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Photothermal spectroscopy of CdInGaS_4 and related compound

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Abstract: Photothermal spectroscopy using high sensitive thermistor is applied to study the heat generation of multinary compounds $\text{Cd}_x\text{InGaS}_{3+x}$ system ($x=1,2,3$) together with the optical absorption. There are differences between the photothermal spectra and optical energy absorption (1-T-R) in $\text{Cd}_x\text{InGaS}_{3+x}$ system and it depends on the composition x .

1. INTRODUCTION

The quaternary layered compound CdInGaS_4 has attracted attention as one of the light-emitting materials for several optoelectronic devices because it shows an intense red photoluminescence even at room temperature[1]. This material has an indirect energy gap of about 2.57 eV at room temperature. Recently, we have found a new material $\text{Cd}_3\text{InGaS}_6$ which shows an intense green photoluminescence at liquid nitrogen temperature[2]. The crystal structure analysis indicated that $\text{Cd}_3\text{InGaS}_6$ was not a single compound but a laminated material composed of CdS and CdInGaS_4 [3]. It is found that the intense green emission in $\text{Cd}_3\text{InGaS}_6$ is closely related with this laminated structure[4]. A number of considerable papers have been published on the investigations of nonradiative processes following by optical absorption in semiconductors.

In this work, we present the photothermal spectra (PTS) of CdInGaS_4 and related compounds in order to obtain informations on the nonradiative processes.

2. EXPERIMENTAL

The crystals of $\text{Cd}_x\text{InGaS}_{3+x}$ system ($x=1,2,3$) were grown from a melt of

Cd, In, Ga and S elements of 99.9999 % purity with stoichiometric composition by normal freezing method in an evacuated quartz ampule. The obtained $\text{Cd}_x\text{InGaS}_{3+x}$ system crystals were cleaved out of the ingot to about 0.5 mm thickness and were cut to the shape of square approximately 7 mm $\times$ 7 mm. The shaped $\text{Cd}_x\text{InGaS}_{3+x}$ system crystals were lapped with polish of Ce_2O_3 powder at surface and were etched with the etching reagent of 30% HCl + 70% H_2O for 1 hour.

For PTS measurements, We employed thermistor as a sensor for temperature increasing (i.e., resistance changes). PTS involves placing a thermistor in close to the sample and measuring temperature changes during sample irradiation with monochromatic light. In order to eliminate the effect of temperature changes of surrounding air, a differential thermistor arrangement was used to compensate for changes of ambient temperature. The thermistor has a nominal resistance of 12 K Ω , a sensitivity of 480 Ω/K at 20 $^\circ\text{C}$. The light source was 500 W short arc xenon lamp. Neither thermistor was irradiated directly by the exciting beam during the PTS measurements which carried out over the wavelength range from 400 nm to 900 nm. Excitating beam was irradiated on the sample for the period of 10 seconds. The PT signal intensity was always divided by the light intensity measured from the PTS of carbon. The optical absorption and reflection were measured by the conventional transmission and reflection methods.

3. RESULTS AND DISCUSSION

Fig.1 shows the normalized PT spectra of $\text{Cd}_x\text{InGaS}_{3+x}$ system as a function of photon energy in the form of semilog plot. It can be seen that these spectra show exponential curve for each case. PT spectrum of CdInGaS_4 shows sudden rise below fundamental absorption edge. On the other hand, PT spectra of $\text{Cd}_2\text{InGaS}_5$ and $\text{Cd}_3\text{InGaS}_6$ show gentle rising in comparison with that of CdInGaS_4 .

Fig.2 shows the total rate of optical energy absorption, $1-T-R$ (T and R are transmittance and reflectance, respectively), of $\text{Cd}_x\text{InGaS}_{3+x}$ system as a function of photon energy in the form of semilog plot. It can be seen that absorption spectra show exponential curve similar to the PT spectra. Fundamental absorption edge of $1-T-R$ are in good agreement with that of PT spectra .

In Figs. 1 and 2, the plots of the logarithms of PT spectra and $1-T-R$ versus photon energy approaches straight line below the fundamental absorption edge for each case. We plot the value of slope PT spectra and $1-T-R$ as a function of composition x of $\text{Cd}_x\text{InGaS}_{3+x}$ system derived from Figs. 1 and 2, as shown in Fig. 3. It can be seen that the PTS and $1-T-R$

Fig. 1. Photon energy dependence of PT spectra in $\text{Cd}_x\text{InGaS}_{3+x}$ system ($x=1, 2, 3$).

Fig. 2. Photon energy dependence of $1-T-R$ in $\text{Cd}_x\text{InGaS}_{3+x}$ system ($x=1, 2, 3$).

Fig. 3. Slope of the PTS and $1-T-R$ for $\text{Cd}_x\text{InGaS}_{3+x}$ system ($x=1, 2, 3$).

Fig. 4. Photon energy dependence of the $\text{PTS}/1-T-R$ in $\text{Cd}_x\text{InGaS}_{3+x}$ system ($x=1, 2, 3$).

show almost the same tendency. The slope of $\text{Cd}_2\text{InGaS}_5$ agrees with that of $\text{Cd}_3\text{InGaS}_6$ and smaller than that of CdInGaS_4 . As mentioned above, the crystal structures of CdInGaS_4 and $\text{Cd}_3\text{InGaS}_6$ (or $\text{Cd}_2\text{InGaS}_5$) are single compound which has a lattice constant of $a=3.85\text{\AA}$ $c=37\text{\AA}$, and laminated compound composed of CdS and CdInGaS_4 , respectively. From the result of Fig.3, we considered that the smaller slope value of PTS and 1-T-R in $\text{Cd}_2\text{InGaS}_5$ and $\text{Cd}_3\text{InGaS}_6$ might be related to laminated structure of CdS and CdInGaS_4 .

In order to examine a rate of optical energy absorption and heat generation we plot PTS/1-T-R of $\text{Cd}_x\text{InGaS}_{3+x}$ system as a function of photon energy, as show in Fig. 4. It can be seen that PTS/1-T-R versus photon energy are nearly constant above the fundamental absorption edge, and approaches the slope below it. From the result, we considered that the rate of nonradiative transition with optical absorption are not constant below the fundamental absorption edges for $\text{Cd}_x\text{InGaS}_{3+x}$ system ($x=1,2,3$).

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