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Shadowgraph studies of laser-assisted non-thermal structuring of thin layers on flexible substrates by shock-wave-induced delamination processes

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ABSTRACT

The laser-assisted microstructuring of thin films especially for electronic applications without damaging the layers or the substrates is a challenge for the laser micromachining techniques. The laser-induced thin-film patterning by ablation of the polymer substrate at the rear side that is called 'SWIFD' – shock-wave-induced film delamination patterning has been demonstrated. This study focuses on the temporal sequence of processes that characterize the mechanism of this SWIFD process on a copper indium gallium selenide (CIGS) solar cell stacks on polyimide. For this purpose high-speed shadowgraph experiments were performed in a pump probe experimental set-up using a KrF excimer laser for ablating the rear side of the polyimide substrate and measuring the shock wave generation at laser ablation of the polymer substrate as well as the thin-film delamination. The morphology and size of the thin-film structures were studied by scanning electron microscopy (SEM). Furthermore, the composition after the laser treatment was analyzed by energy dispersive X-ray (EDX) spectroscopy. The shadowgraph experiments allow the time-dependent identification and evaluation of the shock wave formation, substrate bending, and delamination of the thin film in dependence on the laser parameters. These results will contribute to improve the physical understanding of the laser-induced delamination effect for thin-film patterning.

1. Introduction

The conversion of the energy supply to renewable energy sources represents one of the most important and necessary industrial challenges where solar cells can be play an important role if the fabrication of low-cost, high-efficient and robust solar cells can be improved. For example solar cell on flexible substrates allows the implementation in new applications fields where the laser-based fabrication techniques exhibit an outstanding potential for the fast and easy structuring of thin layers especially on flexible substrates. However, the high thermal sensitivity of the active layer of the solar cell especially from organic semiconductors needs special requirements on the laser method. The direct ablation process for the selective structuring of the solar cell allows the fast patterning of the solar cell especially with ultra-short laser pulses [1], however secondary effects like thermal effects, ripples formation, and debris deposition processes restricted the practical applicability. The indirect laser methods like laser-induced shock waves structuring [2] can be applied for thin-film structuring with a distinct reduction of nuisance secondary effects. This method is called SWIFD (shock wave-induced film delamination) [2]. At SWIFD, the irradiation of the rear side of the substrate induced laser ablation and shock wave formation process, respectively and the mechanical effect (shockwave and bending of the substrate) on the front side result in a delamination effect of thin layers where the laser-induced shock wave in solids is well known [3]. Furthermore, the delamination effect can be discussed in relation to thermal-induced stress effects [4]. This patterning process can be used e.g. for CIGS solar cells [2]. The laser-induced shock wave effect can be used e.g. for the measurement of the adhesion of thin films [5] and for the laser-induced forward transfer (LIFT) of layer systems [6–8]. This study focus on the investigation of the laser induced secondary non-thermal processes that must be considered for the discussion of the SWIFD-mechanism by visualization of the shock and

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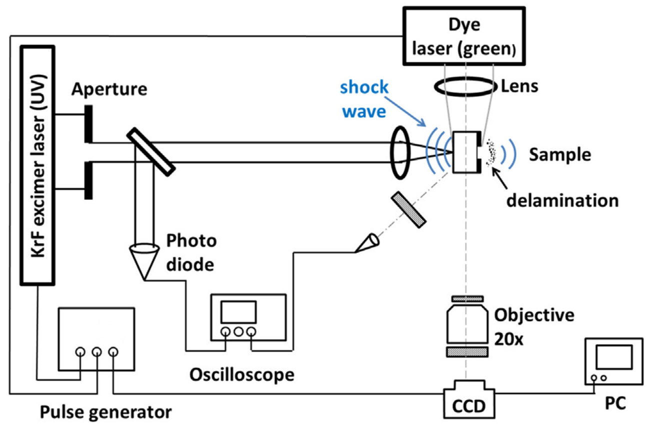

Fig. 1. Schematic illustration of the used pump probe experiment for time-resolved imaging.

forces induced effects. Therefore, the time-resolved shadowgraph measurement that is regularly applied for laser driven mechanical processes [9–12] was used for the examining of the delamination.

2. Experimental set-up

The primer experimental set up in Fig. 1 shows a typical set-up for time resolved shadowgraph photography. For pumping the processes of SWIFD a KrF excimer laser is used that ablates polyimide easily. For imaging the beam of a dye laser illuminates the sample from the side in orthogonal configuration.

For the experiments 25 μm thin polyimide foils covered with a full CIGS solar cell thin-film stack was used. The CIGS solar cell thin films include a indium tin oxide (ITO) front contact and a copper indium gallium selenide (CIGS) film with a entire thickness of 2.2 μm as well as and a 1 μm thick molybdenum (Mo) back. From the material samples were cut by scissors and clamped mechanically. Before the experiments the samples were adjusted to the imaging plane of the CCD camera. The rear side of the polyimide foil was irradiated by a KrF excimer laser pulses of a wavelength of $\lambda = 248$ nm and a pulse duration of $t_p = 25$ ns. The focal spot size was approximately 550 μm. Furthermore, the laser-induced delamination effect was studied by time-dependent optical imaging of the side of the irradiated CIGS solar cell layer system where the sample was irradiated by an electronically delayed N_2 laser pumped Coumarin 153 green dye laser ($\lambda = 543$ nm, $t_p = 1$ ns, $E_p < 1$ mJ). The side view of the layer system was imaged by a 20× objective with optical correction on an external triggered CCD image sensor. The time delay ($\Delta\tau$) between the shock wave induced excimer laser, the illumination dye laser and the recording point of the camera could be adjusted by a freely programmable pulse generator. The delay between the excimer laser and the dye laser as well as the laser fluence stability of the excimer layer was controlled by photo diodes with a delay time of 1 ns where the diode signals were recorded by a 1 GHz oscilloscope. After the shadowgraph experiments the laser irradiated sample surfaces were imaged by scanning electron microscopy (SEM) and their atomic composition was studied by energy-dispersive X-ray spectroscopy (EDX).

3. Results and discussion

The samples were irradiated at a laser fluence of $\Phi \approx 2.8$ J/cm^2 and the images were recorded at different time delays. Except for pulse number studies at fixed laser parameters for each set of experimental properties a fresh not irradiated sample area was used. Hence typically only first pulse images are taken. Shadowgraph images at different time delays after the irradiation with a KrF excimer laser pulse with laser fluence of $\Phi = (2.8 \pm 0.15)$ J/cm^2 are shown in Fig. 2. The images presented manifold laser-induced effects that influence the finally observed thin film patterning. The formation of a shock wave in the air near the laser-irradiated rear side of the sample can be observed at small time delays ($\Delta\tau < 200$ ns). With increasing time delay ($\Delta\tau \geq 500$ ns) the bending of the complete sample and the removal of material from the rear side of the PI substrate can be found. Finally, for even higher $\Delta\tau \geq 750$ ns the thin film delamination and the fly away of the delaminated material can be seen. Further features that can

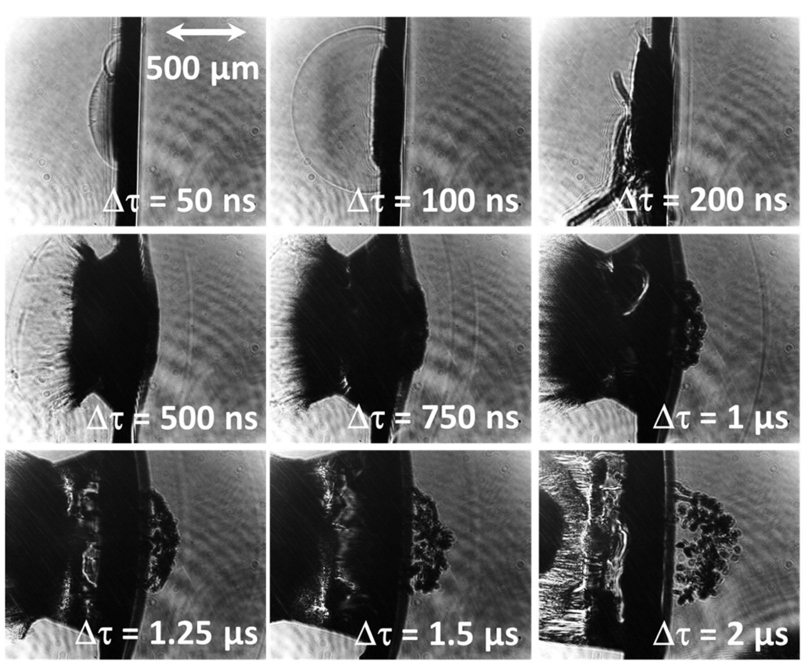

Fig. 2. Shadowgraph images of laser irradiated samples ($\Phi \approx 2.8$ J/cm^2) at different time delays $\Delta\tau$. The laser beam is focused on the rear side of the PI substrate, left side in the images.

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