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Understanding phenomena of thin silicon film crystallization on aluminium substrates

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Abstract

The realization of crystalline silicon thin films on foreign substrates is an attractive alternative to the ingot casting aiming at a reduction in total costs. The purpose of this work is to form polycrystalline silicon films using the crystallization of amorphous silicon deposited on aluminum (Al) substrates. The Al-substrate is used as a catalyzer for silicon crystallization but also as a conductive substrate and as a back reflector for the photovoltaic cell. The crystallization of 1-5 µm thick amorphous silicon films were carried out at a temperature of 550°C and for duration times from 10 to 80 min. The crystallized silicon films were then characterized by Raman spectroscopy, scanning electron microscopy and by electron backscatter diffraction. The analysis show that the annealed layer is composed of two distinct layers: a thin polysilicon film located just above the Al substrate and on the top a thicker layer made of a mix of silicon and aluminum. The thickness of the polysilicon film is found to increase with the annealing time. The crystallization of 5 µm thick amorphous silicon during 80 min resulted in a 1 µm thick polysilicon layer composed of grains of few micrometers in size. The mechanisms of accelerated crystallization are discussed. Such polysilicon films can be used as a seed layer for the growth of a thicker absorbing silicon film for photovoltaic applications.

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1. Introduction

Over the last few decades, the use of thin silicon layers has promoted the manufacture of thin film transistors and flexible solar cells at low prices [1-3]. For example, amorphous silicon (a-Si) has been subject of many research works because it can be deposited at low temperature with a very high deposition rate. Due to its high absorption coefficient owing the disordered structure, very thin amorphous silicon layers (<1 µm) are used [4]. However, since the electrical properties of such materials degrade with time under illumination [5], some research work has been directed towards polycrystalline silicon film fabrication as it is more stable and it shows better electrical characteristics. There are several methods to obtain polycrystalline silicon films such as: Laser Induced Crystallization (LIC), Solid Phase Crystallization (SPC) or Metal Induced Crystallization (MIC). Usually the pulsed lasers, e.g. excimer lasers (308 nm), allow light absorption very close to the surface, up to melting, but prevent heating of the holding substrate [6]. As a result, grains inferior to 1 micrometer in diameter are often obtained because of a high nucleation density caused by fast cooling due to the short pulse duration [7]. The continuous lasers such as the Nd-doubled YAG (532 nm) or the CW-808 nm gave much better crystallinity under high scanning speed [8, 9]. With these CW lasers, the melting is reached and nucleation occurs mainly at the edges causing elongations of the grains to several tens micrometers which are more suitable for the applications mentioned above. On the other hand, direct solid phase crystallization (SPC) enables crystallization of amorphous Si at temperatures far below the melting point. However, long durations lasting several hours are necessary [10-13]. For example, in 2 µm thick amorphous silicon deposited on quartz and annealed at 600°C for 10h, grains around 1 µm in diameter are obtained. The structure contains a lot of defects, which is mainly due to the poor control of nucleation inside the deposited layer. This is suggesting a higher concurrence between the growing grains and a non-perfect match up at the grain boundaries after the process [14]. An alternative to the solid phase crystallization is Metal Induced Crystallization (MIC), which consists in a thermal annealing of a stack of a metal and an amorphous silicon layer. This technique is advantageous because of a reduction in the thermal budget involved. The MIC process can proceed following two different cases: in the first possibility, nucleation occurs at the metal/silicon interface and the growing silicon layer is formed through the precursor silicon layer as observed in the case of silver/silicon interface [15]. In the second case, nucleation occurs at the metal/silicon interface and the growing silicon layer is formed through the metal as observed for instance at the aluminum/silicon interface [16]. As an example of the MIC process, Aluminum Induced Crystallization (AIC) process has received a great interest over the last years [17]. Basically, the AIC process is based on the fact that amorphous silicon, when in contact with aluminium, transforms into crystalline Si at temperatures below the eutectic temperature of Al/Si system, which is much lower than the temperature required for solid phase crystallization of a-Si. This process was thoroughly applied to a stack of very thin Al and Si layers (200-300 nm) deposited on various substrates such as glass, ceramics or coated metals. More concretely, at the onset of the thermal annealing of the Al/Si layer stack, the interaction of silicon atoms with the Al facilitates the dissociation of the amorphous phase and favors the silicon diffusion inside the Aluminium. Once the Si atoms are dissolved in the metal, they nucleate within the Al thin layer. This cluster grows until it is vertically confined within the finite Al layer by the continuous diffusion of Si atoms. The silicon grain continues to grow in lateral direction up to the entire Al is replaced. At a certain time that depends on many parameters, the layers are exchanged and a continuous poly-Si layer is formed on the substrate in place of the Al layer. This process is also called as “aluminum-induced layer exchange process” (ALILE). Some silicon atoms are also crystallized on the top poly-Si layer in the form of small nano-sized Si (nc-Si) crystallites. These islands can be selectively removed to reach the plane crystallized poly-Si layer. Overall, an optimized AIC process carried out at 450-550°C on glass or ceramic substrates resulted in the formation of thin polysilicon films, 100-200nm thick, exhibiting silicon grains up to 20-30 microns [16]. Furthermore, this thin polycrystalline layer is heavily doped with aluminum (1-2 x10^{18} at/cm^3) and can only serve as a seed for a thicker (2-10 µm) absorbing layer formed by high temperature CVD for instance [17]. Despite its interest, very few work however have concerned the AIC process directly on metal sheets because of contamination issues that is detrimental for solar cells [18]. In contrast, textured aluminum plate has been already applied as a substrate for low temperature hydrogenated amorphous silicon solar cells, exhibiting a conversion efficiency of 8 % [19]. Also, micromorph silicon based solar cells made at temperatures below 200°C directly on Al substrate have demonstrated conversion efficiency of 8.7 % [20]. In this case, the aluminum substrate is used as the back contact as well as a reflective supporting substrate.