

Effects of annealing under various atmospheres on electrical properties of Cu(In,Ga)Se₂ films and CdS/Cu(In,Ga)Se₂ heterostructures

T. Sakurai^{a,*}, N. Ishida^a, S. Ishizuka^b, M.M. Islam^a, A. Kasai^a, K. Matsubara^b,
K. Sakurai^b, A. Yamada^b, K. Akimoto^a, S. Niki^b

^a Institute of Applied Physics, University of Tsukuba, 1-1-1 Tennodai, Tsukuba, Ibaraki 305-8573, Japan

^b National Institute of Advanced Industrial Science and Technology (AIST), 1-1-1 Umezono, Tsukuba, Ibaraki 305-8568, Japan

Available online 23 December 2007

Abstract

The effects of annealing under various atmospheres on the electrical properties of Cu(In,Ga)Se₂ (CIGS) films and CdS/CIGS heterostructures were investigated. For CIGS films without CdS, the electrical properties of CIGS degraded under vacuum and O₂ annealing, although such degradations were not observed under N₂ annealing. For the CdS/CIGS heterostructures, the electrical properties of the junctions improved after annealing under all gas ambients. Therefore, CdS films prevent the chemical reactions at the CIGS surfaces and are necessary for effectively annealing the CIGS film. We observed a distinct correlation between the degradation of the electrical properties and increase in the defect density. Finally, we discussed the origin of the defect states.

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Pacs classification: 71.55.Ht; 73.61.Le; 71.20.Nr

Keywords: Cu(In,Ga)Se₂; Defect state density; Admittance spectroscopy; CdS/CIGS interface; Annealing

1. Introduction

Cu(In_{1-x}Ga_x)Se₂-based (CIGS-based) thin-film solar cells have undergone remarkable developments during the past decades, and an efficiency of more than 19% has been achieved in laboratory-scale cells [1]. In spite of such an excellent device performance, however, the electrical properties of CIGS cells have not been fully understood yet due to their complex device structures and multinary compositions [2,3]. In particular, the role of the chemical bath deposition of CdS (CBD-CdS) films and several types of interfaces (e.g., CdS/CIGS interfaces) in influencing the device performance is still unclear, although these characteristics are believed to play an important role in the carrier recombination processes [4]. To clarify the correlation between the electrical properties and device performance of the cells, a study investigating the defects in CIGS is useful. In our earlier study, we obtained the depth information of the defect levels in CIGS, and we found that one of the deep defects that

exist near the surface of CIGS plays an important role in the device performance [5,6]. In this study, we report the effects of annealing under various atmospheres on the electrical properties of CIGS films and CdS/CIGS heterostructures by means of current–voltage (*I*–*V*), capacitance–voltage (*C*–*V*), and admittance spectroscopy measurements. We observed the variation in the electrical properties of these junctions because of annealing under various ambients. Further, a distinct correlation between the electrical properties and defect density was observed for all the samples. Finally, we discussed the origin of the defect states and the role of the CBD-CdS layer formed on the CIGS film.

2. Experimental

Cu(In_{1-x}Ga_x)Se₂ films (*x* ~ 0.5) were grown on Mo-coated soda-lime glass substrates by means of a three-stage process using a molecular beam epitaxy (MBE) system. The copper content of the CIGS films was roughly fixed at around Cu/(In+Ga)=0.8, which was determined by using an electron probe microanalyzer (EPMA). The detailed deposition conditions for the CIGS films have been described elsewhere [7]. After the deposition of CIGS,

* Corresponding author. Tel.: +81 29 853 6150; fax: +81 29 853 5205.

E-mail address: sakurai@bk.tsukuba.ac.jp (T. Sakurai).

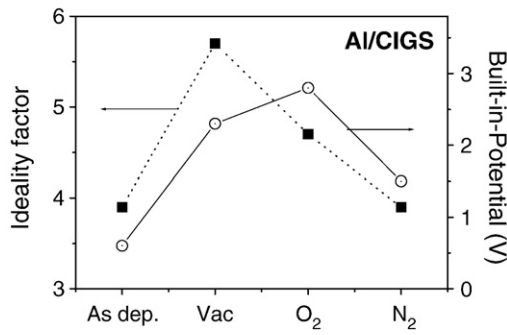


Fig. 1. Dependence of annealing ambient (as-deposited (As dep.), vacuum (Vac.), O₂, and N₂) on ideality factor and built-in-potential for the Al/CIGS Schottky junctions. Annealing was performed at 400 K for 30 min.

the films were immersed in a 0.1 M KCN aqueous solution for 5 min to remove the Cu₂Se segregations [8], which are usually formed on the surface when the composition of the deposited CIGS films is close to the stoichiometric ratio [9]. CdS/CIGS heterostructures were fabricated on the CIGS films by means of the chemical bath deposition of CdS (CBD-CdS). The annealing of the CIGS films and CdS/CIGS heterostructures was done at 400 K in vacuum (10⁻⁵ Torr) and nitrogen (N₂, 10 Torr) and oxygen (O₂, 10 Torr) ambients for 30 min by using the rapid thermal annealing (RTA) method. Al electrodes were grown on the annealed samples to fabricate the junctions used in the electrical measurements.

Admittance spectroscopy measurements were carried out in the temperature range from 10 K to 300 K with the ac frequencies between 1 KHz and 1 MHz using the Agilent 4284A LCR meter. In this analysis, we determine the density of states and activation energy of the traps by varying the ac frequency and substrate temperature [10]. Further, we analyzed the electrical properties of these junctions by means of the capacitance–voltage (*C–V*) and current–voltage (*I–V*) measurements.

3. Results and discussion

3.1. CIGS film without CdS

Fig. 1 shows the dependence of annealing ambient on ideality factor and built-in-potential of Al/CIGS Schottky junctions. Annealing procedures were done at 400 K under various ambient for 30 min. The ideality factor of these junctions is determined by *I–V* relation under a forward bias using the thermionic emission theory, which is given as

$$I = I_s \exp \left[\left(\frac{q(V - IR_s)}{nkT} \right) - 1 \right],$$

where I_s is the saturation current, R_s is the series resistance, k is the boltzman constant, T is the absolute temperature, and n is the ideality factor [11]. The built-in-potential (V_{bi}) is determined from the intercept of the extrapolated $1/C^2–V$ straight line with the V -axis [11]. It was observed that the ideality factor of Al/CIGS was increased compared with as-deposited sample in case of annealing under vacuum and oxygen (O₂) ambient, while that was

not increased by nitrogen (N₂) annealing. It is apparent that the ideality factor strongly affects the device performance since trap-assisted tunneling currents increase with increasing the ideality factor. Therefore, these results indicate that the crystalline (bulk) defects, which cause tunneling currents, are easy to form in the CIGS films by 400 K annealing under vacuum and O₂ ambient. With regard to V_{bi} , it was found that the variation of V_{bi} is similar to that of the ideality factor, namely, V_{bi} tends to increase with increasing the ideality factor. For the vacuum and O₂ annealed samples, however, the shift of V_{bi} showed an opposite correlation with that of the ideality factor since the surface properties of CIGS, which directly correlate with V_{bi} , are relatively changed by the annealing ambient due to the high reactivity of the film surfaces. Thus, the bulk and surface defects, which cause a degradation of the electrical properties, are introduced in the CIGS films by the specific annealing treatment.

To clarify the role of the annealing ambient in the variation in the electrical properties of CIGS, we analyzed the defect density spectra of Al/CIGS obtained from the admittance spectroscopy by using a method introduced by Walter et al. [10], as shown in Fig. 2(a). We found two trap levels denoted as α and ζ , of which activation energies were almost 10 and 250 meV, respectively, for all the junctions. It was observed that the density of these trap levels varied with the annealing ambient. Further, for the junctions with annealing under vacuum and O₂, we found a new peak α' at the activation energy of approximately 90 meV. The densities of these trap levels, which are obtained by integrating over energy using a Gaussian approximation (the shaded portion in Fig. 2(a)), are shown in Fig. 2(b). With regard to the

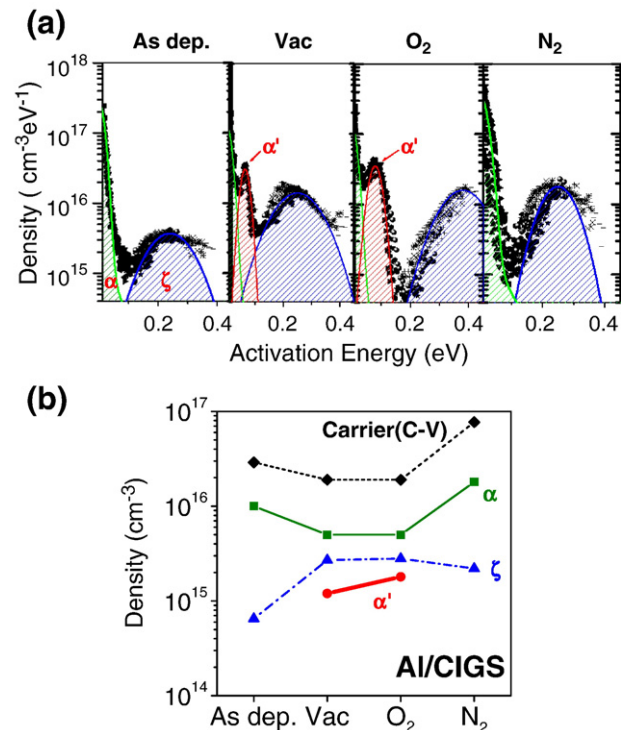


Fig. 2. (a) Defect density spectra for Al/CIGS with annealing at 400 K under various ambients (As dep., Vac., O₂, and N₂) for 30 min. (b) Dependence of annealing ambient on trap level density (α , α' , β , and ζ) for Al/CIGS.

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