication of a solar cell with a considerably lower connecting resistivity, since the adhesive layer was calcined at 300°C. Calculations [6] have shown that a combination of a low connecting resistivity as shown here and improved optical performance (partially solved in this paper) enable tandem solar cell efficiencies in the range of 30% and above.

IV. Summary

In the presented work, the previous developments towards a new, ZnO based TCA for connecting III-V on Si tandem solar cells are moved to a higher technology readiness level. By implementing an initial gluing at lower temperatures, a first actual cell was produced, showing high mechanical stability over the entire wafer. The characterization of the tandem solar cell shows a promising first measured V_{OC} of 1691 mV. The EQE scan showed that both sub-cells are functional and electrically interconnected. In order to achieve a lower connecting resistivity, a second hot pressing can be performed without delamination of the top cell. The back-end processes for fabrication of a tandem solar cell including an anti-reflection coating and grid metallization are currently being optimized for application in a glued tandem device.

One of the limiting parameters of the developed ZnO-based adhesive is the reflectance at the bond layer. In order to reduce the reflectance at the bond interfaces and thus the optical losses, the effect of a textured silicon surface was investigated. The characterization of a textured-planar Si-Si test structure showed that the TCA also bonds textured surfaces and the thickness of the bond layer appeared to be in the same range as for the reference. The optical characterization showed a 5-10% reduced reflectance at wavelengths > 1200 nm compared to a planar Si-Si reference, which should be transferable to the relevant wavelength range of 730-1200 nm. The electrical characterization of the textured-planar test structure showed a new lowest connecting resistivity of 83 mΩcm². Such features can be implemented in coming cell runs to improve the efficiency potential.

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