CIWC-2 2020

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Conference Proceedings Paper

Fabrication of Transparent ITO/Ga-Doped ZnO Coating as a Front Panel Electrode toward Efficient Thin Film Solar Cells

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Received: 29 February 2020; Accepted: 12 May 2020; Published: 13 May 2020

Abstract: Bi-layer coatings from sputtered indium tin oxide (ITO) and gallium doped zinc oxide (Ga:ZnO) were investigated for transparency in the visible range of the electromagnetic spectrum, optical rejection ability in the near infrared spectrum and conductivity for the novel quantum dot based solar cells. The multilayer stack produced at optimal oxygen partial pressure exhibit improved optical properties without to worsen the electrical ones, even after additional oxidation during the reactive sputtering of the metal-oxides. With a mean optical transmittance of 91.3% in the visible region, mean optical rejection greater than 65 % in the infrared range and resistivity lower than $0.4x10^{-2}$ Ω.cm, this coating is good candidate for front panel electrode in the CdS/ZnS core-shell quantum dots based solar cells.

Keywords: optical coatings; transmittance; CdS/ZnS core-shell quantum dots

Introduction

Transparent conductive films are important part of any optoelectronic device. Especially in the solar cell fabrication, the efficiency of the cell is strongly dependent on the quality of the transparent conductive electrode (TCE) [1]. It is preferable if the TCE can transmit more than 82-85% of the visible light with broad band of the transmission characteristic and if it can reject the infrared (IR) and ultraviolet (UV) components. Optical losses reduction is especially crucial for the non-silicon, thin film perovskites cells, where the efficiency is naturally low and each percent of loss decrease is significant for the normal work of the cell. To improve the filtering quality of the TCE, the presence of single layer of indium-tin oxide (ITO) is not enough. The deposition method and the deposition conditions are extremely important for the performance of the TCE. Among the possible methods of TCO deposition, such as CVD, spray pyrolysis, e-beam evaporation, ALD, etc., the most applicable process remains the RF sputtering due to its flexibility in the tuning of the films properties with the process parameters [2]. By the main process' parameters – sputtering voltage (power), sputtering pressure, and deposition rate - it is possible to control the main parameters of the TCO that are important for the solar cell work. Researchers firstly have tried to replace the ITO with ZnO films, because of their thermal stability, non-toxicity and lower cost. However, the absolute resistivity of the ZnO still has been too high as compared to the ITO. Metal dopants, such as Al, In or Ga, have been used to improve the electrical conductivity of the films. Recently, ZnO:Ga-graded ITO electrodes in perovskites solar cells have been reported [3]. Optical transmittance of 95%, resistivity

of 2.3×10^{-4} Ohm.cm, and work function of 4.23 eV have been reported after optimization of the power during DC co-sputtering. However, for an efficient solar cell, it is also necessary to achieve rejection of the IR spectrum (the heating component) in addition to the good transmittance in the visible region and the low resistance. By the authors' knowledge similar study has not been carried out. According to the literature, the effect of the oxygen partial pressure during sputtering on the GZO film's roughness and the complex optical characteristic, including visible and IR range, has not been yet investigated. In this paper we tried to fill this gap by preparing GZO/ITO system by RF sputtering of GZO films at different oxygen pressures and comparison of their surface morphology, transmission spectra in the visible range, reflection spectra in the IR range, resistivity and the energy levels alignment with respect to the promising CdS/ZnS core-shell quantum dots based solar cell.

Methods

Thin films of ZnO doped by Ga (GZO), and ITO/GZO were prepared by RF sputtering of 3 inches-diameter targets on glass substrates. The sputtering voltage was kept constant for all sputtered combinations and it was 0.85 kV for the ITO films and 0.7 kV for the GZO films. The total sputtering pressure in the chamber (argon) was 1.10^{-3} Torr for the sample of single layer GZO and bi-layer ITO/GZO without additional oxidation. For the ITO/GZO1 the total sputtering pressure (argon + oxygen) was 1.10^{-2} Torr due to introduction of oxygen, which corresponds to 10% oxidation. For ITO/GZO2 the total sputtering pressure (argon + oxygen) was 1.10^{-1} Torr due to introduction of more oxygen, which corresponds to 20% oxidation. These relations were previously established and proved for the used vacuum installation (Leybold A400VL) in [4]. The thickness of the single GZO film was 100 nm. After insertion of the ITO the total thickness of the system before the additional oxidation was ~ 100 nm and the separated layers' thicknesses were reduced to avoid optical parasitic effects. Further oxidation imposed to decrease the thickness of the GZO film to 40 nm and respectively to increase the thickness of the ITO film to 70 nm. For the highest oxidation degree the thickness of the GZO was kept to 40 nm and the thickness of ITO was further increased to 85 nm. The films' resistance was measured by four point prober FPP-5000 Veeco. The optical transmittance in the UV and visible range was measured by UV-VIS Spectrophotometer NU-T6PC. The optical reflectance in the IR region was measured by Hitachi U-4100 near infrared spectrophotometer.

Results and discussion

The GZO single layer deposited without additional oxidation of the target showed a resistivity of 1.67×10^{-2} Ω.cm. Insertion of ITO resulted in a resistance decrease to 0.03×10^{-2} Ω.cm. It is known that the electrical properties of the GZO films are improved by introducing ITO film [5]. However, due to the additional oxidation of GZO and the resulting increase of its electrical resistance, it is not possible to keep one and the same thickness for both films. The resistivity of the double ITO/GZO layer increased with increasing the oxygen content during sputtering, however, due to the presence of ITO the variation of the resistance is not great - 0.12×10^{-2} Ω.cm at 10% oxidation and 0.36×10^{-2} Ω.cm at 20% oxidation (fig. 1).

The transmission spectra in broad wavelength region between 190 nm and 800 nm were measured as a function of the oxygen content and the results were shown in fig. 2. It was found that the mean optical transmittance of the strongest oxidized film in the visible region was 91.3% and the transparency for the UV component was approximately 6%. For all GZO coatings the spectra in the near UV region (<400 nm) exhibited very sharp absorption edge. A slight shift of the optical pattern maximums and minimums to the longer wavelengths is noted for the strongest oxidized film ITO/GZO2 as compared to the rest films. This is in consistent with the literature [6] and can be explained with the increased thickness, which is the greatest for this film. Single layer of GZO showed 91 % optical transparency (excluding the glass substrate) in the same region and UV transmittance close to zero, however, its electrical resistance was not suitable for electrode purposes. Then insertion of ITO sublayer without additional oxidation of GZO during sputtering resulted in a slight worsens

of the transmission spectrum in the visible range, resulting in 90.9 % of transmission. Further increase of the oxygen pressure, resulted to 91.2% and 91.3% transmittance.

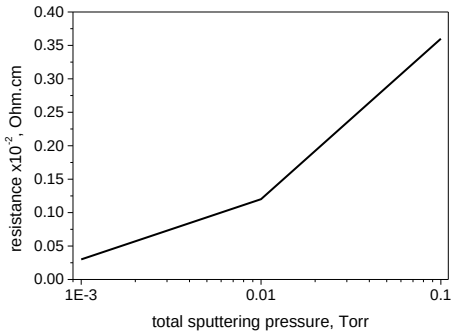

Figure 1. Electrical resistance of the bi-layer system ITO/GZO at different total sputtering pressures due to additional target oxidation.

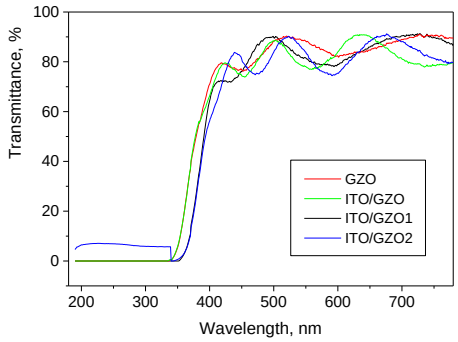

Figure 2. Optical transmittance in the UV-VIS range of single layer of GZO and bi-layer coatings ITO/GZO without and with additional oxidation during sputtering.

Although the difference in the spectra from the visible range is negligible, a significant difference could be noted in the infrared range (fig. 3), where the reflection of the near infrared (NIR) component is crucial for the efficiency of the solar cells.

The influence of the additional oxidizing is clear in the reflectance data in the NIR region. It can be seen that the single layer GZO and ITO/GZO without oxidation exhibited relatively low rejection ability of the IR component – the mean reflectance is 15-20%. Discrimination of the IR wavelengths transmission and optical (respectively heat) rejection greater than 65 % can be seen for the ITO/GZO2 films.

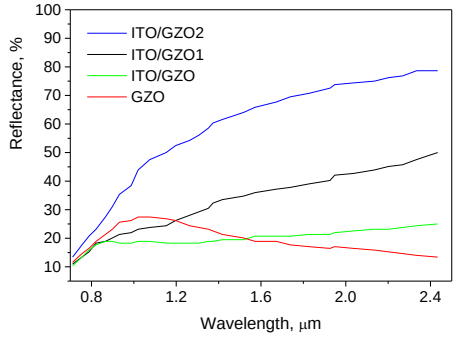

Figure 3. Optical reflection in the NIR range of single layer of GZO and bi-layer coatings ITO/GZO without and with additional oxidation during sputtering.

Conclusion

ITO/GZO by-layered coatings were deposited on glass substrates by RF sputtering at various oxygen contents. The electrical and optical properties of the bi-layer system were strongly affected by the oxidation degree of the GZO layer. The optical data showed that the additionally oxidized GZO films with ITO underlayer exhibited slightly differed mean visible transmittance over 90%, but the difference of the reflection spectra in the IR range is more significant In summary, ITO/GZO2 could serve as a transparent conductive electrode and partially as a heat mirror, or sunshade coating. The future work will be related to impedance spectroscopic study for detailed estimation of the contact properties of solar cells implementing ITO/GZO as a front electrode.

Acknowledgment: This work is supported by the programme for bilateral cooperation Bulgaria-India KP06-India-6.

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