



Optimization of the properties of the molybdenum back contact deposited by radiofrequency sputtering for $\text{Cu}(\text{In}_{1-x}\text{Ga}_x)\text{Se}_2$ solar cells



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ABSTRACT

In this work, we report on using RF sputtering technique for the deposition of the molybdenum back contact for CIGS solar cells. This allows to realize the back and front contact (Aluminum doped ZnO is used for the front contact) with the same deposition system. We demonstrate that deposition pressure controls the morphology and resistivity of the Mo back contact. This latter point has been the subject of apparently contradicting reports in the literature, but an analysis of the deposition parameters versus sputtering mechanisms allow to understand this spread of results. Full cells have been grown onto the various sets of Mo films, and their electrical properties (efficiencies, Voc, Jcc and fill factors) are analyzed and modeled. From this, we obtain the ideality factors (reflecting the amount of carrier recombination) and with the shunt resistances we find these to be the main parameters which explains the different cells performances versus Mo back contact growth conditions. Low temperature photoluminescence is used as a tool to get a deeper insight on the recombination paths in the samples and support the conclusions drawn from electrical measurements: the molybdenum films grown at 1 Pa yields the best cells results, owing to minimized non radiative recombination. Finally, using the optimum Mo films and improving the quality of the top contact allow us to obtain CIGS solar cells with efficiencies of 17%, although the CIGS, CdS and ZnO layers are not still fully optimized.

1. Introduction

$\text{Cu}(\text{In}_{1-x}\text{Ga}_x)\text{Se}_2$ (CIGS) thin films solar cells are continuously improving in performances, with efficiencies around 22.6%, as reported recently by ZSW in Stuttgart [1]. Usually, molybdenum (Mo) is used as back contact, aluminum doped ZnO (AZO) being used as front transparent contact. AZO is, in the huge majority of cases, deposited by radiofrequency magnetron sputtering while Mo is deposited via DC sputtering. In this study, we were interested to check whether high quality Mo back contacts could be made using RF sputtering, in order to use the same equipment as for the deposition of ZnO (which can be interesting in the laboratory context, or for small batch production), and to check for any cross contamination which could result from using the same equipment for front and back contact.

A large number of studies have investigated the optimum growth conditions for Mo using DC sputtering, since the pioneering work of Scofield [2] while fewer studies report on the use of the RF technique [3–10]. For both deposition techniques, deposition pressure and plasma power have been identified as parameters playing a key role. In particular it is clear that deposition pressure controls the molybdenum resistivity, in a quasi linear manner, for the DC process but in the case of

RF sputtering, very different results have been obtained regarding the role of pressure (see for example [4,6,9]). It is well known that this resistivity change is linked to the film microstructure, which is columnar, the column being separated by boundaries that may be voids or oxide, the important point being that the density and porosity of the film changes with deposition pressure. In the case of CIGS growth for solar cells application, the porosity of molybdenum will favor sodium migration from the glass substrate, which strongly and positively impact the electrical properties of the CIGS grown on top of it [11,12]. An optimum has to be found between a low resistivity for the Mo back contact and sufficient porosity to allow for sodium diffusion in the CIGS.

In this paper, we will demonstrate that a good control of Mo conductivity can be obtained in RF sputtering and propose an interpretation of the mechanisms leading to the apparent spread of results in the literature. Finally, molybdenum deposited by the RF technique will be used in the realization of solar cells and the impact of the Mo deposition process on cell performances will be given.

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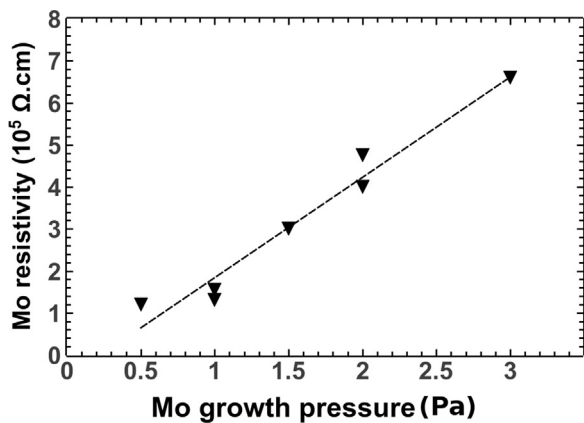


Fig. 1. Molybdenum resistivity versus growth pressure, for a plasma power of 100 W and sample thickness of 500 nm. The dashed line is a linear least squares fit through the experimental values.

2. Experimental

Molybdenum deposition was carried out in a VST services sputtering system, model TFSP-842, equipped with a 13.56 MHz, 600 W RF generator. The equipment is fitted with 3 magnetrons (3 in. in diameter) and is also used for the deposition of undoped ZnO and aluminum doped ZnO in the cells reported here. The pressure for Mo deposition was varied between 0.5 and 3 Pa, using an argon (purity 99.9999%) flow of 20 sccm, with plasma powers of 50 and 100 W. The molybdenum layer thickness has been kept constant at 500 nm in the different samples and cells. Soda lime glass substrate was used as substrate. The system base pressure prior to Mo deposition was $5 \cdot 10^{-5}$ Pa. The electrical properties of the films were measured using an ECOPIA HMS3000 automated 4 point probe system. Morphologies and thicknesses (on cross sections) were determined using a TOPCON ABT-60 SEM system. We had difficulties to obtain clear images of the morphology, although experiments were also duplicated on a TESCAN Vega 3 SEM equipment. We attribute this to the fact that the layers are thin, grown on insulating glass, and the charge buildup reduce the image quality, particularly as the surface detail scale is very small. X-ray diffraction experiments were performed with a Bruker D8 tool equipped with a Gobel mirror on the $\text{CuK}\alpha$ source and a linear detector. Solar cells were grown on top of the molybdenum films by depositing 2 μm of CIGS by 3-stage coevaporation [13] at 540 °C, 50 nm of CdS by chemical bath deposition at 80 °C. The

transparent electrode was formed of RF sputtered undoped ZnO (50 nm, deposited at 3 Pa), then 500 nm AZO (deposited at 0.5 Pa), under argon flow, using the same sputtering system as for Mo. Contacts on the ZnO transparent window are usually made by electron beam evaporation of aluminum. Since we do not have the appropriate tool available and due to the large number of samples grown (12–20 cells for each growth condition), in a first approach top contact was made using a simple indium dot deposited manually. This results in a less efficient carrier collection and minimizes the efficiencies measured (see the spread of series resistances in Fig. 5), but still allow to compare the different samples and growth conditions. Stoichiometry of the CIGS films was monitored using the so-called “End Point Detection” (EPD) process [14], allowing the obtention of a controlled and reproducible copper to group III element ratio in all samples.

The solar cells were characterized at 25 °C under AM1.5 illumination (100 mW/cm²). External quantum efficiency (EQE) measurements were made in the range 350–1300 nm at 25 °C. To further investigate the amount of non-radiative recombination, photoluminescence measurements were performed by exciting the CIGS layer in the solar cell through CdS and ZnO using a 650 nm red solid state laser, sample being kept at liquid nitrogen (LN₂) temperature. A LN₂ cooled InGaAs near IR photomultiplier (Hamamatsu R5509-73) was used to detect the absorber luminescence. External quantum efficiency were measured at room temperature on the solar cells, and the resulting data was fitted according to the procedure described in [15] in order to extract the bandgap energy of the CIGS absorber.

3. Results and discussion

In a first approach, the parameters were tuned to obtain 500 nm thick layers. The growth rate was found to be roughly linear with plasma power and almost independent of pressure, in the investigated range of 0.5–3 Pa. Finally, a power of 100 W was chosen and the role of deposition pressure on the material properties was investigated. In Fig. 1, we report the influence of the pressure on the molybdenum resistivity.

As it can be seen, the resistivity increases linearly with pressure. This is commonly reported for DC sputtering, but there are apparently contradicting information in the literature for the effect of pressure on material properties in RF sputtering: Although most of the papers report an increase of the resistivity with increasing pressure, a very detailed work by Jubault et al. [4] report almost no resistivity change with pressure in the range 2–20 mTorr. It is important to recall here that Mo deposition occurs at low if not ambient temperature; the surface

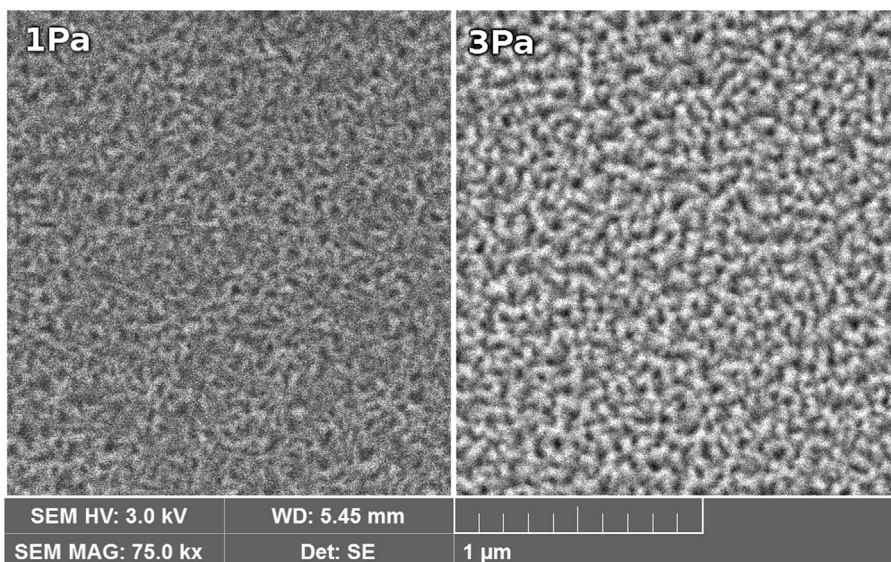


Fig. 2. Surface morphology from scanning electron microscopy for molybdenum layers deposited at 1 and 3 Pa. The layer is clearly more compact at lower deposition pressure.

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