

Intermixing, band alignment and charge transport in AgIn₅S₈/CuI heterojunctions

I. Konovalov*, L. Makhova, R. Hesse, R. Szargan

Wilhelm-Ostwald-Institut für Physikalische und Theoretische Chemie, Universität Leipzig, Linnéstraße 2, 04103 Leipzig, Germany

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Abstract

Possibilities of creating photovoltaic devices using CuI/AgIn₅S₈ heterojunctions are considered. Among other properties, preferential formation of polar (111) surfaces makes n-type AgIn₅S₈ an attractive candidate for absorber layers of top cells in 4-terminal tandem structures. Cu–Ag exchange at the interface with p-type CuI was observed. This intermixing results in an additional component of Ag 3d₅ photoelectron line after deposition of CuI, in the Cu (but not I) contamination of the surface after a chemical removal of CuI, and in a photoelectric sensitivity of the junction at energies below the band gaps. Valence band offsets of 0.4 and 0.5 eV (cliff) were found at interfaces with thin film and bulk AgIn₅S₈, supporting a conduction mechanism through interface recombination. Pinning conflict at the interface between materials with contradictory doping limitations is likely to promote the intermixing.

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1. Introduction

One of the possibilities to improve the efficiency of solar cells is the utilization of tandem structures. In such structures, the solar energy conversion happens in several junctions in parallel, depending on the wavelength of the light. The bottom cell for a two-junction tandem design may be an efficient cell with 1.0–1.2 eV band gap. An optimal top cell has to have a band gap of about 1.5–1.8 eV [1]. Two types of new materials are currently explored for the wide gap cells: sulfide and selenide chalcopyrites and polycrystalline nitride compounds. In wide gap polycrystalline chalcopyrites, problems related to strong interface recombination emerge as soon as the band gap exceeds 1.1–1.3 eV [2,3]. The use of nitrides is related to the success of single crystalline GaN in optoelectronics. However, the grain boundaries in III–V polycrystalline

films are recombinationally active. Contrary, I–III–VI compounds show a small grain boundary recombination velocity, which can be explained by a polar nature of the boundaries [4]. Here, we discuss utilization of a wide band gap semiconductor material AgIn₅S₈ with spinel structure and a band gap [5] of 1.7–1.8 eV. Spinel semiconductors have been successfully used in solar cells before [6–8]. Since these materials are mostly n-type, a p-type buffer layer is necessary instead of the usual for CuInS₂ cells n-CdS buffer. The change of the cell polarity can be tolerated in the tandem cell design e. g. by using separate contacting of the cells (4-terminal tandem device, [9]). Successful application of the p-type transparent semiconductor CuI in solar cells has recently been demonstrated [10]. The band gap of CuI (3.0 eV) is wider than that of CdS, the fact being particularly important for cells active in the blue spectral region. Here, we study the band alignment and the interdiffusion at the AgIn₅S₈/CuI interface, using X-ray photoelectron spectroscopy (XPS). We observe that the electrical properties of the junction match the results of the spectroscopic investigation.

* Corresponding author. Tel.: +49 341 97 36 490; fax: +49 341 97 36 399.
E-mail address: ikono@chemie.uni-leipzig.de (I. Konovalov).

2. Experimental details

2.1. Analytical equipment

For X-ray diffraction analysis (XRD) we used a Seifert XRD 3000 Diffractometer system equipped with Cu K_{α} ($\lambda=1.5405$ Å) X-ray radiation source and a vertical goniometer in the Bragg–Brentano geometry. For X-ray photoelectron spectroscopy measurements, a VG ESCA-LAB 220iXL equipment with a monochromatic Al K_{α} X-ray source was used. No special attention has been usually paid to the absolute calibration of the energy axis of the electron analyzer unless otherwise noted, since only energy differences are of interest here. The deposition of CuI for band discontinuity studies was done in situ in a preparation chamber adjacent to the spectrometer. The measurements of the composition were performed using Bruker S4 Explorer X-ray fluorescence (XRF) analytic system. The self-absorption and secondary excitation were treated according to the matrix coefficient approach. These calculations and intensity analysis have been done using the Bruker software Spectra Plus (Eval). The software is able to consider a thin homogeneous sample and to determine its thickness by evaluating the fluorescence intensity. Current–voltage (I – V) characteristics were measured using an Autolab PGSTAT 10 computer-driven potentiostat. Tungsten halogen lamp was used as a light source for I – V tests under illumination. The differential spectral response has been measured at the Institute for Solar Technologies (Frankfurt (Oder)) using an interference filter monochromator with a bandwidth of 10 nm. The light sources were Xe and tungsten–halogen lamps. The monochromatic light was mechanically chopped and a lock-in measurement of the short circuit current was performed. The temperature controlled sample was subject to a bias light illumination of approximately 1 sun intensity.

2.2. Growth of crystals and films

AgIn₅S₈ single crystalline blocks can be grown using the vertical Bridgman method from elements as described by Orlova et al. [5]. We used binary sulfides as raw material, instead of the elements, in order to decrease the pressure during the growth. 5N binary sulfides Ag₂S and In₂S₃ were put in a quartz ampoule in stoichiometric quantities. The ampoule has been evacuated, tempered at about 500 °C for 2 min and sealed. The growth began in the upper, hotter zone, of the two-zone oven, where the temperature of 1100 °C was maintained. The melt was homogenized for 30 min using mechanical vibration. Subsequently, the ampoule has been moved downwards at a rate of 300 mm/h into the colder annealing zone, where a temperature of 840 °C has been maintained.

Due to a relatively rapid cooling rate and no artificial seeding, the growth process results in a polycrystalline ingot with single crystalline blocks of several millimeters in size.

The crystalline blocks were readily visible, because the surface of the crystal was found to be covered with microscopic crystalline facets. Using optical reflection from the facets, we oriented one of the facet planes in-plane with the sample surface of the powder diffractometer. Fig. 1 shows a diffraction pattern of the sample, with (222) and similar even diffraction lines present. The (222) and (444) reflexes are not among the strongest in a diffractogram of a powdered sample [11], but they are dominant in our case. Therefore, the facets of AgIn₅S₈ crystals are (111) surfaces. Spurious (511) reflexes may originate from a neighbor grain with a different orientation.

Thin films of AgIn₅S₈ were grown from a metallic precursor. Silver and indium films were vacuum-evaporated onto 200 nm Ti bottom contact on glass and sulfurized in sulfur vapor at 400 °C for 10 min. For evaporation, a high vacuum chamber, tungsten boats and a quartz microbalance were used. Details of the precursor formation may be found elsewhere [12,13]. We observed better photoelectric properties of the junction with an In-poor absorber of an overall In/Ag atomic ratio of about 4.05 as determined from XRF measurements, although deviations of the ratio as large as ± 0.1 absolute were found tolerable. Continuous absorber films could be obtained after two subsequent depositions, of 600 nm each. The deposition of the metals was controlled using a quartz microbalance to achieve stoichiometry. The eventual deviations of the atomic ratio were then determined by XRF and corrected through evaporation of a due amount of Ag and a third sulfurization, since we were not able to control the evaporation process precisely enough using the microbalance only. Several absorber layers having occasionally the due atomic ratio without any composition correction generally showed the same properties as the corrected layers of the same composition.

2.3. Junction design

Since AgIn₅S₈ is a typical n-type semiconductor [5,14], we chose a typical p-type transparent in visible semi-

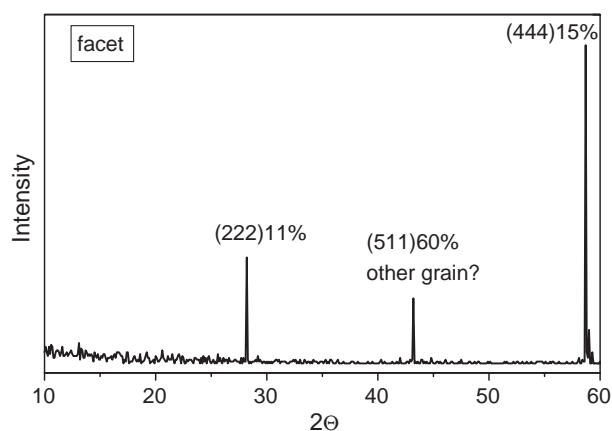


Fig. 1. X-ray diffraction pattern of an AgIn₅S₈ crystal oriented in-plane with a facet.

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