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Fabrication and characterization of wide band-gap $CuGaSe_2$ thin films for tandem structure

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ABSTRACT

The effects of $CuGaSe_2$ (CGS) absorber layers with various film thicknesses have been investigated by using scanning electron microscopy (SEM) and photoluminescence (PL). SEM measurements show that the thickness of the CGS absorber layers grown on Mo-coated soda-lime glass substrate increases with an increase in Ga flux. Moreover, a trend in the grain size and surface roughness can be observed. Single broad peaks centered around 1.64 eV are observed in the PL spectra of the CGS samples. From the excitation power-dependent PL measurement, the single broad peaks are associated with the donor–acceptor pair transition from CGS absorber layers. In addition, we demonstrate that different film thicknesses of CGS absorber layer result in different defect concentration by temperature-dependent PL measurement.

1. Introduction

$Cu(In_{1-x}Ga_x)Se_2$ (CIGS) is the best candidate material for the fabrication of highly efficient thin film solar cells due to its high optical absorption coefficient and its direct band-gap in the absorber layer. In previous reports, CIGS solar cells have shown a maximum lab efficiency of 19.9% with a band-gap of ~1.1 eV [1]. However, it is generally agreed that solar cells need at least an efficiency of 25% to be economical. One alternative to achieve high efficiency is through the realization of a tandem structure by stacking one cell on top of another, which has a different energy band-gap [2].

The ideal band-gap energy for top-cells of a tandem structure is around 1.6–1.7 eV, which makes $CuGaSe_2$ (CGS), with a band-gap of 1.68 eV, an attractive material. However, a serious degradation of efficiency has been observed in the wide band-gap C(I)GS solar cells. High Ga concentration has been shown to increase the number of defects in a cell leading to degradation in cell performance [3]. Even though the CGS solar cells have been developed during the last few years [4–6], more research is needed to better understand how the growth parameters and mechanisms on the growth can be modified to improve CGS thin film performance.

In this study, CGS absorber layers with various film thicknesses were fabricated by a three-stage process using the co-evaporation system. Moreover, we report some interesting results associated with the defects in the photoluminescence (PL) spectra taken from the CGS thin films grown with different growth conditions. The PL spectroscopy is a useful technique to investigate band-gap and defects in semiconductors, and has the benefit of being a nondestructive characterization technique. Especially, we explain that different film thicknesses of CGS absorber layer result in different defect concentration during the formation of CGS thin film by optical spectroscopy method.

2. Experimental procedures

A Mo back contact layer (~1 μm) was grown on soda–lime glass substrate from a 99.95% pure Mo target by using a DC magnetron sputtering system. The CGS absorber layers with various film thicknesses were grown by a three-stage process using the co-evaporation of elemental Cu, Ga, and Se from effusion cells. The temperature of the substrate during the growth was measured by using a thermocouple in contact with the backside of the substrate [7]. In the first stage, Ga and Se were co-evaporated on the Mo/glass substrate at 400 °C to form a Ga_2Se_3 precursor layer. In the second stage, Cu and Se were co-evaporated at 550 °C to grow a Cu-rich $CuGaSe_2$ thin film. The end of the second stage was determined by measuring the substrate temperature drop. In the third-stage, Ga and Se were co-evaporated to complete the growth of the Ga-rich $CuGaSe_2$ thin films. Finally, the sample was cooled in a Se atmosphere to prevent the re-evaporation of Se from the $CuGaSe_2$ layer [8]. In this study, to investigate the effects of various film thicknesses of the CGS absorber layer, Ga flux was controlled for each sample. On the other hand, Cu/Ga ratio was fixed about 0.9 for all the samples.

In photoluminescence (PL) measurements, an argon ion laser with a wavelength of 514.5 nm was used as an excitation source to generate electron–hole pairs. The luminescence light from the

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samples was focused with collection lenses, dispersed using a monochromator and detected by a photomultiplier tube (PMT) detector. For the excitation power-dependent and the temperature-dependent PL, the samples were mounted on the cold finger of a closed-cycle helium cryostat. The PL spectra were taken in the nominal output power range of 10–50 mW with a temperature range of 14–300 K.

3. Results and discussion

Fig. 1 shows the cross-section and plan-view scanning electron microscopy (SEM) images of the CGS absorber layers, including various film thicknesses: 0.68 μm (sample A), 1.36 μm (sample B), and 2.46 μm (sample C). These samples were grown under the same conditions except for different amounts of Ga flux. It is found that the thickness of a CGS film increases with an increase in Ga flux. In addition, a trend in the grain size and surface roughness can be observed. The grain size increases with the CGS film thickness until around 1.36 μm, but pores have appeared along the grain boundaries as shown in Fig. 1b. Moreover, the CGS film of 2.46 μm (Fig. 1c) is rougher than that of sample the 1.36 μm film (Fig. 1b). In the case of CGS films, low Ga flux grown films show continuous grain boundaries and a highly dense surface, whereas high Ga flux grown films show large grain boundaries, many pores and a

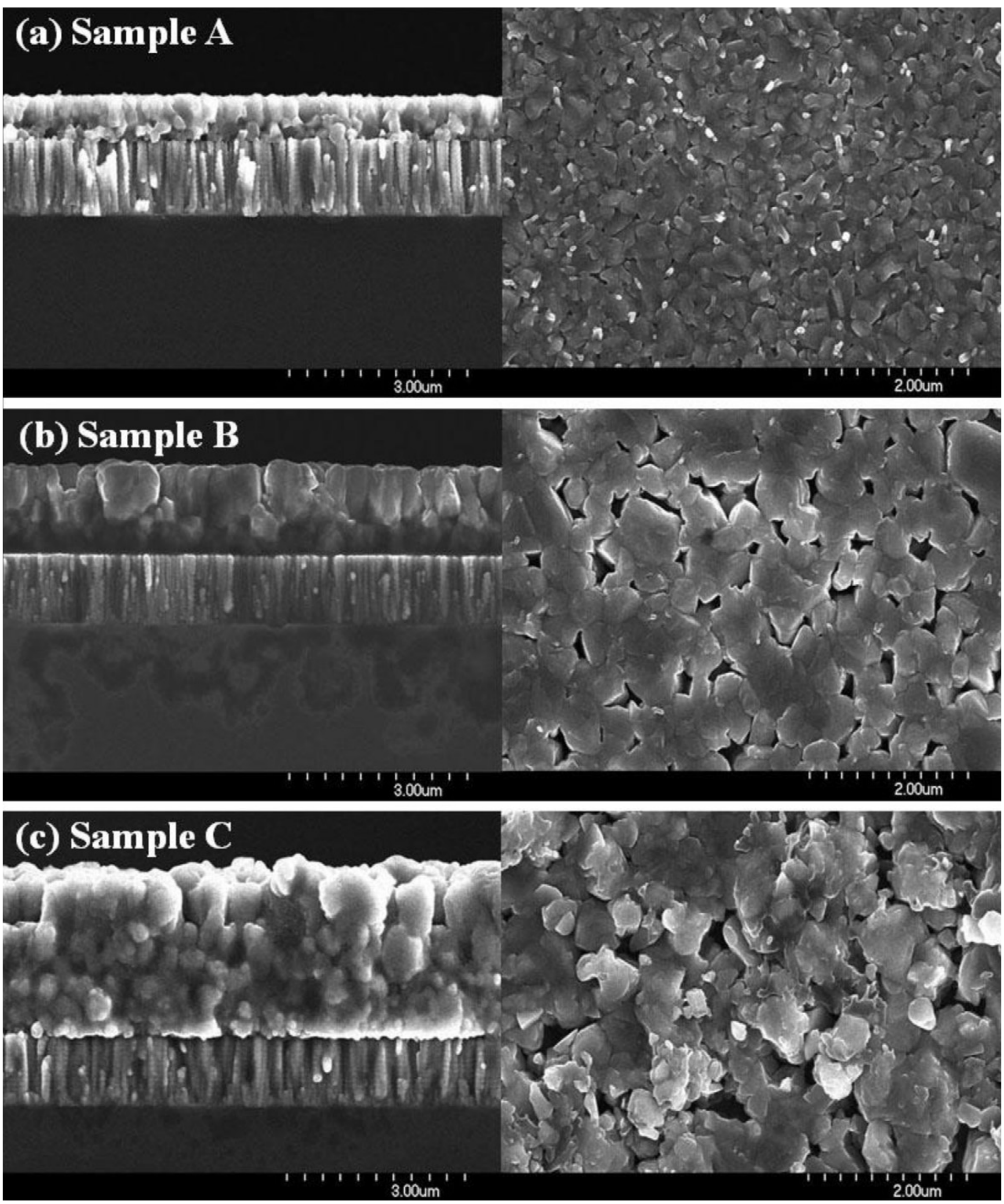

Fig. 1. Cross-section and plan-view SEM images of CGS absorber layer with various film thicknesses. Sample A: 0.68 μm (a), sample B: 1.36 μm (b), and sample C: 2.46 μm (c).