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# Sputter-instigated plasmon-enhanced optical backscattering layer in ultrathin solar cells: Application of GZO in CIGSe material system



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#### ABSTRACT

Recently, realization of ultrathin solar cells is the area of interest of researchers in the domain of cost-effective photovoltaics. This study demonstrates a novel way of generation of plasmonic features in transparent conducting oxide material in the form of Ga-doped ZnO (GZO) thin films to compensate for the loss of optical absorption due to reduced absorber thickness. Through an extensive analysis of photoelectron spectroscopy, spectroscopic ellipsometry, and field emission scanning electron microscope measurements the evaluation of plasmonic features and correlation of them with various metallic and metal-oxide nanoclusters inside GZO thin film and GZO/CIGSe heterojunction interface are carried out. Moreover, we have thoroughly analyzed the applicability of GZO plasmon enhanced thin film as a backscattering layer based on (a) verification of plasmonic behavior in GZO film ( $\sim$ 150 nm), (b) checking on the sustainability of such plasmonic behavior in ultrathin GZO ( $\sim$ 5 nm) layer, (c) investigation of plasmonic feature at the heterojunction, (d) band offset studies at the plasmon-enhanced-GZO/CIGSe heterojunction, and (e) investigating the electrical performance of the junction to verify the linear behavior and resistivity calculation of the heterojunction.

#### 1. Introduction

Cu(In, Ga)Se2 solar cells are promising candidates and have demonstrated photon conversion efficiency value up to 22% (Jackson et al., 2016). Currently, the typical thickness of CIGSe thin films in the thin film solar cell structure is  $\sim 2-3 \,\mu\text{m}$ . It has been proposed that the high efficiencies (> 15%) can only be maintained when the CIGSe absorber is thicker than 1 µm (van Lare et al., 2015a, 2015b). The dominant reason behind the reduction in efficiency is the incomplete absorption of incident light arising from the reduced absorber thickness, which leads to inferior current density. To overcome this scaling limit, exceptional attention has been paid to ultrathin solar cells with thinner absorbers, which in comparison to the thick counterparts, enables the reduction in (a) consumption of rare material such as Indium and (b) resulting manufacturing cost. The ultimate aim is to achieve highly efficient ultrathin CIGS solar cells (with CIGS layer thickness lower than 500 nm). Therefore, light trapping mechanism becomes extremely vital to maintain high efficiency even in ultrathin solar cells. Among numerous light-trapping techniques, metallic nanostructures with sub-wavelength plasmonic features have shown distinct lighttrapping effects in thin film solar cells because they can exhibit either strong scattering or local near-field concentration leading to generation of localized surface plasmons and improves the optical absorption in solar cells (Dabirian and Taghavinia, 2015; de Aberasturi et al., 2015; Hornich et al., 2016; Schmid et al., 2016; Zhou et al., 2015). Few reports on possible options to incorporate plasmonic nanoparticles (noble metals) in thin film solar cell structures are: (a) within transparent conducting oxide (TCO) layers, (b) in absorber layers, and (c) as a plasmon-enhanced backscattering layers (Atwater and Polman, 2010; Garg et al., 2016; Pillai and Green, 2010). Ultrathin metal layers and nanoparticles of noble metals such as Au, Ag, Cu, etc., support collective oscillations of free electrons (plasmonics) at optical frequencies (Garg et al., 2016; Hobbs et al., 2016). Nanostructures of different noble metals have shown tremendous potential in different approaches of light management schemes. The ability of sustained, coherent electronic oscillations in noble metal leads to electromagnetic field confinement and coupling have made plasmonic augmentation an area with very high prospects for light harvesting in photovoltaic devices, photodetectors, optical sensing, drug delivery, and imaging (Echtermeyer et al., 2016; Gjonaj et al., 2013; Hutter and Fendler,

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2004; Kang et al., 2013; Mubeen et al., 2014; Senanayake et al., 2011; Willets et al., 2017). However, few technical and fabrication related issues act as an obstacle to the realization of plasmonic-based commercial grade devices. One of the most prominent fabrication challenges is the compilation of scalable and economically viable techniques for controlled nanostructure patterning. The fact that plasmonic resonances are highly dependent on both shape and geometry of the nanostructure means that design parameters should be precisely controlled throughout the fabrication process.

Few studies are being pursued to characterize the optical properties of various alternative plasmonic materials, and several reports are available worldwide such as metal alloys (Gong and Leite, 2016), transition metal-nitrides (Naik et al., 2011), and heavily doped semiconductors (Calzolari et al., 2014; Kim et al., 2015; Kim, J. et al., 2013b; Pradhan et al., 2014; Zhang et al., 2014). Recent studies have demonstrated that TCOs such as ZnO: Al (AZO), ZnO: Ga (GZO) are promising candidates as plasmonic materials because they exhibit metallic behavior and smaller losses as compared to those for noble metals (Calzolari et al., 2014; Kim, H. et al., 2013; Kim et al., 2015; Kim, J. et al., 2013a; Pradhan et al., 2014; Zhang et al., 2014). So far, there are only limited successful demonstrations where TCOs have been used as plasmonic materials, such as in semiconductor plasmonic quantum dots, and plasmonic modulators (Faucheaux et al., 2014; Kim, H. et al., 2013; Kim et al., 2015; N'Tsame Guilengui et al., 2012). However, the region of interest in the electromagnetic domain for photovoltaic and photodetector devices is the ultraviolet (UV)-visible (Vis)-infrared (IR) region. Therefore, the generation of widely tunable plasmonic features in TCOs, which are the integrated parts for a broad range of applications such as solar cells, photodetectors, etc., and incorporation of these TCOs in the solar cell structure, is the motivation for this study.

Here, we report a novel approach to generate plasmons in GZO thin films. By utilizing dual-ion beam sputtering (DIBS) system, plasmonic generation in TCOs in the wide spectral range of 1.87–10.48 eV is reported in our previous studies (Awasthi et al., 2017, 2016, 2015; Garg et al., 2018). In continuation with our earlier studies on sputter-instigated plasmon generations, we have further extended our investigation on assessing the application of plasmon enhanced TCO layer

in thin film solar cell material system as a backscattering layer as depicted in schematic of the device structure shown in Fig. 1(a). Additional studies have been performed including (a) verifying the plasmonic behavior in GZO thin film ( $\sim 150\,\mathrm{nm}$ ), (b) confirming the sustainability of such plasmonic behavior in ultrathin GZO ( $\sim 5\,\mathrm{nm}$ ) layer, (c) investigating the plasmonic features at the heterojunction, (d) band offset studies at the plasmon enhanced-GZO/CIGSe heterojunction, and finally (e) investigating the current-voltage (I-V) characteristics for the verification of linear behavior and resistivity calculation of the heterojunction.

#### 2. Experimental section

CIGSe and GZO thin films with a thickness of 150 nm are deposited at 300  $^{\circ}$ C on Si and glass substrates using DIBS system by utilizing commercially available CIGSe (99.99% pure) and GZO target inside the DIBS chamber, utilizing a radio frequency (RF) primary deposition source. In addition to the RF primary ion source, direct-current coupled assist-ion source, consisting of a plasma of Ar+ ion, is turned on to reduce columnar growth and thereby boost growth uniformity and the film adhesion. Additionally, in order to realize the GZO/CIGSe heterojunction, a 5 nm layer of GZO is grown on 150 nm thick CIGSe thin films. A detailed description of the DIBS deposition system is reported elsewhere (Garg et al., 2018; Kumar et al., 2017).

The crystalline quality of GZO and CIGSe thin films are analyzed using Rigaku SmartLab system with Cu K $\alpha$  radiation (1.54 Å). For studying the morphological properties of samples, field-emission scanning electron microscope (FESEM) based on Supra55 Zeiss lens is used. Elemental properties, band offset and plasmonic properties of the samples are examined by ultraviolet photoelectron spectroscopy (UPS). X-ray photoelectron spectroscopy (XPS) measurements are performed to analyze the core level peaks for band offset analysis. Optical properties, such as dielectric functions, dispersion curves, and optical absorption are analyzed by using M-2000D J.A. Woollam spectroscopic ellipsometry (SE). I-V measurements are performed using Keithley 2612 source meter and Everbeing cryogenic probe station.

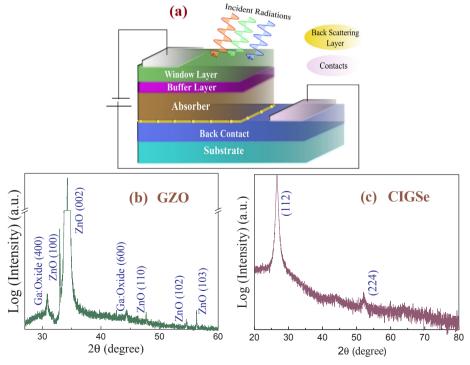


Fig. 1. (a) schematic diagram of the device structure with a backscattering layer, XRD spectra of (b) GZO and (c) CIGSe thin films, respectively.

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