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Journal of Luminescence 119-120 (2006) 551–555
JOURNAL OF LUMINESCENCE
www.elsevier.com/locate/jlumin

Effect of interface on luminescence properties in ZnO/MgZnO heterostructures

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Available online 28 February 2006

Abstract

A set of ZnO/MgZnO heterostructures with well widths, L_w, varying from 2 to 20 nm has been grown by plasma-assisted molecular-beam epitaxy (P-MBE). We present a room-temperature (RT) study of the well-width-dependent photoluminescence (PL) spectra and carrier lifetimes in ZnO/MgZnO heterostructures. Bi-exponential process is seen at RT time-decay curves of the luminescence from the well layers. The fast process is from the recombination of the free exciton, while the slow process is attributing to the recombination of the localized exciton in the interface of ZnO and MgZnO. It is also confirmed from the PL peak shift of time-resolved PL spectra. With decreasing the well thickness, the fast process gradually increases and dominantly contributes to the PL spectrum due to the interface improvement.

PACS: 68.65.Fg; 79.60.Jv; 78.47.+P; 32.70.Jz

Keywords: ZnO/MgZnO heterostructure; Interface effect; Time-resolved photoluminescence; Decay time

1. Introduction

Recently, ZnO-based semiconductors are attracting increasing attention as promising candidates for optoelectronic applications in ultraviolet (UV) regions [1,2]. Because ZnO/MgZnO heterostructure can realize double confinement actions for electrons and photons in optoelectronic devices, there are many reports about the growth and exciton-related lasing process with low threshold in the ZnO/MgZnO structure [3–5]. However, the emission mechanisms of this structure are still

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doi:10.1016/j.jlumin.2006.01.056

limited [6,7]. Understanding the emission mechanism of the ZnO/MgZnO structure is necessary for further improving the performance of the optical devices. Time-resolved photoluminescence (TRPL) spectroscopy provides a method for the direct measurement of exciton and carrier lifetimes in semiconductors and semiconductor structures. Lifetimes are critical to device efficiency and therefore overall device performance. Since most optical devices are used at room temperature (RT), the RT behavior of the systems at high carrier concentration is directly relevant for device operation. In this letter, we present a RT TRPL study of ZnO/MgZnO structure, through which we confirm the original of ZnO well layer. The photoluminescence (PL) and exciton lifetimes are effected by interface state obviously. The effect depended on the well thickness is systematically studied.

2. Experiment

Epitaxial films were grown by plasma-assisted molecular beam epitaxy (p-MBE) on *c*-plane sapphire. The growth temperature is 550 °C. After epitaxy of a 100-nm-thick $Mg_{0.10}Zn_{0.90}O$ barrier layer, a various of ZnO well layers, the thickness of which are vary from 1 to 20 nm, were sandwiched between the buffer layer and a 60 nm $Mg_{0.10}Zn_{0.90}O$ capping layer. The detail of the growth has been reported elsewhere [8].

In this article, we report on the studies of three samples: 2, 4, 20 nm well thickness heterostructures (referred as sample A2, A4, A20, respectively). The conventional PL measurements were performed using the 325 nm line of a He–Cd laser operating at a pump level of 50 mW. A JY Raman spectrometer equipped with a charge-coupled device was employed to detect the luminescence signals.

The TRPL was measured using the time-correlated single-photon counting technique with FL920-fluorescence lifetime spectrometer, and 300 nm line of nF900 nanosecond flashlamp was used as excitation source with a system resolution of 12 ps. The measurement system had lifetime range from 100 ps to 50 μs.

3. Results and discussion

The structural quality of these heterostructure was evaluated using X-ray diffraction (XRD) (which is shown elsewhere [8]). Fig. 1 shows the normalized PL spectra and absorption spectra of the samples with different well width at 300 K. For the absorption spectrum of sample A20, a high-energy absorption edge and a low-energy step (arrow in the figure) are observed, which are attributed to the absorption from MgZnO and ZnO layer, respectively. With decreasing the well width, the absorption from ZnO layer weakens. For sample A4, the absorption is a gentle curve. With decreasing the well width further, for sample A2, the absorption become sharper. For the RT PL spectra, two PL bands corresponding to the two absorptions are observed, respectively. The stronger emission at about 3.3 eV (a) and a weaker emission at about 3.5 eV (b) are attributed to the recombination from the MgZnO and ZnO layer, respectively. The absorption spectra and PL spectra confirm the formation of ZnO/MgZnO heterostructure. The PL peak (a) at low-energy side, which is from the radiative recombination of ZnO well layer, has a large blueshift (~40 meV) with decreasing well thickness from 20 to 2 nm. This phenomenon is attributed to the quantum confinement effect. The observed broadening of the linewidth for all samples should originate from

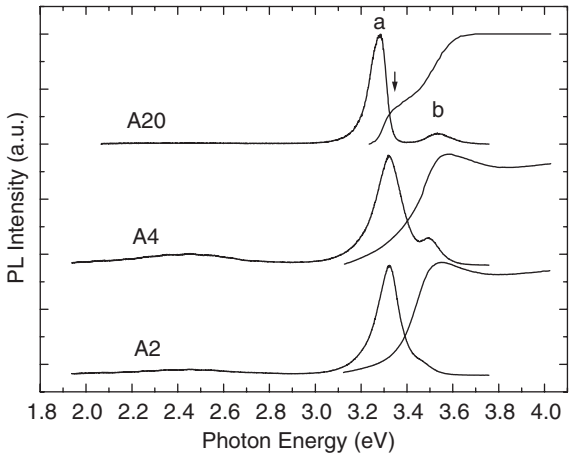

Fig. 1. The PL of the samples with different well thickness at RT.

the existence of interface-localized exciton emission. The temperature-depended PL spectra of A20 are measured as shown in Fig. 2. From the spectra, we find the intensity of peak (b) quenching rapidly with the increase in temperature while the intensity of the peak (a) quenching slower. The peak (a) position has a blueshift at the same time. It is considered that the localized exciton of MgZnO obtain enough active energy to delocalized and relax into the ZnO layer with increasing the temperature. For peak (a), the buleshift of peak position is due to the localized exciton of ZnO well layer delocalized to free exciton. It can be seen in Fig. 1, as ZnO layer thickness decreasing, the luminescence from MgZnO layer becomes weaker and weaker, which is attribute to the interface state reduce with the decrease of ZnO well layer thickness [9]. The PL from localized exciton of MgZnO and ZnO weaken.

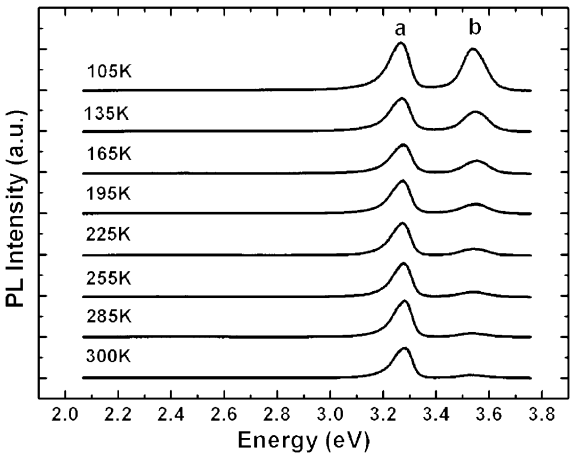

Fig. 2. The temperature-depended PL spectra of A20 (105–300 K).

Fig. 3 shows the time-decay curves of the luminescence from ZnO well layer at RT. The time-decay curves can be well described by a bi-exponential decay for all samples. For sample A20, a bi-exponential function fits the decay profile well with a reduced chi-square value, χ_r^2, of 1.024 compared with a single-exponential fit with χ_r^2 of 2. Time constants from the fit are 0.58 and 3.82 ns. Similarly, time constants of sample A4, A2 are 0.43 ns, 0.14 ns for fast process; and 3.43 ns, 1.83 ns for slow process. The short lifetime of 0.58 ns from sample A20 is comparable to the recombination lifetimes of 0.1 ns at 293 K for free excitons in ZnO films [9]. Such a long lifetime indicates very low density of non-radiative defects

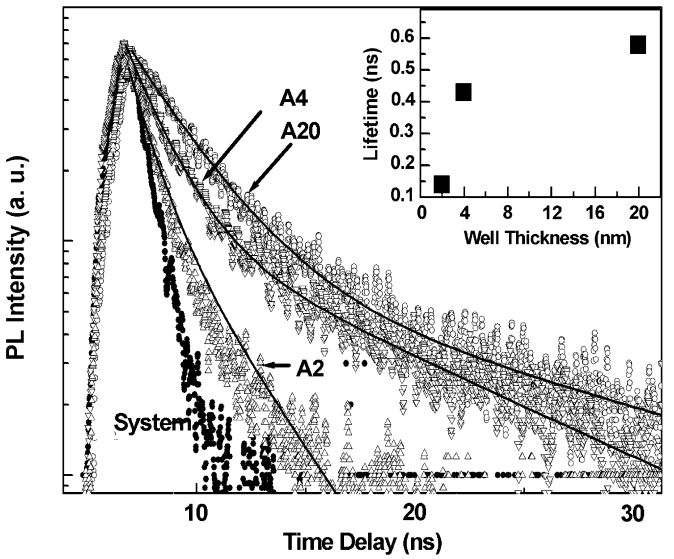

Fig. 3. Time-decay curve spectra of samples with different well thickness (A20 (○), A4 (▽), and A2 (△)). The solid curves are the least-squares fit of data with two exponential decay, $I(t) = I_1 \exp(-t/\tau_1) + I_2 \exp(-t/\tau_2)$. The inset shows the lifetime vs. well thickness for the three samples.

in well intra-layers. The slow lifetime observed in our quantum well is about several times of the fast lifetime that is contributed to free exciton. This clearly shows that the slow decay processes are not from the free state exciton. The slowdown in the exciton relaxation must be due to the trapping of exciton in potential fluctuation in the interface. Sugawara [10] has shown theoretically that lifetimes of such excitons are several times that of the free excitons. This kind of decay may suggest two different recombination channels. We prefer the following explanation: the well layer PL is from two components. The fast decay is assigned to the free exciton recombination. The slow decay is assigned to the localized exciton recombination. These phenomena have been seen in GaAs single quantum wells at 20 K [11]. The reduce of interface state with decrease of the well layer thickness, as have been described above, can also be confirmed with TRPL. The slow components from the interface-localized exciton contribute 69.1%, 60.4%, 33.4% of the PL signal for sample A20, A4, A2, respectively. The inset of Fig. 3 shows the luminescence lifetime as a function of well width. The shorter lifetime with decrease well thickness can be explained by the effect of the strong quantum confinement in thin well structure.

To further clarify the origin of recombination, the time-decay spectra were measured. Fig. 4 shows TRPL spectra monitored at various time after pulsed excitation of sample A2. The whole spectra are integrated during the same time interval, and are normalized in intensity. From the figure, with the increase in the decay time, the PL peak position shifts toward the low-energy side. The energy difference of the peak between (a) 0 ns and (d) 1.5 ns is about 30 meV. It confirms that the PL spectrum is consisted of two parts (free exciton at high-energy side and localized excitons at low-energy side) as explanation above. The recombination of free exciton is faster than localized exciton, with increasing time the free exciton recombination, the slow process of localized exciton dominant. This phenomenon existing at RT is attributing to the ZnO with high-exciton binding energy (60 meV), for localized excitons the binding energy is about 30 meV, the exciton and localized exciton is existed even at RT.

4. Conclusions

In summary, a set of ZnO/MgZnO heterostructures with well widths, L_w, varying from 2 to 20 nm has been grown by plasma-assisted molecular-beam

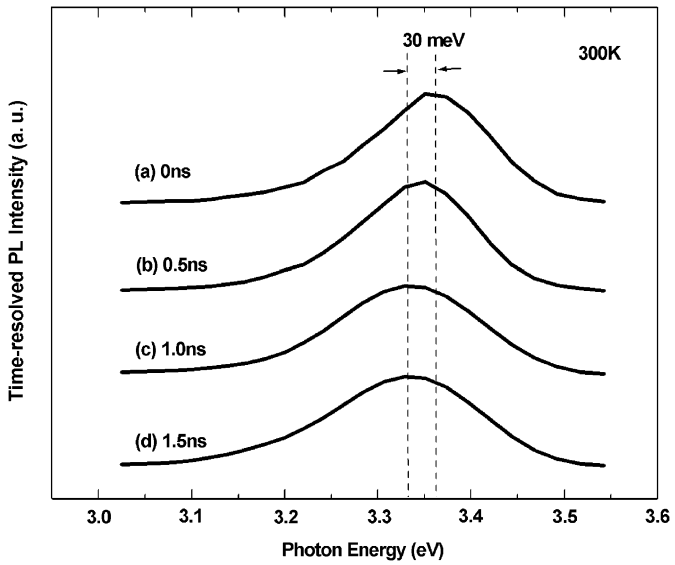

Fig. 4. Time-resolved PL spectra of sample A2.

epitaxy. RT TRPL has been observed in ZnO/MgZnO heterostructures. Bi-exponential process is seen at RT time-decay curves of the samples, which attribute to the fast process from the recombination of the free exciton, while the slow process from the recombination of the interface-localized exciton. In particular, it was shown that a decreasing of the well width results in reduction interface state. The localized exciton recombination is lower proportion in thin well structure.

Acknowledgments

This work is supported by the "863" High Technology Research Program in China, under Grant No. 2001AA311120, the Key Project of National Natural Science Foundation of China under Grant No. 60336020, Foundation for Prominent Youth Awarded by NNFCC, No. 60429403, the Innovation Project of Chinese Academy of Sciences, the National Natural Science Foundation of China under Grant Nos. 60278031, 60376009, and 50402016.

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