Laser structuring of thin films for flexible CIGS solar cells

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ABSTRACT

Thin-film solar cells (TFSCs) still hold unlocked potential for achieving both high efficiency and low manufacturing costs. The formation of integrated interconnects is a useful way of maintaining high efficiency in small-scale solar cells by their connection in series to form a module. Laser scribing is widely used for scribing a-Si- and CdTe-based TFSCs to form interconnects. The optical properties of the ternary copper-indium-gallium (di)selenide (CIGS) compound are well suited to the solar spectrum, with the potential to achieve a high photoelectrical efficiency. However, since it is a thermally sensitive material, new approaches for the laser-scribing process are required, to eliminate any remaining heating effects. For flexible CIGS solar cells on non-transparent substrates (metal foils or polymer), the scribing process faces additional challenges. This is one reason why ultrashort laser pulses yield better results in terms of scribing quality and selectivity. The modelling of laser energy coupling and an extensive characterization of laser scribes allow approaches to be developed for laser scribing of CIGS solar cells on flexible polymer substrates. The measured high efficiency of the resulting high-speed laser-scribed, integrated CIGS mini-modules proved the capability of this approach.

Introduction

Thin-film solar cells (TFSCs) based on copper, indium, gallium and selenium – abbreviated $Cu(In,Ga)Se_2$, or CIGS – have received increasing attention because of achievable photoelectrical conversion efficiencies as high as 27% [1], which is the highest among the different TFSC types. Experimentally verified efficiencies are 20.1% for rigid CIGS solar cells [2] and 17.1% for flexible ones on polyimide [3]. Despite the currently lower efficiency, there is a lot of interest in flexible CIGS solar cells because of the cost-saving roll-to-roll processing and the lower thermal budget for fabrication. Further advantages of flexible CIGS solar cells with relevance to applications are their excellent power-to-weight ratio, a good resistance to radiation (space application [4]) and their flexibility, allowing building integration.

Within the CIGS solar module fabrication process by monolithic integrated interconnection (MII), the efficiency is lower, currently reaching values from 12.1% to 15.9% [5]. Low-loss series interconnections therefore hold significant potential for the fabrication of high-efficiency solar modules. Among other aspects, precise and damage-free scribing of thin films for the interconnections is necessary in order to maximize the effective area and to minimize the losses due to contact resistances and shunts at the interconnections.

Industrial processes for module fabrication call for high speed, low cost, high throughput and high reliability. Scaling up the CIGS thin-film solar cell production and reducing the production costs below US$1/$W_p$ are essential requirements for the future development of the entire fabrication process of CIGS modules. Laser processing is one of the key technologies in reaching this goal [6].

To achieve the high module efficiency over large areas, the module area must be divided into small segments that are interconnected in series in order to maintain a low current while limiting the serial resistance losses within the thin films and the interconnections. The standard way of producing MIIs includes alternating steps of layer deposition and layer patterning. Structuring is accomplished by three scribing processes called P1, P2 and P3, which perform the definition of the length of the solar cell, the formation of the contact area, and the disconnection of the front contact. However, in the alternative approach of external integrated interconnection (EII), the whole film stack is fabricated first and is scribed afterwards. In consequence, the challenges are different for MII and EII.

Currently the scribing of the films is often performed by mechanical tools that provide a rather limited performance in

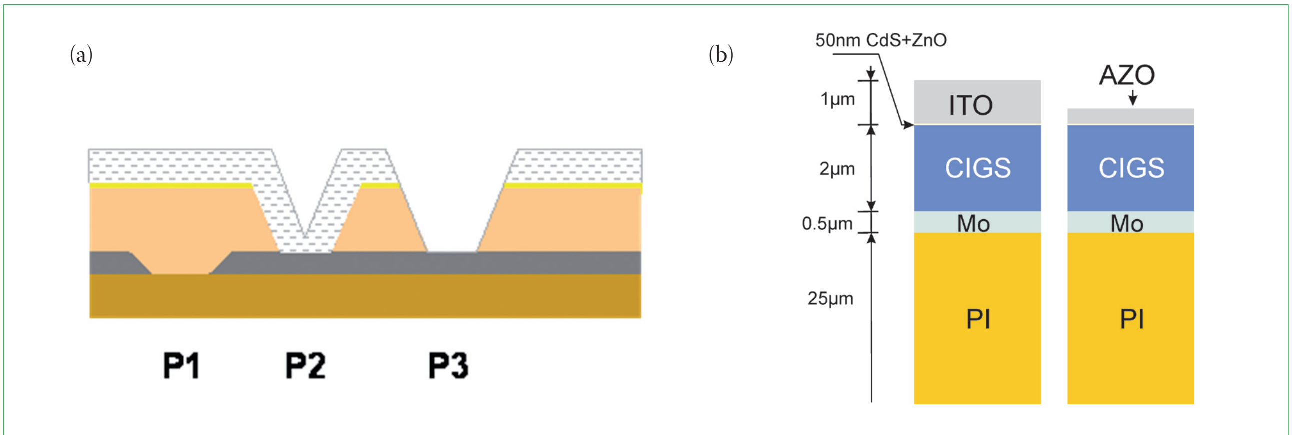

Figure 1. (a) Standard monolithic integrated interconnect (MII) scheme, with P1, P2 and P3 scribing processes performed between deposition steps. (b) Typical layer structure of a CIGS solar cell with a transparent top contact made of ITO or AZO (ZnO:Al).

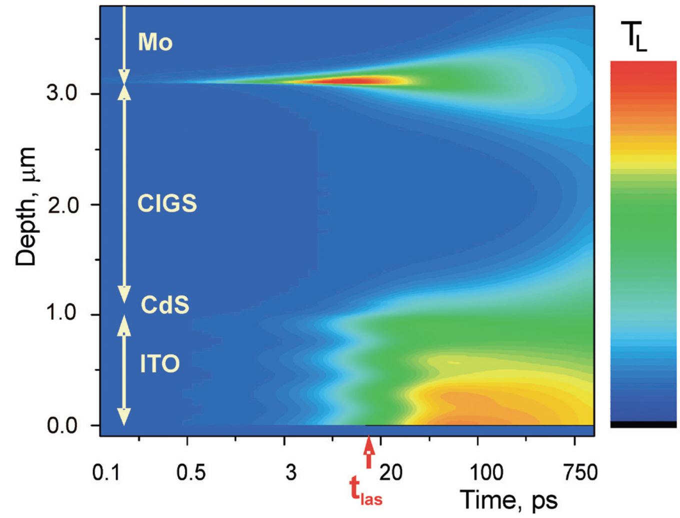

Figure 2. Spatial (bottom to top) and temporal (left to right) evolution of the lattice temperature inside the CIGS structure. A laser pulse with a duration of 10ps (indicated in red on the axis) struck the CIGS PV structure from the bottom (laser fluence = 1J/cm²; wavelength = 1064nm).

terms of the scribing width, chipping of the films, tool degradation and scribing speed. However, this technique is still in use, especially for scribing thermally sensitive CIGS films with the processes P2 and P3 [5]. Hence, the use of mechanical tools, despite their shortcomings, provides evidence that laser scribing of such thin-film stacks is still challenging for current laser technology. Different approaches for the development of laser-scribing processes in CIGS solar module fabrication by MII are known. The scribing of the molybdenum film (P1) by laser is widely accepted [7,8]. However, a nanosecond laser may produce cracks in the molybdenum and this must be avoided.

> "Only ultrashort pulsed lasers appear to be capable of patterning CIGS films without any undesirable thermal effects."

There are no established processes for the laser scribing of all the films of CIGS solar cells owing to the thermal sensitivity of the material involved. Because of this sensitivity, laser processing may lead to material modification or cause interface damage by, for instance, the phase transition of semiconducting $CuInSe_2$ to a metallic state. Nanosecond lasers have been used in trials, but only ultrashort pulsed lasers appear to be capable of patterning CIGS films without any undesirable thermal effects [9], although p-n junction damage during the scribing process, even with ultrashort laser pulses, has been reported [10]. On the other hand, the laser-induced conversion of semiconducting CIGS to a metallic phase has been utilized for the fabrication of P2 interconnects using nanosecond lasers with the so-called 'welding' process [11].

Most of the research into the use of picosecond lasers for scribing CIGS solar cells has been carried out using layers deposited on a rigid glass substrate [12,13]. For solar cells based on glass substrates, laser scribing can be performed by irradiation through the glass substrate due to the micro-explosive effect [14]. The use of molybdenum both as a back contact and as the polyimide substrate in CIGS solar cell manufacturing makes it impossible to scribe through the substrate side because of the optical characteristics of the support. The laser-scribing processes of CIGS on flexible metal [11] and polymer substrates [10,15,16] are still challenging. The development of front side laser scribing is therefore essential for the implementation of industrial module fabrication. Fig. 1(a) depicts a schematic cross section of the interconnection area of an MII; the typical layer structure of a CIGS solar cell is shown in Fig. 1(b).

The goal of this study was to develop a flexible and rapid laser technology that is compatible with a roll-to-roll production line for the precise structuring of CIGS solar cells. Modelling of laser light absorption, energy coupling and heat distribution dynamics was realized after a

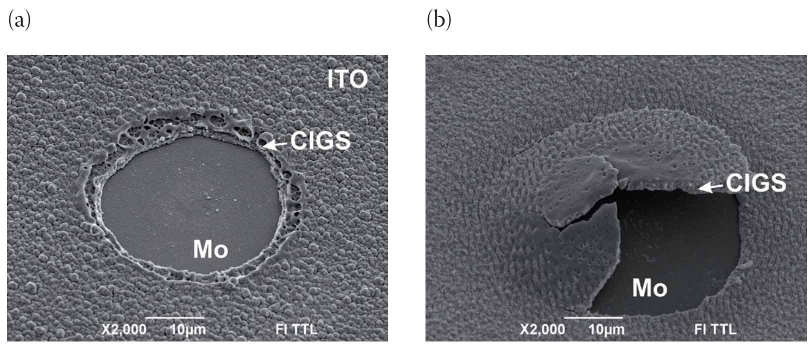

Figure 3. SEM images of craters ablated in (a) ITO/CIGS/Mo/PI and (b) ZnO/CIGS/Mo/PI solar cell structures. The effect of selectivity in energy coupling with the material removal quality is illustrated: CIGS can be cleanly removed using 1064nm in (a), but it is not fully removed using 1572nm in (b).

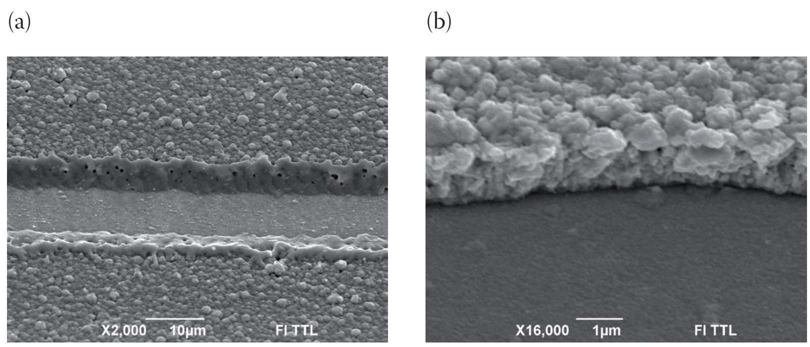

Figure 4. P3-type laser scribes in a CIGS PV structure: a single pass at 200mm/s has been performed using a wavelength of 1064nm in (a), and 1572nm in (b).

picosecond laser pulse was applied to the thin-film structure. Lasers with the picosecond and femtosecond pulse durations were used in the scribing of the CIGS TFSCs deposited on a flexible polymer substrate. An evaluation of the laser-scribe quality, an elemental analysis, and investigations of the local electrical properties of solar cells near the laser-scribing zone, together with efficiency and parallel resistance measurements, are presented for both pulse durations.

Modelling of energy coupling and laser-induced stress

Modelling of the laser-induced processes is important for interpreting the experimental results. Since different physical processes are involved, the finite-element method provided by COMSOL Multiphysics packages was utilized to simulate the process.

The quantity of laser energy coupled inside the films can be found by solving the Helmholtz beam propagation equation, which takes into account absorption and reflection of electromagnetic waves [8]. The wavelength and the complex refractive index $\bar{n}$ (given by $\bar{n} = n + ik_{ext}$, where n is the real part of the refraction index and k_{ext} is the extinction coefficient) mainly define the energy coupling from the laser-generated photons within the thin-film stack. Reflection at the thin-film interfaces causes periodical modulation of the absorbed laser energy along the beam propagation direction, especially for wavelengths where the films are partially transparent, for instance 1064nm. The selectivity of the laser-scribing process can be controlled as a result of the localization of the coupled laser energy at the inner interfaces.

By using the two-temperature model and the calculated absorption of a laser pulse, a numerical simulation of the temperature distribution in the CIGS solar cell structures was performed as a function of the wavelength of the laser radiation in order to determine the removal mechanisms of the transparent conductor and CIGS layers. Simulation results for CIGS solar cells with an ITO top contact are presented in Fig. 2.

Visible light is absorbed by the absorber layer itself. For infrared wavelengths, the high temperature at the CIGS–molybdenum interface remains for a few hundred picoseconds, while the CIGS layer itself is kept cold. This can prevent thermal degradation of the CIGS material.

Because the laser-induced stress increases in proportion to the thermal gradient, the large temperature gradients ΔT of up to 6000K can result in strain values of the order of tens of GPa within the different layers. The irradiated area of the sample experiences plastic deformations and can be fractured under laser-induced thermal strain [14]. Because localization of the strain by local absorption near the interfaces is beneficial, the infrared laser beams can be very useful for scribing thermally sensitive thin-film stacks by using spallation. The SEM images in Fig. 3, showing experimental CIGS ablation with wavelengths of 1064nm and 1572nm, present evidence of the laser-induced mechanical removal processes.

In the case of Fig. 3(a), laser pulses of 1064nm wavelength were sufficiently absorbed at the CIGS/Mo interface, and a clean exposure of the Mo layer was observed. In the case of Fig. 3(b), using a 1572nm wavelength, the laser pulse energy was not sufficient to fully remove the CIGS layer.

Scribing and laser wavelength

Scribing of the CIGS structure with the thick ITO top contact was performed using the picosecond laser (pulse duration 10ps) with different wavelengths. Scribing with a 1064nm wavelength caused clean exposure of the Mo layer (Fig. 4). This result agrees with theoretical assumptions that a large amount of the 1064nm irradiation is absorbed at the CIGS-Mo interface, enabling good layer removal selectivity. The 532nm wavelength induced the formation of a melt area at the edges of the scribe due to the high absorption of this wavelength at the CdS-CIGS interface. The CIGS material was

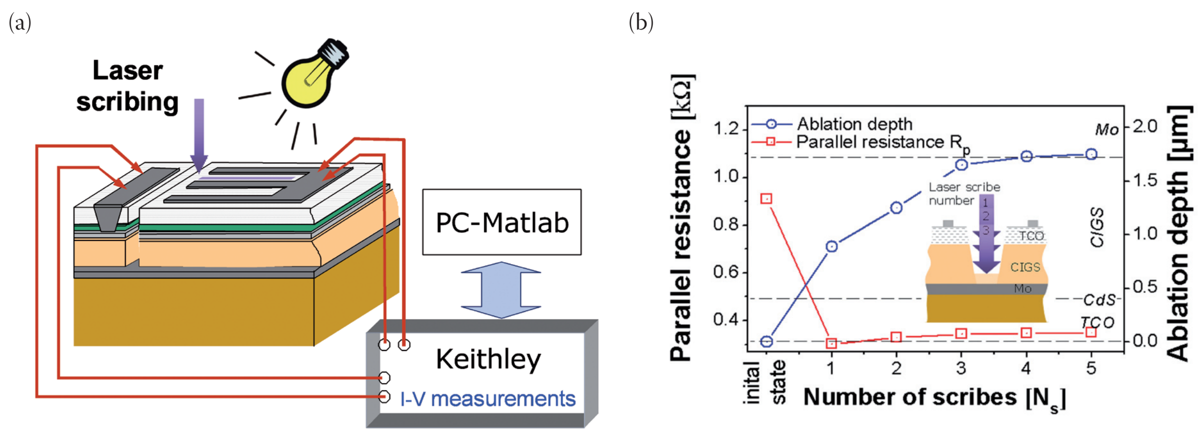

Figure 5. Quasi in situ measurements of a CIGS TFSC for the optimization of the scribing process with an ultrashort pulse laser ($t_p \approx 150$fs): (a) experimental set-up; and (b) parallel resistance during sequential laser scribing (laser fluence of 0.786J/cm² and a pulse overlap of 77%).

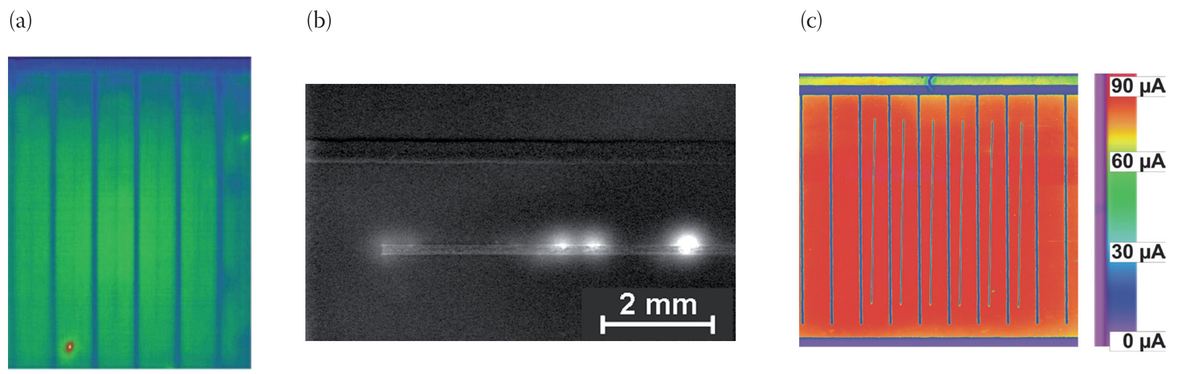

Figure 6. (a) LIT image of the laser-scribed area of a CIGS solar cell. The bright spots indicate film deposition defects, and the blue lines are the contact grid; laser scribes are located in between the vertical contact grid. (b) DLIT image of the overlapping laser scribes near a contact finger of a CIGS solar cell. (c) LBIC photocurrent map of the solar cell area after picosecond laser scribing.

removed by a direct laser ablation process, causing an increase in thermal effects. A 355nm radiation can produce high-quality scribes; however, gentle ablation with low pulse energy is required and multiple scans are necessary.

> "Scribing with a 1064nm wavelength caused clean exposure of the Mo layer."

A laser wavelength of 1064nm was found to be optimal for the P3-type scribe formation in the thin-film CIGS structure. By increasing the scanning speed to 900mm/s, it was possible to selectively remove only the ITO layer without noticeably affecting the absorber layer underneath.

Layer selectivity and depth control are crucial for the high-quality scribing of complex TFSCs. To investigate the remaining layer structure after picosecond and femtosecond laser scribing, SEM and X-ray energy dispersion spectrometer (EDS) analyses were applied [17]. EDS analysis did not detect any splashes of molten molybdenum on the edges of the scribed trench, and all the layers still had sharp interfaces. This demonstrates the potential of ultrashort lasers for high-quality selective thin-layer structuring.

Raman spectroscopy of the laser-affected area

CIGS is a thermally sensitive material, and laser scribing can induce structural changes close to the scribes. The formation of a metallic phase in the CIGS material may cause an internal shunt formation and a reduction in solar cell performance [18]. Raman spectroscopy is a sensitive tool for measuring local disorder in solid materials. Raman spectra were acquired by spatial resolution of 2µm at different locations relative to a laser scribe. Alterations in Raman spectra were obvious in the melt area (walls) as well as outside the trench, where the film was irradiated with wings of the Gaussian beam. However, those changes in spectra did not reveal any evidence of the secondary metallic phase $CuSe_x$ formation during the picosecond laser scribing, as no new lines appeared in Raman spectra close to 262cm^{-1}. These measurements confirmed the ability of picosecond lasers to scribe CIGS TFSCs with low thermally-induced structural changes of the material near the scribing zone.

Photoelectrical characterization of laser-scribed CIGS solar cells

To optimize the laser scribing of CIGS solar cells, measurements of the laser-induced modifications of the electrical characteristic are needed; ex situ and in situ characterization techniques should therefore be applied. The main objective is to evaluate the alterations in solar cell photoelectrical properties after laser scribing.

Electrical measurements of laser-scribed CIGS TFSCs

Fig 5(a) shows the experimental set-up utilized for quasi in situ measurements during the laser scribing. This allowed the rapid collection of reliable information

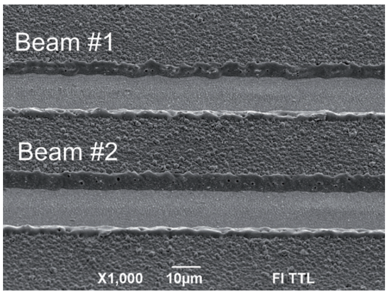

Figure 7. SEM image of parallel P3 scribes in a CIGS PV cell. Distance between them can easily be controlled by optics.

about the changes of the solar cell characteristics due to the laser scribing. Together with the capabilities of the laser-scribing workstation, the scribe geometry and the optical image of the scribe, as well as the electrical properties extracted from the *I-V* curves, can now be evaluated quasi in situ for the assessment of the laser and thin-film interactions and for optimization of the laser-scribing process of TFSCs.

The sequential multi-pass scribing of the CIGS TFSC with ultrashort laser pulses (λ = 775nm, t_p = 150fs) caused a sudden drop in the parallel resistance (R_p) due to shunt formation, as shown in Fig. 5(b). This happened during the first scribe, which resulted in a laser ablation depth of nearly one micron. However, further scribes did not change R_p greatly, as further laser pulses hit only the modified CIGS material. The results show that even femtosecond pulses can introduce thermal modification of CIGS if the film is absorbing energy and is not completely removed during scribing.

Electrical measurements of the solar cell characteristics give no information about the reasons for the laser-induced modifications or about their distribution. Thus, localized studies using electro-optical techniques can help in understanding the laser-induced defect formation, and are required to study the correlations of electrical defects to chemical, physical and topographical properties at the scribing edge. Suitable techniques for high-resolution electro-optical measurements are laser beam-induced current (LBIC) mapping, electroluminescence (EL) imaging and dark lock-in thermography (DLIT) in the reverse current direction.

Lock-in thermography (LIT) has proved to be a valuable technique for non-uniformity diagnostics in crystalline and polycrystalline solar cells [19]. It utilizes AC infrared imaging of a device, where the temperature is affected by an external AC voltage of the same lock-in frequency. The thermography images represent the local current losses.

To detect any short-circuiting caused by laser ablation, LIT measurements were performed in the area close to the scribes. A typical LIT image for the CIGS cell scribed using the infrared 1064nm radiation of a picosecond laser is shown in Fig. 6(a). The film deposition defects are clearly visible and cause a short-circuit current leak at the examined surface. However, no change in the surface temperature was observed for the picosecond or femtosecond scribing regimes. No significant internal shunt formation was detected during laser scribing with either pulse duration. The IR camera was optimized for large-area observation; the resolution was too low to observe an area of the order of a scribe width, and small defects may not have been detected.

The high-resolution DLIT technique in the reverse current direction is well suited to analyzing localize shunts at the edge of laser scribes [10]. The DLIT image in Fig. 6(b) clearly shows that the laser-induced defects are located only on one side of the scribe, and that no defects, for example due to speed reduction, occur at the end of the scribe. The localization of the defects might be due to inadequate overlapping (hatching) in making a wide scribe or to the non-symmetric beam profile. Both localized and distributed shunts are found at the laser-scribe edges. Current investigations show, however, that the efficiency of thin-film $Cu(In,Ga)Se_2$ modules can also be reduced as a result of laser scribing with ultrashort laser pulses.

Laser beam-induced current is a non-destructive optical technique used for defect detection in semiconductors. The LBIC image consists of measurements of the total local current, flowing out through contacts, that is induced by the local laser beam irradiation. The diode laser beam was focused into a 50µm spot and this defined the spatial resolution of the technique. Laser scribing was performed between the fingers of the front contact. The LBIC map of the areas close to the laser scribe (see Fig. 6(c)) showed a uniform distribution of current in the area close to the scribe lines made with the picosecond laser. The dead area near the scribe was minimal, although the resolution of the measurements did not allow an investigation of the defect formation on a micro-scale at the edge of the ablated trench.

PV performance after laser scribing

Complete working solar cells from a prefabrication stage, with an average efficiency of 10.7% and an active surface area of $32cm^2$, were scribed using optimal process parameters. The total length of the laser scribes was 360mm in all cases. The photoelectrical efficiency and parallel resistance measurements were taken before and after laser scribing to evaluate the influence of the laser scribing on the solar cell performance. Reference cells not subjected to laser scribing were also used to monitor degradation of the experimental cells. The measurements were performed using irradiance of the standard global spectrum AM 1.5 and $1000W/m^2$ intensity. The initial photoelectrical efficiency of the CIGS solar cells used in experiments was about 10.7%.

The photovoltaic efficiency tests after laser scribing revealed a minor decrease in the solar cell performance and parallel resistance during optimized laser scribing with picosecond and femtosecond pulse durations. The average drop in efficiency of the CIGS solar cells after laser scribing was 0.35%, which also included the effect of a reduction in active area due to scribe width.

Issues in the industrial implementation of CIGS laser scribing

The industrial implementation of the laser-scribing technologies developed is not yet practical, because of issues concerning reliability, process speed and the realization of flexible substrates in the R2R concept. The reliability issue relates to the remaining effects of laser scribing on the PV performance of the cells, even using ultrashort lasers, and the acceptance of the variation in film thickness and composition. To produce rolls of flexible cells in a single run, a high scribing speed is necessary from an economic point of view. The separate locations of the laser scribing processes (P1, P2, P3) for MIIs along the production line cause an obstruction or a stoppage in the technology implementation. Further progress in flexible CIGS solar cells may therefore benefit from new interconnect architectures. Those issues will be addressed next.

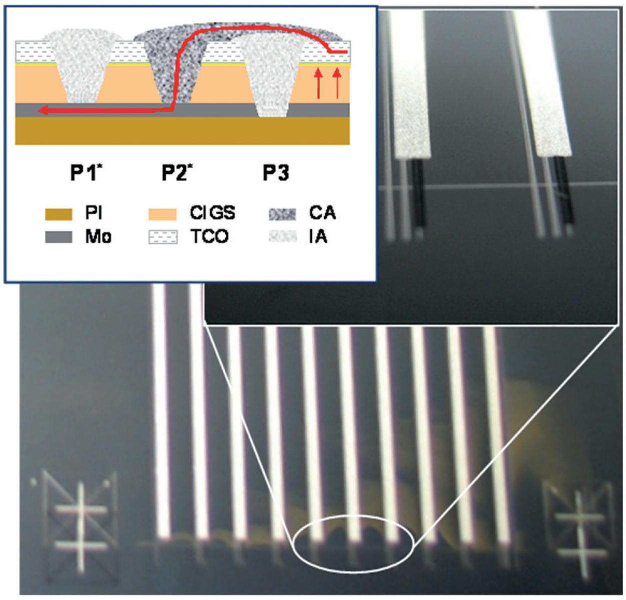

Figure 8. CIGS modules fabricated by the external integrated interconnection (EII) scheme using the R2R laser-scribing process.

> "Laser scribing in the multi-layered structures of modern TFSCs requires high selectivity in the ablation of the films."

Reducing the modified area by the use of shaped laser beams for scribing

Laser scribing in the multi-layered structures of modern TFSCs requires high selectivity in the ablation of the films, without adjacent material being affected. The focused spot of a Gaussian beam that is limited by a beam diameter contains only 86.5% of the laser beam power, and the intensity on the boundary is only 13.5% of the peak intensity. The energy in the wings causes modification of the surrounding material. This type of spatial beam profile does not therefore permit a distinct boundary of the scribes to be achieved. Specialized optical elements are able to transform the smooth Gaussian profile into the square-shaped flat-top beam [20]. The utilization of a top-hat profile beam reduced the laser-modified area on the edges of the ablated scribe by 34% because of uniform irradiation within a limited area, with sharp edges on the boundaries. Moreover, the width of the scribe was less sensitive to instability in the laser pulse energy for the flat-top profile beam. Reliability of the scribing process with regard to the material variation was improved.

Utilization of the full laser power by parallel scribing

Industrial scribing applications demand cost-effective, high-speed processes that are able to be easily integrated into existing production lines. In most cases the best scribing results were achieved when the laser power was a fraction of the total available power of the source. One way to increase productivity and exploit the full power of the laser is to split the beam and then scribe, in parallel, a few lines simultaneously with the same system. The typical distance between integrated interconnects in TFSCs is 5–10mm, which means that several segments of the solar cell fit into the working field of the galvo scanner, and a common beam-guiding system can be used. Moreover, simultaneous scribes for advanced external interconnects can be realized in a single pass, keeping a tight distance control between separate scribes of the same interconnect. The 'dead area' can be reduced significantly with this approach.

The scribing of CIGS TFSCs with four parallel beams was realized by working with a single scanner head and installing a beam splitter in the experimental set-up [21]. The overall power required for the scribing is much lower than the maximum power of the laser used, and the processing can be easily realized using four or more parallel beams. The distance between focused parallel laser beams was adjusted by the splitting angle, which depended on the optical set-up. The same scribing conditions (focusing, power) were maintained for parallel beams, with an excellent repeatability of the laser-scribed trenches (Fig. 7). This illustrates the potential for optimizing laser-scribing processes for CIGS solar cells while meeting the demands of an industrial implementation in terms of process speed and economics.

The process parameters were very close to those obtained for the single-beam configuration. As only about one-tenth of the full laser power was required for the P3 scribing in CIGS solar cells, the approach can be expanded to the ten-beam processing using a picosecond laser. By using a single laser for multiple simultaneously scribed lines, a substantial reduction in the processing cost may be realized.

External integrated interconnections and CIGS modules

By using the EII approach [22], CIGS TFSCs on flexible substrates were interconnected by laser scribing and screen printing of conductive adhesive to fabricate mini-modules. For the interconnection, EII (a cross section is shown in Fig. 8) requires three parallel scribes, which can be made simultaneously as discussed above. However, the requirements of the laser-scribing process for EII are even more demanding than those of the scribing process for MII, as damage to all films and interfaces of the TFSC must be avoided for all scribes from P1 to P3. This is a challenge because the full CIGS stack has to be scribed and requires specific optimized laser-scribing processes.

The complete EII process for CIGS module fabrication comprises:

1. Deposition of the complete stack of CIGS solar cell material.
2. Laser scribing with ultrashort laser pulses.
3. Formation of the interconnection by screen printing of a silver-containing conductive adhesive.

The benefits of this EII technology are: (1) the reduced demand on the scribing procedure concerning the requested overlay accuracy; (2) the division of the fabrication process into thin-film deposition and laser-scribing steps, whereby both the thin-film deposition and the laser scribing can be optimized without interference; and (3) the opportunity to use the same process for CIGS thin-film deposition as for CIGS solar cell fabrication. The laser-scribing procedure in particular can be improved by developing parallel scribing schemes with adapted laser fluences, scribing widths and number of scribe repetitions.

Laser-scribed flexible CIGS thin-film modules, illustrated in Fig. 8, with an output voltage of ~4.5V and an efficiency of ~10% were fabricated. A relative precision and overlay accuracy to external markers of better than 5µm and 10µm, respectively, have been measured within the module area of 20×20cm^2. The rather extended interconnection area is a result of the limited precision of the screen printing, which potentially offers scope for further increases to be made in module efficiency in the future.

Conclusions

The modelling of laser energy coupling and an extensive characterization of laser scribes have facilitated the development of a high-speed laser scribing of CIGS solar cells on flexible polymer substrates. The selection of the appropriate laser wavelength allowed the energy coupling to be kept in a well-defined region at the interface between layers. The high absorption at the inner interface of the layers triggered a localized temperature increase. The transient stress caused by the rapid temperature rise led to peeling of the films rather than evaporation.

Although the quality of laser scribes is promising, most of the processes are too slow in real production lines. The reliability of the scribing process is still an open issue as well. However, to address these issues, a parallel-processing approach has been developed, and the beam-shaping technique was applied to increase the quality of scribes.

"A new interconnect architecture similar to the external integrated interconnection may make it easier to adapt the laser-scribing technology to the roll-to-roll production of flexible solar cells."

The picosecond lasers used in scribing the thin-film CIGS solar cells exhibited high potential for industrial applications. A new interconnect architecture similar to the external integrated interconnection may make it easier to adapt the laser-scribing technology to the roll-to-roll production of flexible solar cells.

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