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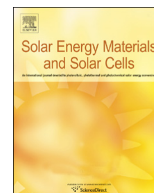

# Sputtered metal oxide broken gap junctions for tandem solar cells

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ABSTRACT

Broken gap metal oxide tunnel junctions have been created for the first time by sputtering. Using a ceramic ZnO–$SnO_2$ target and a reactively sputtered copper target we deposited $ZnSnO_3$ and $Cu_2O$ for the n-type and p-type layers, respectively. The band edges and work functions of these materials are suitable for favorable alignment with the bands of copper indium gallium selenide (CIGS) for a tandem CIGS-based solar cell applications. Total junction specific resistivities under 1 Ω-cm$^2$ have been achieved with Ohmic current–voltage (*I–V*) characteristics pointing to a broken gap band alignment. Low temperature *I–V* measurements confirmed the lack of traps at the interface despite other measurements pointing to an interface where bands overlap. $Cu_2O$ films contained copper inclusions, but they were shown, by conductive atomic force microscopy, not to be the dominant paths for conduction across the junction. Post-deposition annealing of junctions demonstrated thermal stability up to 300 °C, and the ability to improve conduction and influence device material's electron affinity by rapid thermal anneal (RTA). Optical transmission over 78% below a band gap of 2.4 eV was attained for as-deposited films.

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## 1. Introduction

Increasing the performance of photovoltaic (PV) devices is extremely important for their widespread use for solar-to-electric energy conversion. Unless multiple exciton generation or hot electron extraction provides an alternate route, the conventional PV devices, based on a single semiconductor absorber, are constrained by the Shockley–Queisser efficiency limit [1]. This limit is approximately 32% for a single-junction device constructed with a semiconductor absorber that has the ideal band gap (~1.1–1.4 eV). Tandem two-junction solar cells, which can achieve up to 42% efficiency [2] are of increasing interest as the performance of single-junction solar cells approaches this limit [3]. Unconcentrated epitaxial tandem solar cells have demonstrated an efficiency of 31.1% [4]. However, tandem devices must be made of inexpensive polycrystalline thin films to lower their cost. Of the existing and commercialized high-efficiency thin-film PV technologies, $CuIn_xGa_{1-x}Se_2$ (CIGS) has the highest efficiency. Various tandem solar cells based on CIGS and related materials have been proposed. Modeling that makes realistic assumptions about traps and various parasitic effects indicates that efficiencies as high as ~30% can be achieved as identified by Song et al. and others [5–7].

A key issue for the fabrication of a monolithic tandem cell is the series electrical connection between the two p–n junctions that allows charge carriers to move across this junction via tunneling to provide continuous current flow between the two devices. Holes from the top solar cell and electrons from the bottom solar cell flow towards each other and recombine, providing current continuity. A metal layer is the simplest and would allow recombination, but metals block light transmission to the lower cell. Epitaxial tunnel junctions have been deposited by molecular beam epitaxy (MBE) [8] and metal organic chemical vapor deposition (MOCVD) [9], and laser ablation have produced Broken Gap Junction (BGJ) devices [10]. The first two approaches deposit epitaxial crystalline films with heavily doped p- and n-type layers which improve intraband tunneling by minimizing the depletion region [11], while laser ablation processing is not widely adopted in industry. A BGJ[1] device is a solution to these issues. Specific resistivities ($R_c$), defined as the slope of the *J–V* curve at zero voltage, as low as $10^{-7}$ Ω-cm$^2$ have been reported [12], a value far lower than that needed for PV applications. An $R_c$ of 1 Ω-cm$^2$ used in a tandem device with a short circuit current of 20 mA/cm$^2$, would lead to a voltage drop less than 20 mV at the optimum operating point.

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[1] Broken gap junction.

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While tunnel junctions in epitaxial III–V compound semiconductor based solar cells have been successful, making a tunnel junction in thin film PV devices based on polycrystalline CdTe or CIGS layers is extremely difficult. Fast grain boundary diffusion [13,14] during the high temperature steps needed to fabricate the top absorber layer will rapidly deplete dopants from the vicinity of the junction, dramatically increasing $R_c$. Another problem is identifying the materials that can be used for tunnel junction. For example, most metal oxides are transparent in the visible and have been used to form heterojunctions with CIGS type absorbers but deep levels in oxide semiconductors dramatically increase resistance as trapped charge alters the internal electric fields. Another difficulty with transparent conducting oxides is the difficulty of finding p-type semiconductors with doping high enough to be effective while still aligning to the bands of the upper and lower cells.

Thermal budget is an extremely important consideration in developing a monolithically integrated tandem device. The bottom cell and the tunnel junction must both survive the thermal cycle required to form the top cell. In addition to the tunnel junction diffusion problem, if the bottom cell is made with a conventional CIGS stack Cd will diffuse from the CdS buffer layer into the CIGS, counter doping the absorber. If the diffused dose is high enough, it can destroy the pn junction. This occurs at 250 to 300 °C, depending on the time at temperature [15]. A ZnS buffer appears to be only slightly better. Since there is no known way to make a high performance top cell at such low temperatures, the bottom device will have to be modified in some manner. To begin to address the thermal budget concern, we have annealed the broken gap junction at temperatures between 300 °C (a temperature at which the lower CIGS device might survive) and 600 °C (a temperature at which high quality CIGS can be made) to simulate possible top cell processing. The intent is to see if the broken gap device would be degraded as a conventional tunnel junction would. Modification of the lower device to survive the top cell processing remains an open problem.

## 2. Operation and material selection

For a BGJ device, the fundamental requirement is that the electron affinity of the n-type layer must be equal to or greater than the sum of the electron affinity and the band gap of the p-type layer (Fig. 1b) [16]. The chemical doping levels of the two layers are relatively unimportant. Electrons in the valence band of the p-type side are at a higher energy than unoccupied states in the conduction band of the n-type side. At equilibrium these electrons will flow from the p-type side to the n-type side autodoping the junction. This charge transfer creates accumulation layers at the interface leading to band bending (shown in Fig. 1c) and introducing a narrow energy barrier. Under forward bias, electrons injected into the n-type material recombine with holes injected into the p-type material, giving rise to current through the junction. More importantly, under reverse bias, the electrons in the valence band of the p-type layer are injected into the n-type layer conduction band, while the holes in the valence band of the n-type layer are injected into the valence band of the p-type layer. Consequently, the junction is Ohmic. Numerical simulations of a metal-oxide broken-gap heterojunction by Song et al. between p-$Cu_2O$ and n-$In_2O_3$ suggest that low-resistances can be achieved even with moderate dopant concentrations.

In addition to the requirements on band alignment, the films must be optically transparent, relatively stable, and good conductors [17,18]. The need to minimize optical absorption over the solar spectrum would seem to favor wide band gap materials. However, in tandem solar cells most of the light with energies greater that the band gap energy of the upper cell will be absorbed before reaching the BGJ. Thus, the band gap must only be greater than about 1.7 eV. The films must be stable at ~500 °C, the temperature needed for depositing a high quality top cell absorber [19]. Stability in air would make processing substantially easier. Therefore this article will focus on heterojunctions between transparent conductive oxides (TCOs) [20] as potential BGJ materials. The resistivity of the TCO is less of a concern since, for a series specific resistivity of 1 Ω-cm$^2$ or less, materials with resistivities as high as $10^4$ Ω cm can be used in 40 nm thick films.

There is a paucity of p-type TCOs. $CuAlO_2$ and $Cu_2O$ are the only currently known p-type TCOs which have work functions around 5 eV [21], and band gaps well above 1.7 eV, the optimal band gap for the top cell of a tandem device. Band alignment to these materials reduces the options for the n-type layer to $GaInO_3$ and $ZnSnO_3$ [22]. In this work, zinc stannate was chosen because it has a wider band gap than $GaInO_3$, is thermally stable at high temperatures [20], and avoids the use of indium.

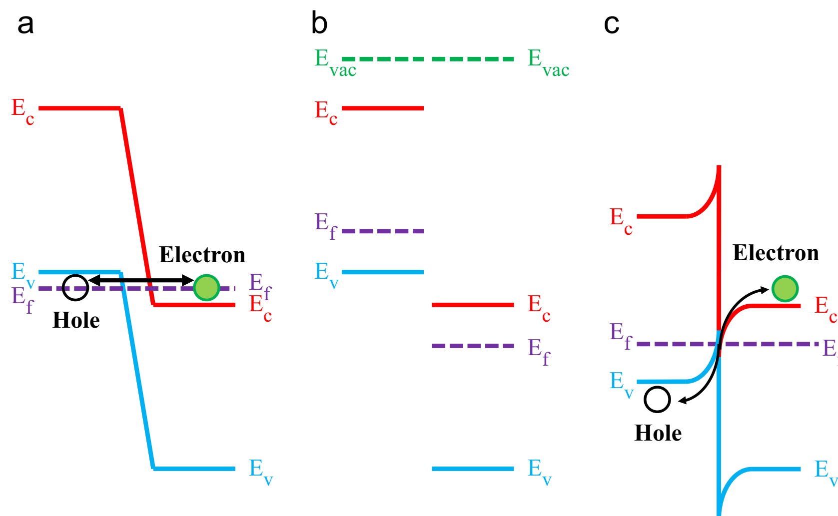


**Fig. 1.** Barrier tunneling vs. broken gap: (a) A conventional $p^+/n^+$ tunnel junction at equilibrium; and a broken gap junction before (b) and after (c) equilibrium. In both cases the valence band maximum of the p-type semiconductor is at a higher energy than the conduction band minimum of the n-type semiconductor at equilibrium. In the $p^+/n^+$ tunnel junction this is achieved by doping. In the broken gap junction, the valence band maximum of the p-type semiconductor is at a higher energy than the conduction band minimum of the n-type because of the difference in the electron affinity of the two materials.

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