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journal homepage: www.elsevier.com/locate/solmatBand diagram construction of CdTe/Sb₂Te₃ interface using synchrotron radiation

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ABSTRACT

The CdTe/Sb₂Te₃ interface was prepared by co-evaporation on an ultra-thin glass substrate and studied “*in-situ*” by photoelectron spectroscopy using a tunable synchrotron radiation photon source. The results indicated that the as-deposited antimony telluride was a mixture of Sb–Te phase and Sb₂Te₃ structure. The band diagram showed a conduction band offset (ΔE_{CB}) of 0.65 eV at the interface. A thin layer of Sb₂Te₃ came into being on the surface of CdTe with a subsequent 300 °C anneal. The sputter depth profile suggested that the annealed interface was actually constituted by the “substrate CdTe/interface Sb₂Te₃ donors/inner Sb–Te mixture/surface Sb₂Te₃” structure. The band diagram gave ΔE_{CB} = 1.35 eV for the annealed interface. It was found that there was a thin barrier of 0.2 eV induced by Sb₂Te₃ donors for the hole transportation at the interface. To create a thin p⁺ layer between CdTe and the back electrode, a 50 Å thick Sb₂Te₃ layer was used for the back contact to CdTe thin film solar cells. *J–V* characteristic showed that this thin p⁺ back contact greatly enhanced the cell performance.

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1. Introduction

CdTe thin film solar cells have been widely investigated because of their high efficiency and low cost [1–7]. The highest conversion efficiency reported till now is 20.4% [8]. However, forming an ohmic contact to CdTe is an issue due to its high work function and self-compensation. One method to overcome this problem is to introduce a p⁺ region that provides required electrical properties between CdTe and the metal back electrode. Usually copper or copper-containing material is incorporated to form this p⁺ region, but CdTe solar cells with such back contacts are not stable because of the diffusion of Cu and the formation of the related defects cross the junction [7,9]. It is found that Sb₂Te₃ could form a stable back contact to CdTe, and an efficiency of 14.6% for CdTe solar cells with Sb₂Te₃ has been reported [6,9–12]. Our previous work has built the band diagram of CdTe/Sb₂Te₃ interface prepared under normal conditions (*ex-situ*, 6×10^{-4} Pa) using XPS (Al K α) and UPS (He I, $h\nu$ = 21.2 eV) and emphasized the role of Sb in CdTe thin film solar cells [13].

In this paper, we would build the band diagram of this interface fabricated under much more constrained conditions (*in-situ*, 1×10^{-7} Pa) using the synchrotron radiation technology firstly. The *in-situ* fabrication could get rid of the reactions with oxygen and unknown contaminants as a result of exposure to air in the course of the sample handling. Unlike the traditional light which

could only radiate unchangeable energy, the synchrotron radiation light is tunable over a large energy range, thus it provides the way to maximize the photoelectron productivity by tuning the incident energies to the binding energies of target atoms, in addition to its high brightness. It is synchrotron radiation photoelectron spectroscopy that has large advantages in terms of fine energy resolution and high surface sensitivity [14,15]. Then we would focus on improving the band diagram of the CdTe/Sb₂Te₃ interface to enhance the device performance by utilizing synchrotron radiation photoelectron spectroscopy. Our previous paper discussed in detail the role of Sb in CdTe solar cells [13], but mentioned few on the modification of the interface. Actually, by carefully controlling the thickness of Sb₂Te₃ layer it forms a stable p⁺ layer between CdTe and the back electrode and allows the carriers to tunnel through as well. To determine the optimal thickness of the primary contact layer, we would build the CdTe/Sb₂Te₃ interface band diagram by firstly stepwise deposition and a subsequent sputter depth profile. Finally, Au electrode [11,12] as the secondary contact was evaporated on the Sb₂Te₃ layer to complete the fabrication of CdTe solar cells on the ultra-thin glass substrates.

2. Experimental details

Glass/ITO/CdS/CdTe sample used in this study was produced at our lab. The CdS and CdTe layers were deposited by chemical-bath deposition and close-spaced sublimation onto 0.2-mm-thick borosilicate glass to nominal thickness of ~180 nm and ~2.26 μ m, respectively.

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This was followed by a wet CdCl_2 treatment. It should be noted that ultra-thin glass substrates in this work are used for synchrotron-radiation-measurement convenience. The contact formation of CdTe solar cells was investigated in an UHV system at the Surface Physics End Station of National Synchrotron Radiation Laboratory of the University of Science and Technology of China. The surface analysis chamber equipped with an argon ion gun was directly connected to a preparation chamber, which allowed for different layers preparation and the sample transfer without exposure to air. Prior to the Sb_2Te_3 deposition, the sample surface was etched with a mixture of nitric and acetate acid to remove the surface contaminants, and was then transferred immediately into the analysis chamber to clean the surface with an AG5000 ion sputtering gun. Due to the large difference in the saturated vapor pressures of components direct vacuum evaporation or sputtering of the bulk material is not appropriate, resulting in a compositional gradient along the film thickness or a re-sputtering effect of the constituent elements of the targets alloys [16,17]. Thereafter, Sb_2Te_3 was stepwise deposited onto the CdTe surface by co-evaporation with a base pressure of 1×10^{-7} Pa. The temperature of the Sb and Te (both 99.999% purity from Alfa Aesar) were maintained at 355 °C and 142 °C, at which the deposition rates were measured at 0.7 Å/min and 1.1 Å/min, respectively. A subsequent 300 °C anneal at the rate of 10 °C/min was conducted on the sample after a time of 64 min deposition, and then a sputter with up to 3 keV argon ion without artificially changing the composition at 0.0015 mA/cm² giving a typical sputter rate of 0.5 Å/min was performed. The 75 eV synchrotron radiation beam line was used to characterize the clean CdTe substrate and the specimen after each single Sb_2Te_3 deposition and sputter step. The photoelectron initial kinetic energy was detected by a VG ARUPS10/2 angle resolved electron energy analyzer. All the spectra were calibrated with the core level line of Au 4f_{7/2} emission peak (83.8 eV). Finally, 300-nm-thick Au was evaporated on the Sb_2Te_3 layer (with 30 min deposition and then 300 °C annealing) to complete the preparation of CdTe solar cells on the ultra-thin glass substrates. The resulting photovoltaic devices were characterized using the light *J-V* measurement under simulated AM1.5, 100 mW/cm² illumination at 25 °C.

3. Results and discussions

3.1. As-deposited CdTe/Sb₂Te₃ interface

Fig. 1 shows the Te 4d, Sb 4d and Cd 4d emissions of the step-by-step deposited Sb_2Te_3 films onto the clean CdTe substrate layers. Being contained both in the substrate layer and overlayer,

Te can be detected from the very beginning of the deposition to the end, in the course of which the binding energies (BEs) of Te 4d vary little, only 0.15 eV to the lower end from the initial 39.4 eV. The Cd 4d signal decreases with adding coverage of Sb_2Te_3 . Correspondingly, the Sb 4d intensity increases. The emission shapes do not change with coverage, indicating no chemical shifts. However, the core level line of Sb 4d shifts 0.55 eV to the lower binding energy from the initial 32.15 eV, whereas the binding energies of Cd 4d vary little. Substrate signals, namely Cd 4d emission, are no longer observable for the largest coverage of 64 min, *i.e.*, the Sb_2Te_3 layer caps the substrate completely. The valence band spectra for the sample after stepwise deposition of Sb_2Te_3 thin films on CdTe are also given in Fig. 1. By linearly fitting to the photoemission onset at low binding energy end, the valence band maximum (VBM) was determined. It shows the VBM shifts 0.65 eV gradually from the very beginning 0.2 eV to a lower binding energy with the final deposition of Sb_2Te_3 .

To confirm the valence band offset the binding energies of the core level lines with respect to the VBM of bulk materials have to be determined. The binding energy with respect to VBM, $E_{\text{B}}^{\text{VBM}}$, has been determined to be 10.3 eV for Cd and 32.05 eV for Sb, respectively. Using these data, the evolution of VBM referring to the Fermi level, $E_{\text{VBM}}-E_{\text{F}}$, was extracted. The clean substrate CdTe and thick Sb_2Te_3 overlayer were taken as reference points. The results are shown in Fig. 2. Due to the coverage of Sb_2Te_3 the CdTe E_{F} approaches rapidly to VBM from the beginning 0.2 eV after 4 min deposition, while the Sb_2Te_3 E_{F} separates fast from VBM. A further 0.2 eV of $E_{\text{VBM}}-E_{\text{F}}$ is observed for 4 to 16 min deposition. These values are in good agreement with our previous work [13]. These shifts are reasonable. For CdTe growth and surface etching the CdTe/Sb₂Te₃ interface are Te rich. In such surrounding Sb can substitute Cd to form low-energy $\text{Sb}_{\text{Cd}}^{\text{d}}$ donors [1]. These donors would compensate the V_{Sb} acceptors in the p⁺-type Sb_2Te_3 and therefore decrease the doping level of the Sb_2Te_3 overlayer. However, the $E_{\text{VBM}}-E_{\text{F}}$ does not change much with further deposition. Meanwhile, no $E_{\text{VBM}}-E_{\text{F}}$ from Cd side could be derived from Fig. 1 due to the dying of Cd 4d emission. That means the thickness of the cover layer exceeded the space charge region of the CdTe/ Sb_2Te_3 heterojunction and the surface sensitive photoelectron could not be affected with further deposition.

The valence band discontinuity, ΔE_{VB} , between the CdTe substrate layer and the Sb_2Te_3 overlayer is evident as shown in Fig. 2, which is determined to be 0.5 ± 0.1 eV using the statistical average from all points over 0.5 min and 4 min. The principal experimental uncertainty and the scatter of the individual data points were both 0.05 eV. Therefore the error margin was determined to be 0.1 eV.

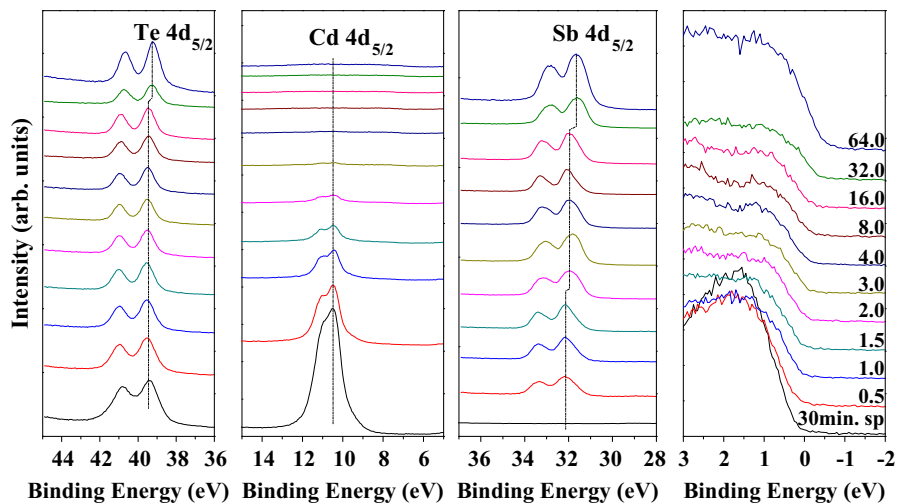


Fig. 1. Te 4d, Sb 4d and Cd 4d emission lines of the step-by-step deposited Sb_2Te_3 films onto the clean CdTe substrate layer as well as the valence band spectra.

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