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Metal-oxide broken-gap tunnel junction for copper indium gallium diselenide tandem solar cells

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ABSTRACT

A metal-oxide broken-gap heterojunction between p-Cu_2O and n-In_2O_3 is proposed and studied for use as a tunnel junction in polycrystalline $CuIn_{1-x}Ga_xSe_2$ (CIGS) based tandem solar cells using numerical device simulation. Specifically, a tandem solar cell with a $CuGaSe_2$ (CGS) absorber top cell and a $CuInSe_2$ (CIS) absorber bottom cell was considered. The ballistic transport model explains well the carrier transport in the broken-gap heterojunction. Broken-gap heterojunctions provide linear current–voltage characteristics even if one side is lightly doped and the junction resistance is much lower than typical values of series resistance in solar cells. However, electron affinities of metal oxides for broken-gap band alignment may induce an energy barrier between the tunnel junction and the top and bottom solar cells, degrading performance. These barriers can be reduced using buffer layers. For example, NiO and graded In_2O_3–ZnO buffer layers are proposed beneath CGS top solar cell and above the CIS bottom solar cell, respectively. With these buffer layers, the efficiencies of the top and bottom cells are 17.5% and 6.5% respectively. The modeled efficiency of the CGS/CIS tandem solar cell is 24.1% and there is virtually no significant efficiency loss due to the presence of the broken-gap junction.

1. Introduction

Thin film multi-junction solar cells are among the most promising structures for third-generation photovoltaic (PV) devices because they have the potential to convert sunlight to electricity with high efficiency and at low cost [1,2]. Single-junction thin film solar cells show low cost [3] and high efficiency [4] compared to other solar cells. However, single-junction solar cell efficiencies are constrained by the Shockley–Queisser limit [5]. Further improvements require the use of multi-junction PV device architectures [1]. Crystalline multi-junction solar cells hold the world record efficiency [6], however, they are expensive to produce due to the need for epitaxial growth [7]. It would be desirable to make multi-junction solar cells based on low-cost thin polycrystalline films such as CdTe and $CuIn_{1-x}Ga_xSe_2$ (CIGS).

In multi-junction PV devices two or more solar cells share the solar spectrum. The top cell should be transparent below the bandgap of its absorber and connected to the bottom cell with transparent contacts. When two solar cells are stacked monolithically, a reverse biased p–n junction forms at their interface. Typically tunnel junction (TJ) diodes are used between the solar cells to solve this problem.

This connection is one of the obstacles to making multi-junction solar cells based on polycrystalline films. The TJ requires a very heavily doped p–n junction with steep doping profiles at the interface [8]. However, the dopant concentration profile control in polycrystalline films is much more difficult to achieve owing to fast grain boundary diffusion [9,10]. Furthermore, some of the top cell layers require a high temperature deposition process to produce large grains and low trap densities. Thus, significant dopant diffusion in the bottom solar cell and inter-diffusion near the tunnel junction can occur. Furthermore, doping in most polycrystalline PV materials such as CIGS occurs via point defects, making precise doping concentration control very difficult and compensation common. Finally, it is impossible to form heavily doped layers of both types due to compensation effects [11,12]. To avoid these difficulties, multi terminal methods have been demonstrated [13,14]. In CIGS solar cells, $MoSe_2$ on ZnO:Al, SnO:F and In_2O_3:Sn (ITO) make good ohmic contacts with CIGS absorber, however low transmittance is a problem [15–17]. More fundamentally, tandem devices formed by stacking cost more since one loses the advantages of monolithic integration.

In this article, we examine the possibility of using metal-oxide broken-gap heterojunctions as the tunnel junction in multi-junction CIGS solar cells. We investigated the I–V characteristics of p-Cu_2O/n-In_2O_3 junction. Early experimental results show ohmic behavior at a similar junction made from p-Cu_2O/n-$ZnSnO_3$ [18]. Here we also investigate monolithic CIGS-based tandem PV device with p-Cu_2O/n-In_2O_3 junction where the top and bottom cell absorbers were $CuGaSe_2$ (CGS) and $CuInSe_2$ (CIS) respectively, including the band

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alignment of broken-gap heterojunctions with top and bottom solar cells.

It is important to point out that major obstacles exist to realizing a high-efficiency tandem cell. The top (i.e. wide bandgap) cell is critical to the efficiency. Its larger V_{OC} means that it must provide about two thirds of the total power. CGS has a suitable band gap for the top solar cell in tandem devices. However, the best efficiency of CGS solar cells is only about 10%. This is due to a large bulk trap density [19] and poor band alignment between CGS and the buffer layer [20,21] which lead to a low open circuit voltage [22,23]. A second obstacle is thermal budget. The deposition of a low defect density wide bandgap absorber typically requires high temperature. A pore structure forms in the CdS upon annealing leading to a heavy diffusion of Ga into ZnO layer. Ga-assisted Cd diffusion may be responsible for the pore formation [24]. Furthermore, diffusion of Cd in the lower device counter dopes the absorber leading to a severe drop in open circuit voltage once the device is fully depleted [25]. The next possible problem is the use of CIS in the lower device. Although CIS has not demonstrated an efficiency as high as CIGS, Solar Frontier recently demonstrated a low Ga content 20.9% cell with a bandgap of only 1.05 eV [26]. Since this low concentration of gallium only increases the desired top cell from 1.65 to 1.70 eV, this problem has largely been resolved. As a result of the first two technical limitations, the device efficiency predicted here cannot be realized today. However, progress is being made. Not only are the efficiencies of CGS solar cells rising [27,28], but alternative wide bandgap CIGS-based materials are also being explored [29]. Shafarman has been developing $Cu_xAg_{1-x}In_yGa_{1-y}Se_2$ [30] and has recently demonstrated relatively high efficiency 1.4 eV devices. We have demonstrated very low trap density absorbers at 1.6 eV using UHV-deposited $CuIn_xAl_yGa_{1-x-y}Se_2$ [29,31], but have not yet demonstrated a high efficiency device. In this work we focus on the effects of the broken-gap heterojunction under high performance conditions.

2. Metal oxide broken-gap heterojunction

While high conductivity transparent conducting oxides (TCOs) [32,33] are used as window layers in solar cells [34] they are difficult to heavily dope both electrons and holes making them poor candidates for tunnel junctions [35,36]. When the doping concentration is increased and Fermi level reaches the formation energy of a compensating defect, the net carrier concentration saturates in an effect called pinning [21]. Thus, one cannot form a heavily doped p-type film from materials such as ZnO, SnO_2, In_2O_3, Ga_2O_3, and CdO. Similarly, one cannot form a heavily doped n-type film from Cu_2O, NiO and $CuMO_2$ (M=Al, In, Cr and Ga).

It is possible to use a heterojunction of an n-type and a p-type metal oxide for a tunnel junction. A broken-gap metal-oxide heterojunction is a promising design. This involves a junction between two dissimilar materials where the conduction band edge of the n-type material is equal to or lower in energy than the valence band edge of the p-type material as shown in Fig. 1. Carriers can easily move across an interface with this band alignment by a nonlocal interband transition. As a result the junction has linear current–voltage (I–V) characteristics [37] even without heavy doping on both sides.

The electrical characteristics of the broken-gap heterojunction have been studied extensively in InAs/GaSb junctions [37–41]. The valence band edge of GaSb is between 0.48 and 0.53 eV higher than that of InAs. The band overlap ($\Delta\chi$ as shown in Fig. 1) is between 0.12 and 0.17 eV since band gaps of InAs and of GaSb are 0.36 eV and 0.71 eV, respectively [38]. The transport properties depend on the band bending near the junction interface. When an n-InAs/p-GaSb approaches flat band, the I–V characteristics are linear [38–40]. A resistance as low as $10^{-7}\ \Omega\,cm^2$ can be achieved [41] suggesting no significant contribution to R_{series}.

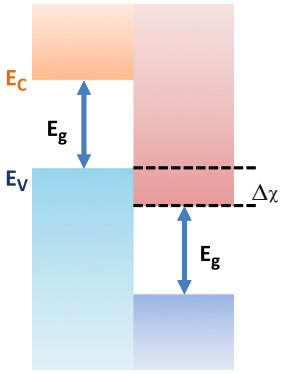

Fig. 1. Band alignment of broken-gap heterojunctions.

When designing a broken-gap tunnel junction, the electron affinities of the materials are the most important properties. Low and high electron affinities are desired for the n- and p-type materials, respectively. P-type TCOs are rare. Cu_2O and $CuAlO_2$ are two of the most attractive materials for broken-gap tunnel junctions since they have the lowest electron affinities among the p-type TCOs. In n-type TCOs, SnO_2, $GaInO_3$ and $ZnSnO_3$ are the most likely to form broken-gap band alignment with other TCOs for a similar reason [33].

There are few studies on broken-gap heterojunction with metal oxide films. Tanaka et al. studied the heterojunctions formed between Cu_2O and several different n-type TCOs including ZnO, ITO and In_2O_3 [42]. Unlike junctions with other n-type TCOs, the p-Cu_2O/n-In_2O_3 junction showed linear I–V characteristics while the p-Cu_2O/n-ITO junction showed nonlinear I–V characteristics with a low energy Schottky barrier [42]. The band alignment of these ohmic p-Cu_2O/n-In_2O_3 junctions was not studied, however it can be estimated from the I–V results and the band alignment of the Cu_2O/ITO junction, and the electron affinity difference between ITO and n-In_2O_3. Recently, Deuermeier et al. analyzed the band alignment of p-Cu_2O/n-ITO by X-ray photoemission spectroscopy (XPS). They found that the conduction band edge of n-ITO is 0.2–0.7 eV higher than the valence band edge of p-Cu_2O depending on the ITO deposition conditions [43]. According to research results from multiple groups [33,44] the work function of In_2O_3 is 0.2–0.8 eV lower than that of ITO. From these results, it can be estimated that p-Cu_2O/n-In_2O_3 junctions have a broken-gap band alignment. The linear I–V characteristic supports this inference.

3. Device simulation

Numerical simulation for the tunnel junction in a complete monolithic tandem solar cell requires current continuity and the Poisson equation, as well as a self-consistent treatment of quantum mechanical transport in the tunnel junction. However, well-known solar cell simulations do not have this capability. Thus, previous tandem device modeling has simply treated the tunnel junction as an ohmic contact between the top and bottom solar cells [45,46]. Here we use DESSIS – a commercially available multi physics device simulation tool for numerical simulation. Quantum transport at the broken-gap junction is solved by the Wentzel–Kramers–Brillouin (WKB) approximation. Device simulation is performed by self-consistently solving the current continuity equation, the Poisson equation and WKB approximation near the broken-gap junction interface. Optical absorption in the stacked multiple layers is calculated by transfer matrix methods [47].