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High Spatial Resolution Analytical Electron Microscopic Investigation of Femtosecond-Laser-Induced Crystallization of a-Si:H Films

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Laser induced crystallization of thin amorphous semiconductor films is regarded as an attractive technique for fabrication of thin film transistors in active-matrix liquid-crystal displays [1,2] and of high-efficiency low-cost thin-film solar cells [3,4]. Typically nanosecond or microsecond lasers are utilized to crystallize such films via a rapid melting and solidification process. Ultra short laser pulse interaction with material introduces nonlinear photon energy absorption and nonequilibrium thermodynamics that are anticipated to govern the interactions [5-7]. Ultra short laser interactions with solid provide precise and low threshold fluence for material ablation [8-10]. It has been demonstrated that by controlling the laser fluence and number of pulses impinged on hydrogenated amorphous silicon thin films (a-Si:H), one can crystallize and texture the film at the same time [11]. This one step processing could have tremendous technological importance for fabrication of high efficient thin film solar cells and photo detectors. However, further in-depth investigation is necessary to understand the relations between structural changes, surface texturing and functional properties. This work is aimed to study the structural evolution of a-Si:H films after being exposed to a single femtosecond laser pulse using high spatial resolution field-emission analytical electron microscopy (FE-AEM) combining conventional and phase contrast electron imaging, selected-area electron diffraction (SAED) with energy-dispersive X-ray (EDX) and parallel electron energy-loss (PEEL) spectroscopies.

The samples for the study were a-Si:H single junction p-i-n type of solar cells fabricated on glass substrates using standard PECVD technique. The samples were placed on a x-y stage and treated using a 120 femtosecond Ti : Sapphire laser system (Spectra Physics, Spitfire). The system delivered pulses at a repetition rate of 100 Hz and a wavelength of 800 nm. The laser fluence was maintained at 0.24 J/cm^2. The beam spot on the sample was circular with beam diameter of 0.76 mm. The samples were scanned in such a way that each point on the surface was exposed only to one laser shot. After that the specimen were collected on the microscopy grids. Bright-field and dark-field (BF-/DF-) TEM/ SAED/(HR) TEM, EDXS and PEELS of the films were done on a JEOL JEM-2010F field-emission AEM operating at 197 kV with a high-tilt pole piece (C_s=1.0 mm and 0.23 nm resolution by points) and utilizing a Gatan Imaging Filter, model 678 and an Oxford Pentafet ultrathin window EDX detector with resolution of 136 eV (MnKα). PEEL spectra were acquired using a 0.6 mm entrance aperture in the image mode with 0.05-0.1 eV/pixel dispersion and 1.0 eV resolution at the zero-loss peak (FWHM). (HR)TEM/SAED revealed largely amorphous structure in the initial film (FIG. 1a). In the processed film, diamond type cubic Si crystalline sphere-like surface (and defect) structures and c-Si nanocrystals of different sizes and orientations grown in the amorphous matrix were found (FIGs. 1b and 1c). Nanoprobe EDXS performed using nominal probe size varying from 2.4 nm to 0.5 nm indicated some surface oxidation of the film during laser processing. First derivatives of low-loss PEEL spectra (FIG. 2a) demonstrate a 0.4 eV high-energy

shift of the bulk plasmon at 16.7 eV indicating an increase in density of the processed film. The net $SiL_{2,3}$-edge (FIG. 2b) also shows a femtosecond-laser-induced low-energy shift of the ELNES features at 103.4 eV and 111.6 eV reflecting variations in the symmetry-projected density of s, p and d conduction band states at Δ_1, L_1 and L_3 in the Brillouin zone.

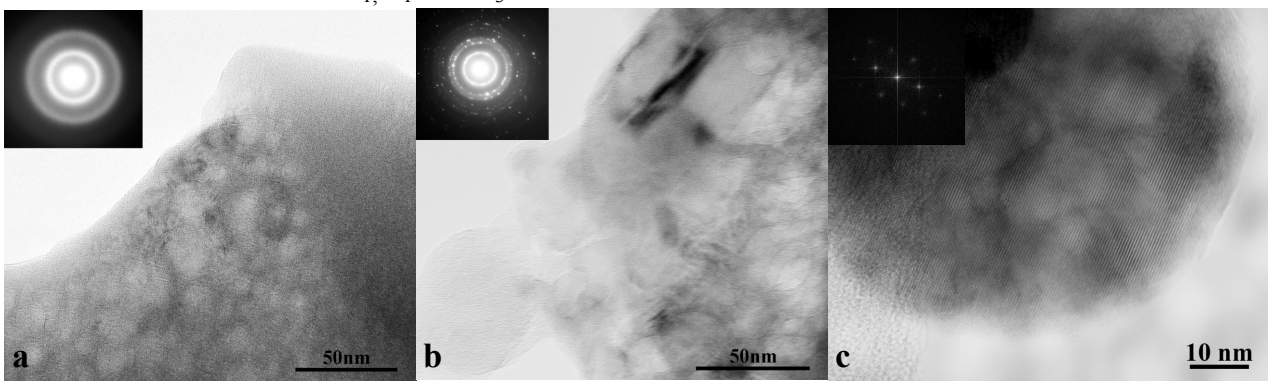

FIG. 1. TEM micrographs of (a) initial a-Si:H film and (b, c) femtosecond laser-processed a-Si:H film. Insets in (a, b) show corresponding SAED patterns. (c) HRTEM image and Fourier transform (inset) of a processed crystalline spherical particle demonstrating {111} and {220} lattice fringes.

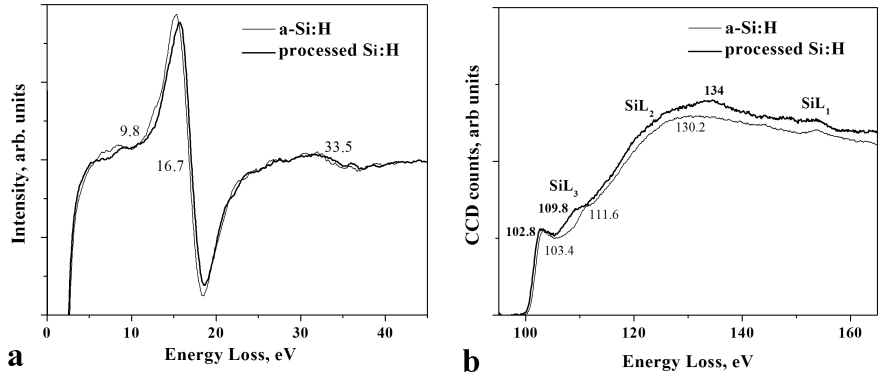

FIG. 2. PEEL spectra of untreated and femtosecond laser processed a-Si:H film. (a) First derivatives of low-loss PEEL spectra show the bulk plasmon (16.7 eV), surface (9.8 eV) and double (33.5 eV) plasmons. (b) The net $SiL_{2,3}$-edge.

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