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Preferential {100} grain orientation in 10 micrometer-thick laser crystallized multicrystalline silicon on glass

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ABSTRACT

Liquid phase crystallization of 10 µm thin silicon layers on glass substrates was performed with a line-shaped continuous wave laser beam. The process window was investigated in terms of the scanning velocity of the laser, the pre-heating of the specimens and the applied laser intensity. We have identified the entire process window, in which large-scale crystallization without deformation or destruction of the substrate and cracking of the silicon layer can be obtained. The grain orientations of the resulting Si layers were analyzed using both electron backscatter diffraction (EBSD) and X-ray diffraction (XRD). The influence of the critical crystallization parameters on the grain orientation of the silicon film was examined. EBSD and XRD measurements show that a preferential {100} surface texture and {100} and {101} orientations in scanning direction of the laser can be achieved if appropriate crystallization parameters are used. This texture formation is accompanied with a substantial decrease of high angle grain boundaries.

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1. Introduction

Recently, polycrystalline silicon (poly-Si) films received renewed attention as an attractive material for the next-generation of Si thin-film solar cells. Commonly, solid phase crystallized (SPC) poly-Si is employed for such devices, which suffers from a number of shortcomings such as small grains, large defect concentrations [1,2], and the absence of a surface texture [3]. Some of these limitations can be overcome by using continuous wave (cw) diode laser crystallization of silicon films on foreign substrates. This method is also known as liquid phase crystallization (LPC) and produces poly-Si films that are composed of large laterally grown grains of several cm in length and a few mm in width [3–6]. Because of the large grains the electrical properties of LPC poly-Si are mainly determined by dislocations, impurities and certain grain boundaries that can introduce localized defects in the band-gap [7–10].

For enhancing the power conversion efficiency of solar cells a surface texture that allows the incorporation of light-trapping structures is desirable. A {100} orientation allows for direct integration of light trapping using KOH based etching, thus increasing the absorption length significantly. Secondly the resulting pyramidal facets of {111} orientation are an ideal prerequisite for the deposition of an a-Si:H (i/n +) or a-Si:H (i/p +) hetero-junction with low interface state density. Until now, a preferential surface orientation for poly-Si on glass substrates has only been reported for excimer laser

crystallization of silicon [11,12]. The type of texture is determined solely by the thickness of the silicon layer. Below a critical value of about 40 nm texture formation is governed by surface energy anisotropy and a {100} texture forms. On the other hand, for thicker Si films the kinetics of the crystallization process predominate and the resulting texture shows a {111} orientation [11].

Recently, the influence of the deposition temperature and cw-diode laser annealing on the surface texture and microstructure of thick poly-Si films were investigated. Depending on the deposition temperature of the starting material a {100} and {111} surface textures of up to only 20 and 45% were obtained, respectively [13]. In this work, we demonstrate for the first time that a strong {100} surface texture can be achieved with cw-laser crystallization by using appropriate crystallization parameters. This is accompanied with a substantial decrease of high angle grain boundaries.

2. Experimental

The sample preparation consisted of the following steps. As substrate Corning Eagle XG glass with a size of $5 \times 5 \text{ cm}^2$ was used. After thorough cleaning with an alkaline cleanser in an ultrasonic bath a reactively sputtered silicon oxide layer with a thickness of 200 nm was deposited from a 6" 6 N-Si target using a 13.56 MHz RF magnetron sputtering system. The gas flow was set to 100 sccm with an O₂/Ar-ratio of 0.07. The power and the pressure of the process were 1000 W and 0.01 mbar, respectively. The substrate temperature was set to 150 °C. This silicon oxide layer serves as a diffusion barrier [3,14]. In a second step nanocrystalline-silicon layers were deposited to a thickness of







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10 µm by electron beam evaporation of float zone silicon. The deposition temperature amounted to 600 °C. P-type was achieved by adding boron from an effusion cell. The doping concentration was in the range of 10^{16} – 10^{17} cm⁻³. Subsequently, the samples were crystallized using a line-shaped cw laser from LIMO with a wavelength of $\lambda = 808$ nm \pm 10 nm. The laser had a top-hat profile along its line width of 31 mm. Across the line the profile was Gaussian with a full width half maximum of 0.177 mm. With this configuration an optical intensity of 0.5–18.5 kW cm⁻² was available for the crystallization process. The specimens were mounted on a movable heater stage. Crystallization was performed at elevated temperatures and in ambient atmosphere by moving the samples under the laser beam with a velocity of up to 50 mm/s.

The crystallized samples were characterized using scanning electron microscopy (SEM), electron backscatter diffraction (EBSD), and X-ray diffraction (XRD) measurements. For EBSD measurements a cold field emission gun with an acceleration voltage of 25 kV and an emission current of 20 μ A were used. The samples were cut to a sample size of 1 × 1 cm² and mounted on a sample holder. EBSD micrographs were taken on an area of 800 × 800 μ m² with a lateral resolution of 5–10 μ m. The XRD measurements were performed on a five-circle ETA diffractometer using a Cu K_α radiation with a wavelength of $\lambda = 1.54056$ Å. The primary beam cross-section was 1.5 mm. For the pole figures angle dispersive XRD measurements in symmetrical Ψ mode were performed. The counting time was set to t = 2 s for each step. To obtain information on the {111}, {220}, {311} and {400} lattice planes the 2 θ angles were set to 28.5, 47.3, 56.1, and 69.1°, respectively.

3. Results

Fig. 1 shows cross-sectional secco-etched SEM micrographs of poly-Si samples that were crystallized with optical laser-intensities that varied between $I_{opt.} = 1.904$ and 2.073 kW cm⁻². The crystallization was performed at a substrate temperature of $T_{sub.} = 700$ °C and a scan velocity of $v_{laser} = 3$ mm/s. Such high substrate temperatures are of advantage since they decrease the thermal gradient during crystallization and thus help avoiding the formation of cracks in the poly-Si layer and glass substrates.

At low laser intensity [Fig. 1(a)] the poly-Si sample consists of some grains that are separated by highly defective regions. At the interface to the SiO₂ coated substrate at the bottom of the micrographs the poly-Si film is highly disordered. With increasing I_{opt.} the disordered regions between the grains and the Si/SiO₂ interface decrease and ultimately disappear at an optical laser-intensity of 2.073 kW cm⁻² [Fig. 1(d)], as soon as elongated grains are formed, extending through the whole absorber thickness. A similar behavior was observed for laser scanvelocities up to $v_{\text{laser}} = 15 \text{ mm/s}$. However, with increasing v_{laser} the laser intensity had to be adjusted to larger values. The data are summarized in Fig. 2. The circles indicate the transition from as-deposited nanocrystalline to small-grained poly-Si [see left inset of Fig. 2]. Although the specimens are completely crystallized with grains extending from the disordered interface region close to the substrate to the surface the grains are embedded in a highly disordered Si matrix [see Fig. 1(a)-(c)]. The triangles and squares comprise the parameter space where crystallization leads to the formation of elongated grains with a grain width of several mm. The grain length is mostly limited by the size of the specimens or the length of the crystallization scan [see right inset of Fig. 2]. When I_{opt.} exceeds values indicated by the squares the silicon melt converges into a branching structure. This dewetting effect becomes increasingly dominant as the laser intensity is raised. Although laser crystallization can be performed with scan velocities $v_{\text{laser}} \leq 1 \text{ mm/s}$ the energy deposited in the sample stack raises the temperature beyond the softening point of the glass substrates causing deformation. Moreover, the demands of the diffusion barrier are increased. At elevated scanning velocities $v_{\text{laser}} \gg 15 \text{ mm/s}$ the



Fig. 1. Cross-sectional secco-etched SEM micrographs of laser crystallized poly-Si films on glass substrates (bottom). The samples were pre-heated to 700 °C and subsequently crystallized with a scan velocity of $v^{\text{laser}} = 3 \text{ mm/s}$ at the indicated optical laser-intensities, $I^{\text{opt.}}$.

crystallization process is unstable. The only known possibilities to enhance the crystallization range in terms of $\Delta I_{opt.}$ and v_{laser} are the utilization of alternative high temperature stable interlayers [15,16] or to use SiO₂ capping layers [5]. Nevertheless cracking of the substrate can occur due to tensile stress and the silicon growth is disturbed leading to a dendritic silicon growth [16].

The optical laser-intensity required to obtain large elongated grains also depends on the substrate temperature during crystallization. In Fig. 3 the minimum optical laser-intensity to obtain large elongated poly-Si grains is shown as a function of T_{sub} for scan velocities of $v_{laser} = 3$ and 10 mm/s. With increasing T_{sub} , the optical laser-intensity required to obtain large elongated grains decreases. However, when T_{sub} < 650 °C

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