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Properties of deep-level defect in Cu(In, Ga)Se_2 thin films

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Abstract
The properties of the deep-level defect which locates at around 0.8 eV above the valence band in Cu(In, Ga)Se_2 thin films with different Ga content was investigated by transient photo-capacitance measurements and the time-resolved photoluminescence. It was deduced that the 0.8 eV deep defect plays as a carrier recombination center and influences the efficiency of CIGS solar cells. The influence of the deep defect becomes more significant as the Ga content increases.

1. Introduction

Cu(In_{1-x},Ga_x)Se_2 (CIGS) is an excellent material for the application to thin-film solar cells. To enhance the performance of CIGS solar cells, however, it is necessary to understand the carrier recombination mechanism, which is caused by the deep defects in photoabsorber layers. In our previous study we have pointed out that the defect level centered around 0.8 eV from the valence band may act as a recombination center at room temparature [1]. In this study, we try to clarify whether the 0.8eV-defect works as a recombination center by the time-resolved photoluminescence (TRPL) and transient photo-capacitance (TPC) measurements.

2. Experimental

Polycrystalline CIGS thin-films were grown on Mo-coated soda-lime glass substrates by a three-stage process using a molecular beam epitaxy (MBE) system. After CdS deposition on CIGS, TRPL measurements were performed. For the measurements of TPC, cell structure was fabricated by deposition of i-ZnO and Al doped ZnO. The device parameters of CIGS thin-film solar cells are listed in Table I. CIGS films with Ga content (x) varied between 0.30 and 0.80 were prepared to investigate the relationship between the defects and the PL properties. The PL measurements were carried out at room temperature with a confocal laser scanning microscope using a 635-nm diode laser (Scientex OPG-3300) for the excitation. A pulsed light source with a pulse width and repetition rate of 100 ps and 2-5 MHz was used for the excitation in the TRPL measurements. The TPC measurements have been carried out in the temperature range of 60 to 340 K with a fill pulse bias of 0 V and a reverse bias of 0.5 V. The pulse width and duration are fixed at 50 ms and 1 s, respectively.

Table I. CIGS device parameters used in this study measured under AM1.5 illumination at 25 °C

Ga/(Ga+In) (x)	J_{sc} (mA/cm^2)	V_{oc} (V)	FF	Efficiency (%)
0.30	29.74	0.687	0.78	15.89
0.40	27.88	0.730	0.77	15.67
0.52	26.48	0.700	0.73	13.57
0.62	24.39	0.718	0.72	12.67
0.76	22.35	0.738	0.71	11.15
0.80	20.30	0.731	0.68	10.14

3. Results and discussion

Fig. 1 (a) shows the PL decay curves for CIGS films with Ga content varied from 0.30 to 0.80. For each film, the signal was detected at the maximum of the PL emission. A bi-exponential relationship between the PL intensity and the time was used to fit the PL decay and the minority carrier lifetime was evaluated using the slow emission component which approximately corresponds to the values observed under low intensity irradiation [2]. Figure 2 (b) shows the relationship between the minority carrier lifetime and the Ga content, the

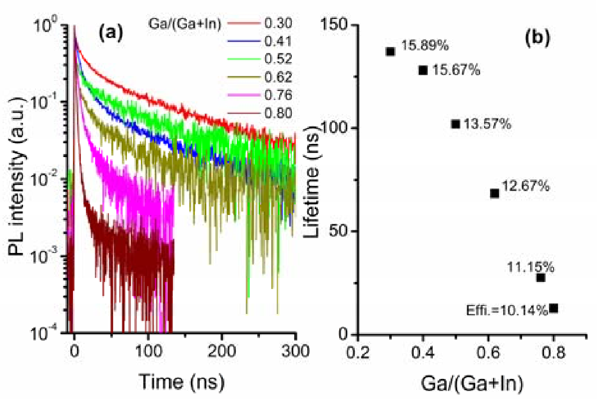

Fig. 1 (a) PL decay curves for CIGS films with Ga content varied from 0.30 to 0.80. (b) Dependence of PL decay lifetime on the Ga content with the conversion efficiency.

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efficiency of corresponding solar cell device was also presented. It can be obviously seen that high device efficiency corresponds to long minority carrier lifetime.

Usually, the PL decay properties can be interpreted by radiative decay which is caused by interband recombination and nonradiative decay which is assumed to be described by Shockley-Read-Hall (SRH) statistics [3]. The measured PL decay lifetime is given by the reciprocal sum of the lifetimes caused by the two decay processes as equation (1),

$$\frac{1}{\tau_{TRPL}} = \frac{1}{\tau_{rad}} + \frac{1}{\tau_{SRH}} \qquad (1)$$

Where τ_{TRPL} is the measured PL decay lifetime, τ_{rad} is the lifetime of radiative recombination under low intensity irradiation or in the low carrier injection regime and is described by equation (2),

$$\tau_{rad} = \frac{1}{BN_A} \qquad (2)$$

Where B is the radiative recombination coefficient and N_A is the net majority carrier density. τ_{SRH} is the lifetime of non-radiative recombination via deep defects and in low carrier injection regime can be described by equation (3),

$$\tau_{SRH} = \frac{1}{N_T v_{th} \sigma} \qquad (3)$$

where N_T is the defect density, v_{th} is the thermal recombination velocity and σ is the capture cross section of minority carrier by the defects.

Assuming a radiative recombination coefficient of B=8×10^{-11} cm^{-3}s^{-1} and a thermal velocity v_{th}=10^{7} cm/s at room temperature [4] and taking a doping density, N_A, of the order of 10^{16} cm^{-3} for all samples, as identified by capacitance-voltage measurement, the radiative lifetime for CIGS can be expected to be longer than 1 μs, which is much larger than the obtained lifetimes as shown in Fig. 1. Thus the minority carrier lifetime was considered to be dominated by non-radiative recombination via deep defects, while considering the similar thermal recombination velocity and capture cross section for the same deep defect, the relationship between the deep defect density after normalized and Ga content could be obtained.

A deep defect locates at 0.8 eV above valence band maximum (VBM) for all Ga content of CIGS has been found [5] and it was proved to be a recombination center at room temperature by a two-wavelength photo-capacitance method [6]. In our previous work, a new characterization method for relative 0.8eV-defects density has been proposed from the transient photo-capacitance (TPC) data [7], and using the characterization method, the defect density was found to increase with increasing Ga/III ratio. To clarify whether the 0.8eV-defect works as a recombination center or not, the normalized defect density depending on the Ga content and the normalized defect density extracted from the TRPL were compared as as shown in Fig .2.

We can see a very similar tendency between the relative defect density with Ga content estimated from the TPC and TRPL measurements. So it may be deduced that the 0.8 eV deep defect plays as a carrier recombination center. The cause of the degradation of CIGS-cell performance in higher Ga content may be due to the 0.8-eV defect, and it is necessary to decrease the defect density to improve the cell efficiency.

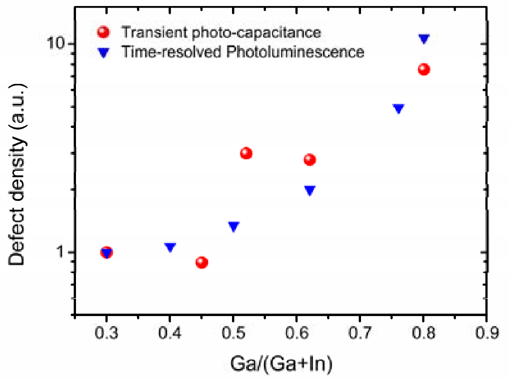

Fig. 2 Comparison of the normalized relationship between deep defect density and Ga content extrapolated from TPC measurement (circle) [7] and TRPL (triangle) measurement, respectively.

4. Conclusions

The properties of the deep-level defect which locates at around 0.8 eV above the valence band in Cu(In, Ga)Se_2 thin films with different Ga content was investigated by TPC and TRPL measurements. It was deduced that the 0.8 eV deep defect plays as a carrier recombination center and influences the efficiency of CIGS solar cells. The influence of the deep defect becomes more significant as the Ga content increases.

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