Analysis of microstructure and surface morphology of sputter deposited molybdenum back contacts for CIGS solar cells

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Abstract

Magnetron sputter-deposited molybdenum (Mo) thin films are widely used as back electrodes in thin-film copper indium gallium (di)telluride (CIGS) solar cells. In order to fabricate high-efficiency CIGS solar cells, the properties of the Mo back electrode need to be tailored. These properties are associated with the microstructure of the films and thus highly dependent on the deposition conditions. The microstructure of the Mo back electrode is critical to the adhesion and the efficiency of the device. In this work, approximately 900 nm thick single-layer and bilayer Mo films are deposited at different pressure conditions while keeping the power constant at 2 kW. The films are characterized by different methods for a detailed microstructure and morphology understanding. X-ray diffraction measurements reveal that the Mo grain orientation in the [110] direction is enhanced by the bilayer stack design, which is favourable for back contact applications in high-efficiency CIGS solar cells. The surface roughness of the bilayer Mo films is found to increase slightly with increasing bottom layer thickness, and is generally higher than that of single-layer Mo films.

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1. Introduction

CIGS based solar cells are a promising thin-film photovoltaic (PV) technology [1]. One of the most attractive features of CIGS solar cells is the high efficiency and the low-cost potential. Recently, ZSW in Germany has reported a record efficiency of 21.7% [2], which is comparable to the performance of market-dominating multi-crystalline silicon wafer based solar cells [1]. The first step in making a CIGS solar cell is to deposit a metallic rear contact on a glass substrate, which is a critical step for the cell performance. Various metals have been studied as back contact for CIGS solar cells, including Au, Ag, Ni, Mo, Ti, Cu and Al [3-6]. Among these investigated materials, magnetron sputter-deposited molybdenum (Mo) is considered as the best choice of rear contact for CIGS solar cells, because of its low electrical resistivity, high stability and lower contact resistivity with the CIGS layer by the formation of a thin MoSe2 layer at the Mo and CIGS interface [7, 8]. However, the adhesion of Mo films on glass substrates, which is a critical requirement for back contact applications, is determined by the microstructure of the Mo layer [9, 10]. The microstructure of the Mo layer also affects the adhesion of the CIGS absorber to the Mo film [11]. Hence, it is important to tailor the properties of the Mo back contact to fulfill these requirements and to achieve high-efficiency CIGS solar cells.

Several groups have reported the deposition of Mo bilayer stack designs for back electrode applications in CIGS solar cells [12-17]. The most widely studied topic is to find the balance between good adhesion and low electrical resistivity. But there are only a few reports about the influence of the properties of single layers on the performance of the bilayer Mo films. Salome et al. have reported the optimization of Mo bilayer by varying the bottom Mo layer thickness in order to achieve good adhesion, but the overall thickness of the Mo stack is different for each deposition condition [15]. Hence, in this study we made an attempt to prepare bilayer Mo films consisting of different thickness ratios of a high pressure deposited bottom layer and a low pressure deposited top layer, while keeping the total layer thickness constant at ~900 nm. We made an attempt to develop a solid understanding of the effect of the individual layer thicknesses on the overall properties of the bilayer Mo films, such as microstructure and surface morphology.

2. Experimental details

Mo films were deposited onto 3 mm thick soda-lime glass substrates (SLG) by the direct current (DC) magnetron sputtering method (FHR Anlagenbau GmbH, Germany). A planar Mo target was used to deposit the Mo films (size: 9 cm × 51 cm, purity: 99.999%). During deposition, the power applied to the target was kept constant at 2.0 kW and the chamber pressure was controlled by adjusting the Ar gas flow: 330 sccm at 6.0×10^{-3} mbar (high pressure, HP) and 50 sccm at 1.5×10^{-3} mbar (low pressure, LP). The films were deposited at room temperature conditions. Mo films with thicknesses of about 90 nm and about 900 nm were deposited at both high pressure (HP) and low pressure (LP) for detailed characterization purposes. Bilayer Mo films of approximately 900 nm thickness consisting of different thickness ratios of HP deposited bottom layer and LP deposited top layer were also deposited, as shown in Fig.1.

Fig. 1. Schematic of the bilayer Mo back electrode design: The bottom Mo layer was deposited at HP (6.0×10^{-3} mbar) while the top Mo layer was deposited at LP (1.5×10^{-3} mbar). The total thickness of the films was approximately 900 nm.

In order to understand the density of the films, X-ray reflectivity (XRR) measurements were carried out for ~90 nm single-layer Mo films deposited at both HP and LP conditions. Typical XRR measurements were performed at the X-ray demonstration and development (XDD) beam line facility at the Singapore Synchrotron Light Source (SSLS). The experimentally measured XRR curves were theoretically simulated to determine the density (\(\rho\)), roughness (\(\sigma\)) and
thickness \(d\) of the films. X-ray diffraction (XRD; Bruker D8) measurements were carried out to determine the crystal structure of the films. Grazing angle geometry was used to record the XRD patterns of the Mo (900 nm) films, which is very sensitive to the surface structures of thin films. The surface morphologies of the Mo films were investigated by atomic force microscopy (AFM).

3. Results and discussion

3.1. X-ray reflectivity measurement of single-layer Mo films

XRR measurements are carried out for single layer Mo films deposited at LP and HP. Figure 2 shows the XRR measurement curves recorded for these \(-90\) nm thick single-layer Mo films. While comparing the curves of the LP and HP deposited single-layer Mo films, it can be seen that the oscillation amplitude for the Mo film deposited at HP decays rapidly; this is an indication for a high surface roughness of the films deposited at HP. It was observed that the sample deposited at LP possesses a high density \((10.02\, \text{g/cm}^3)\), which is closer to the bulk density of Mo \((10.2\, \text{g/cm}^3)\), and a very smooth surface with a root mean square (RMS) roughness value of 1.58 nm. In contrast, the film deposited at HP possesses a low density \((9.25\, \text{g/cm}^3)\) and a relatively higher surface roughness value \((3.64\, \text{nm})\). This observation can be attributed to the energy differences of the sputtered particles arriving on the SLG substrate at different deposition pressures. At higher deposition pressure, the sputtered particles will have more collisions with the Ar molecules; this leads to a reduced kinetic energy when arriving at the surface of the substrate.

![Fig. 2 XRR measurement of ~90 nm single-layer Mo films deposited at (a) low pressure and (b) high pressure conditions.](image)

3.2. Microstructure measured by XRD measurement

The crystalline structure of the Mo back electrode stacks were studied by XRD in a grazing angle setup. All samples were measured for a fixed inclination angle of \(2^\circ\) and the XRD spectrum was recorded from \(20\) to \(75^\circ\). From the measurements, we found two pronounced peaks centred at \(2\theta = 40.5^\circ\) and \(73.7^\circ\) (see Fig. 3). These two peaks are indexed to Mo \((110)\) and Mo \((211)\), respectively. It should be noted that the single-layer Mo film deposited at LP (Fig. 3a) shows a better crystallization with a stronger and sharper peak at \(2\theta = 40.5^\circ\) compared to the single-layer Mo film deposited at HP (Fig. 3b). This can be attributed to the high kinetic energy of the sputtered particles at LP. In the case of bilayer Mo films, as shown in Fig. 3, they show pronounced \((110)\) preferred orientation while the intensity of the \((211)\) orientation is negligible. The observed preferred orientation in \([110]\) direction is a typical feature for body-
centred cubic (BCC) structures [11]. The grain orientation in [110] direction is more favourable for preparing high-efficiency CIGS solar cells. Previous studies showed that the single crystal of (110) orientated Mo back contact can significantly enhance the (220)/(204) orientations of the CIGS layer, resulting in improved PV efficiency of the CIGS solar cells [18, 19].

![XRD patterns of single-layer and bilayer Mo films.](image)

Fig. 3. XRD patterns of single-layer and bilayer Mo films. The scanning range is centered on the Mo (110) and Mo (211) orientation.

3.3. Surface morphology

The surface morphology of the bilayer Mo films was studied by AFM. The results are shown in Fig. 4. The AFM measurements reveal that, with increasing bottom Mo layer thickness, the root mean square (RMS) surface roughness of the films gradually increases from 7.2 to 10.4 nm. The observed increase can directly be attributed to the roughness of the bottom Mo layer (deposited at HP). It should also be noted that the size of the elongated grains of the bilayer films decreases with increasing bottom layer thickness, which is due to the small grain size of the HP deposited bottom Mo layer.
Fig. 4. AFM 2D images of bilayer Mo films with different thickness ratios of bottom layer and top layer: (a) 90 nm/810 nm, (b) 180 nm/720 nm, (c) 270 nm/630 nm, (d) 360 nm/540 nm and (e) 450 nm/450 nm.

4. Conclusion

The main purpose of this study was to clarify the impact of the thicknesses of the individual layers (HP deposited bottom layer and LP deposited top layer) on the microstructure and the morphology of bilayer Mo films. Bilayer Mo thin films with different thickness ratios of bottom layer and top layer were investigated. From XRD measurement it was observed that compared with single layer Mo films, the bilayer Mo film showed more pronounced grain orientation in [110] direction. Simultaneously the intensity of (211) peak also found decreased. Mo grain orientation in [110] direction is favorable for back contact applications in CIGS solar cells. However, the surface roughness of the bilayer Mo films was found to increase with increasing thickness of the HP deposited bottom layer, but the increase was still within the acceptable limits [20, 21]. The bilayer stack design developed in this study thus seems to have strong potential for improving the efficiency of industrial CIGS solar cells.

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