

Carrier dynamics in $\text{Zn}_x\text{Cd}_{1-x}\text{O}$ films grown by molecular beam epitaxy

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Abstract. In this work, the carrier dynamics in $\text{Zn}_{1-x}\text{Cd}_x\text{O}$ thin films with different Cd contents grown by molecular beam epitaxy system have been investigated using photoluminescence and time-resolved photoluminescence measurements. The carrier lifetime can be estimated from the PL decay curve fitted by triple exponential function. The emission energy dependence and temperature dependence of the PL decay time indicate that carrier localization dominate the luminescence mechanism of the ZnCdO alloy semiconductor.

1. Introduction

Recently, ZnO -based semiconductor materials have been rapidly and widely developed for laser diodes (LDs) and light-emitting diodes (LEDs), owing to their tunable band gap by alloying with MgO to reach ultraviolet region or by alloying with CdO to achieve the visible region [1-4]. For the application in the region from ultraviolet to green light spectra, the ZnCdO alloy is useful to fabricate ZnCdO -related heterostructures or quantum well structures, which are the key elements in ZnO -based light emitters and detectors [5]. Therefore, it is quite necessary to investigate the optical characteristics in the ZnCdO alloys, such as the properties of band gap engineering, exciton behavior, and luminescence. The luminescence behavior provides not only a fundamental insight into recombination process but also information about the carrier localization.

In this work, we investigate the luminescence characteristics of ZnCdO thin films with different Cd contents grown by plasma assisted molecular beam epitaxy (PA-MBE) system by temperature-dependent photoluminescence (PL) and time-resolved photoluminescence (TRPL) spectra.

2. Experimental

The investigated $\text{Zn}_{1-x}\text{Cd}_x\text{O}$ films were grown on the *c*-plane Al_2O_3 substrates by SVT Associates molecular beam epitaxy system equipped with conventional effusion cells for evaporation of elemental Zn (6N) and Cd (6N). Oxygen (5N5) was supplied via an rf-plasma source after additional gas



purification. In order to reduce the lattice mismatch between $\text{Zn}_{1-x}\text{Cd}_x\text{O}$ and Al_2O_3 , the $\text{Zn}_{1-x}\text{Cd}_x\text{O}$ films were firstly grown at 350 °C following a 70 nm thick ZnO buffer layer grown at 650 °C. The Cd concentration of $\text{Zn}_{1-x}\text{Cd}_x\text{O}$ was controlled by adjusting the Cd/Zn beam pressure. The Cd concentrations applied for the four ZnCdO samples are approximately estimated to be 0.06, 0.08, 0.10, and 0.13, respectively by using the energy-dispersive X-ray spectroscopy (EDX) measurement. More details about the growth procedures have been described in the previous work [6].

The PL and TRPL were measured by using a diode laser (PicoQuant), which produces light pulses with 50 ps duration and a repetition rate of 1 MHz. PL measurement was setup using a spectrometer (Zolix omni- λ 500) with a grating of 1200 grooves/mm, and detected using a photomultiplier tube (PMT). TRPL was performed using the technique of time-correlated single-photon counting (TCSPC). The luminescence decay was detected with a high-speed PMT, followed by a computer plug-in TimeHarp counting card, which was triggered with a signal from the diode laser. Janis Research Model CCS-150 and LakeShore Model 321 temperature controller were used to carry out the temperature-dependent PL and TRPL spectra.

3. Results and discussion

Figures 1(a)~(d) show the normalized PL spectra recorded at 12 K of ZnCdO thin films with Cd content of 0.06, 0.08, 0.10, and 0.13, respectively. It is observed that the peak position of PL spectrum shifts to lower energy with increasing Cd content. To further clarify the luminescence mechanisms in ZnCdO thin films the carrier lifetime were measured by time-resolved PL technique. The inset in figure 1(a)~(d) display the luminescence decay profile monitored at the PL peak and the PL decay curve can be fitted by a triple exponential function as [7]

$$I(t) = A_1 * e^{\left(\frac{-t}{\tau_1}\right)} + A_2 * e^{\left(\frac{-t}{\tau_2}\right)} + A_3 * e^{\left(\frac{-t}{\tau_3}\right)} \quad (1)$$

As shown in the insets of figure 1, the red solid curve described from equation (1) can well fit the experimental data and to obtain the value of τ .

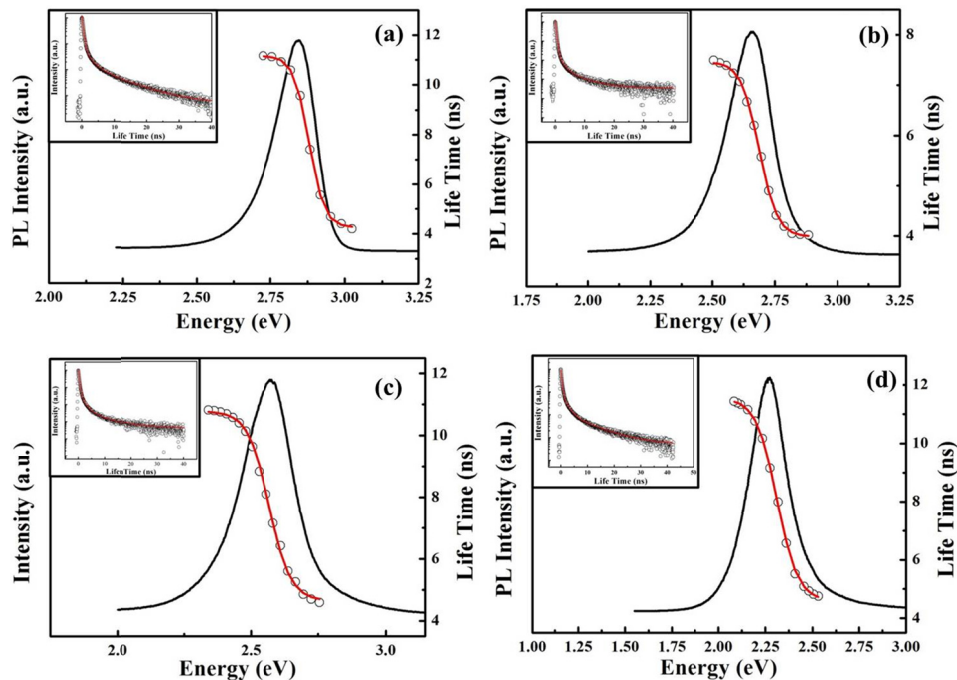


Figure 1. 12 K PL spectrum of ZnCdO thin films with Cd content of (a) 0.06, (b) 0.08, (c) 0.10, and (d) 0.13. The open circles display the emission-energy-dependent lifetime.

The estimated PL lifetime as a function of emission energy is further plotted as open circles in figure 1. The emission energy dependence of carrier lifetime shows that the luminescence decay is faster on the high energy side than the low energy side of PL spectrum. The decrease in the lifetime with increasing emission energy has been known as the characteristic of a localization effect and attributed to the localized carriers transfer process to lower energy states [8,9]. The decay rate of localized carriers is expressed as the radiative recombination rate plus the rate of transfer to lower energy sites, which results in the observed lifetime decreasing as the emission energy increases. The relationship between lifetime and emission energy can be expressed by [8,9]

$$\tau(E) = \frac{\tau}{1 + \exp\left[\left(E - E_{me}\right) / E_0\right]} \quad (2)$$

where τ is the radiative recombination lifetime, and E_0 is the characteristic energy for the depth of the localized states. E_{me} is a definite energy for the decay time that equals the transfer time, which is similar mobility edge. The red solid line in figure 1 indicates the best fit according to equation (2), and the obtained values of E_{me} , and E_0 are listed in table 1. The value of E_{me} decreases from 2.88 eV to 2.15 eV as increasing Cd content. In addition, the obtained E_0 for the $\text{Zn}_{0.94}\text{Cd}_{0.06}\text{O}$ sample is 25.7 meV and that increases to 55.2 meV for the $\text{Zn}_{0.87}\text{Cd}_{0.13}\text{O}$ sample.

Table 1. The values of E_{me} , E_0 , and E_a estimated from equation (2) and (3), respectively.

	E_{me} (eV)	E_0 (meV)	E_a (meV)
$\text{Zn}_{0.94}\text{Cd}_{0.06}\text{O}$	2.88	25.7	24.8
$\text{Zn}_{0.92}\text{Cd}_{0.08}\text{O}$	2.69	36.2	35.9
$\text{Zn}_{0.90}\text{Cd}_{0.10}\text{O}$	2.57	40.4	40.1
$\text{Zn}_{0.87}\text{Cd}_{0.13}\text{O}$	2.31	55.2	54.6

The lifetime τ of the PL decay as a function of the reciprocal temperature, for the ZnCdO thin films with Cd content of 0.06, 0.08, 0.10, and 0.13, are further plotted in figure 2(a)~(d), respectively. It decreases slowly at low temperature region, but rapidly at the high temperature region. The temperature-dependent lifetime can be fitted using the function [10]

$$\tau(T) = \frac{\tau_0}{1 + C * \exp(-E_a / kT)} \quad (3)$$

where τ_0 is the lifetime at low temperature, the coefficient C is a fitting constant, and E_a is the thermal activation energy. It is known that the carriers would be delocalized from the localized states in thermalization process, which results in the reduction of the carrier lifetime. Therefore the activation energy E_a means the localization energy of the localized carriers [10]. The values of E_a are estimated by fitting using equation (3) shown as the solid line in figure 2 and listed in table 1. The obtained values of E_a are in agreement with the values of E_0 , which confirm the delocalization of carriers from the localized states in the thermalization process.

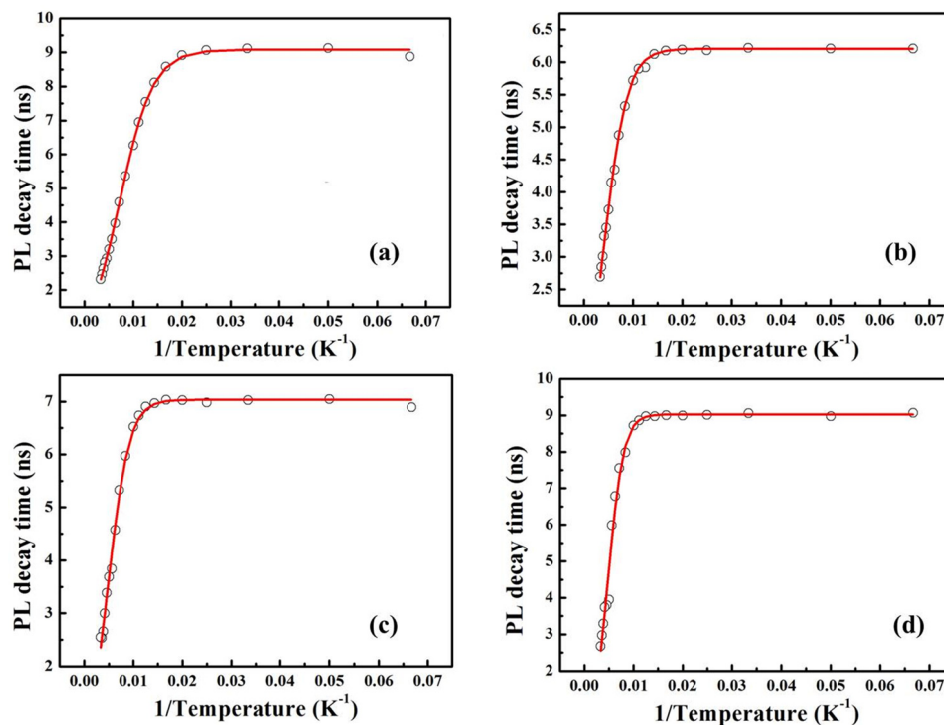


Figure 2. Arrhenius plot of the PL decay time of ZnCdO thin films with Cd content of (a) 0.06, (b) 0.08, (c) 0.10, and (d) 0.13. The solid line is a curve fitted using equation (3).

4. Summary

We have investigated the luminescence mechanism of the $\text{Zn}_{1-x}\text{Cd}_x\text{O}$ films with $x=0.06, 0.08, 0.10$, and 0.13 by PL and TRPL measurements. The emission-energy-dependent PL decay time shows that the PL decay is faster on the high energy side than on the low energy side of PL spectrum, exhibiting the characteristic of a localization effect. The temperature-dependent PL decay time presents that the carriers would be delocalized from localized states due to thermal activation. Our results indicate that the carrier localization would be increased with increasing Cd content, which dominates the luminescence mechanism in ZnCdO semiconductors.

5. References

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