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Enhanced light absorption in nanotextured amorphous thin-film silicon caused by femtosecond-laser materials processing

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ABSTRACT

Efficient thin-film solar cells balance the reduced absorption occurring in thin absorber layers by means of various photon management strategies that often involve randomly nanotextured interfaces. We report on broadband absorption enhancement in nanotextured amorphous silicon processed by femtosecond laser materials processing. As identified by micro-Raman spectroscopy and surface profilometry, the absorption of a single femtosecond amplifier laser pulse (30 fs, 795 nm, 75 mJ cm^{-2}) creates a thin nanotextured micro-crystalline surface layer. Optical microscopy in transmission and reflection geometry reveals a broadband absorption enhancement in the visual spectrum range for the nanotextured area. Scattered light spectroscopy in combination with spectral interferometry indicates that light trapping for about 100 fs is achieved in the femtosecond-laser processed amorphous silicon area and thus is responsible for the observed enhanced absorption and locally enhanced Raman yields. Thus fs-laser materials processing offers an interesting pathway towards advanced photon management in amorphous silicon based thin-film solar cells.

1. Introduction

Efficient thin-film solar cells compensate the low single layer absorption by advanced photon management schemes to effectively utilize the incident sun spectrum. A large variety of photon management strategies is employed, such as for example the integration of randomly or periodically textured interfaces, plasmonic antennas, or the use of multi-junction solar cells. The established technology in the area of thin-film silicon solar cells assembled in superstrate configuration is the application of randomly textured transparent conductive oxides (TCO) [1–4]. It is assumed that light scattering at the corrugated TCO/a-Si:H interface leads to longer light propagation pathways in the absorber layer and thus enhances absorption. Based on the assumption that the surface corrugation of the absorber layer is the essential ingredient for this photon management strategy any nanotexturing method will be of interest. The formation of highly absorbing nanotextured interfaces, i.e. black silicon, via femtosecond (fs)-laser materials processing is already known for crystalline silicon [5,6]. In this case fs-laser induced surface photochemistry is employed to create a highly corrugated surface topography that efficiently captures incident light and leads to almost perfect absorption via the moth eye effect [7]. Recently, also for a-Si:H, enhanced light absorption in fs-laser crystallized layers was reported [8,9]. Various mechanisms, such as fs-laser induced defect band formation and light trapping in structural spikes, were suggested to explain the enhanced absorption [8].

Here we investigate the fs-laser induced absorption enhancement in a-Si:H layers directly using time resolved light scattering spectro-microscopy and indirectly via the Raman yield enhancement. The investigation confirms that fs-laser materials processing of a-Si:H represents a promising approach to prepare nanotextured absorber layers with enhanced light absorption and shows that light trapping is essential for absorption enhancement.

Optical microscopy in transmission and reflection geometry, micro-Raman spectroscopy, and time resolved light scattering experiments are carried out to analyze the material modification induced by a single amplified fs-laser pulse with thin a-Si:H films. More detailed information regarding the laser material processing of c-Si and a-Si:H with different hydrogen contents is given elsewhere [10–13]. It is shown that the crystallization process in the a-Si:H layer creates nanoscale surface corrugations in coincidence with enhanced absorption that is identified in optical microscopy in transmission and reflection mode. Broadband coherent scattering light spectro-microscopy is used to assess

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the light trapping properties of fs-laser modified areas. The results reveal that an enhanced light trapping time is responsible for the absorption enhancement in the fs-laser material processed areas as it is seen in optical microscopy. This demonstrates that the application of femtosecond-laser pulses enables both (i) material and surface modification for the development of efficient absorber materials and interface designs and (ii) determination of the lifetime of trapped light in layers with a nanotextured interface.

2. Material and methods

Hydrogenated amorphous silicon (a-Si:H) is deposited by plasma enhanced chemical vapor deposition (PECVD) on a quartz glass substrate at room temperature. Due to the low deposition temperature the hydrogen content of the film is about 30%. The Fs-laser materials processing (Fig. 1a) was performed using a single amplified laser pulse selected by a Pockels cell switch from the 1 kHz fs-pulse train of a commercial multipass Ti:sapphire amplifier (Femtopower PRO CEP). A plano-convex lens (focal length $f=1$ m) was used to focus the almost Gaussian shaped laser beam on the surface with a spot diameter of $d=420\,\mu$m ($1/e^2$ width of the intensity profile). The laser beam profile at the position of the film was determined using a laser beam profiler (TaperCam-UCD12 DataRay Inc.). 290 nm thick a-Si:H layer (Fig. 1) was processed using a peak fluence of 75 mJ cm^{-2}, i.e. a peak fluence above the thresholds for the onset of optical properties changes (about 30 mJ cm^{-2} [12]) and material ablation (about 40 mJ cm^{-2} [12]).

The single shot modified areas were characterized by optical microscopy in transmission and reflection geometry, step height profilomerty (Dektak 6M Stylus Profiler Veeco), imaging ellipsometry at 658 nm, scanning electron microscopy (SEM), micro-Raman spectroscopy (Horiba LabRAM ARAMIS), and analysis of the scattered light in combination with spectral interferometry to reconstruct the temporal evolution of the scattered field [14]. It was shown in a previous work that for the same irradiation conditions as they are used in the present investigation the amplified fs-laser pulses are efficiently absorbed in a thin surface layer because of a nonlinear absorption mechanism. This leads to fs-laser induced material modifications such as Si–H dissociation and crystallization that have been recently investigated [12] and that are not further discussed here.

The measured intensity profile of the fs-laser materials processing laser in combination with spatially resolved techniques such as micro-Raman spectroscopy (1–2 μm Raman laser spot diameter) allows analyzing the fluence dependence of the material modification by recording spatially resolved signals across individual spots. Qualitative depth information of the material modification is obtained from micro-Raman spectroscopy using different excitation wavelengths (473 nm and 633 nm) with different penetration depth profiles. The crystallinity of the fs-laser materials processed layer is determined via spectral decomposition of the Si–Si vibrational mode around 500 cm^{-1} Stokes shift in the amorphous component at 480 cm^{-1}and the microcrystalline component at 505 cm^{-1} and 520 cm^{-1}. The crystalline fraction is obtained as the sum of the microcrystalline yields normalized to the Raman yield in the 500 cm^{-1} spectral range. Besides the evaluated crystalline fraction from spectroscopic measurements the Raman signals associated with the Si–H vibrational modes (2000–2100 cm^{-1} Stokes shift) are used for the characterization of the hydrogen content.

The experimental setup for scattered light spectroscopy and spectral interferometry is illustrated in Fig. 2. For materials processing and time-domain spectro-microscopy of the scattered light, different femtosecond lasers with different parameters were applied. For a simple identification, the term “fs-laser” is used in the context of laser materials processing and the term “ultrashort laser pulse” is applied in case of scattered light spectroscopy. Ultrashort laser pulses (25 fs duration, 790 nm center wavelength, 8 mW average power, and 80 MHz repetition rate) are focused with a parabolic mirror ($f=15$ mm) onto the surface of the sample at an angle of incidence of 45°, achieving a focal spot size of less than 2 μm ($1/e^2$ width). The scattered light is collimated using the parabolic mirror. In the present investigation, as selected by the position of the aperture, scattered light at 22° with respect to the specular reflected beam is recorded. As the sample was mounted on a translation stage, the laser focus was scanned

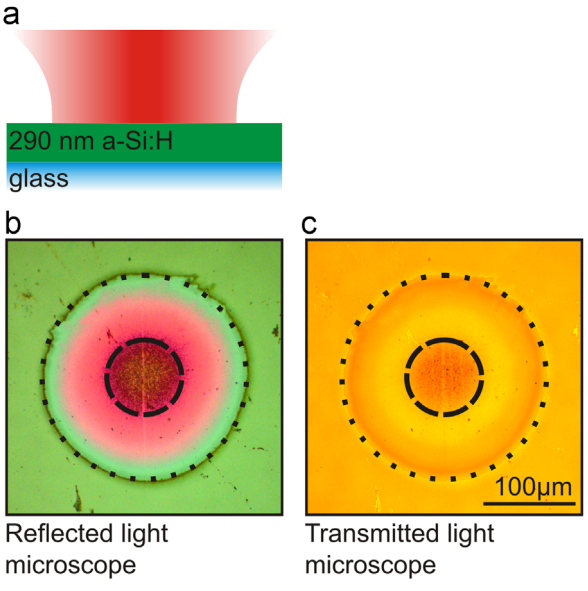

Fig. 1. (a) A Fs-laser materials processing. A single amplified fs-laser pulse (center wavelength of 795 nm, pulse duration of 30 fs, pulse fluence 75 mJ cm^{-2}, and focus diameter of 420 μm) impinges on a 290 nm thick a-Si:H layer (30% hydrogen content) deposited on a glass substrate. (b,c) Optical microscopy images of the fs-laser processed spot recorded in reflection (b) and transmission (c) geometry. The dashed line encloses the center region in which an absorption enhancement is observed at the center of the amplified fs-laser pulse treated area. The dotted line indicates the rim of the ablation region.

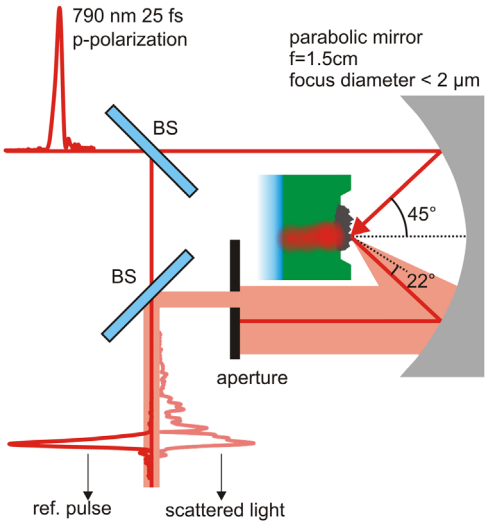

Fig. 2. Spectro-microscopy setup for scattered light characterization and spectral interferometry measurement. Scattered light is selected with respect to the scattering angle by an aperture and coupled to a single mode fiber. For reconstruction of the temporal evolution of the scattered electric fields, spectral interference measurements are performed using the original ultrashort laser pulse as a reference. The cross-hatched region in the schematic sample cross section indicates the microcrystalline layer forming because of the fs-laser materials processing.