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Super-radiation processes of $ZnO/Mg_{0.1}Zn_{0.9}O$ SQW under high-density excitation

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Abstract

ZnO /$Mg_{0.1}Zn_{0.9}O$ single-quantum well (SQW) with 1.5 nm ZnO well width was grown on sapphire substrate by plasma-assisted molecular beam epitaxy. The absorption spectrum of the sample at room temperature (RT) shows a sharp absorption edge from MgZnO barrier layer in the ultraviolet region. A stronger exciton emission at 3.392 eV from the ZnO well layer was observed in the photoluminescence spectrum at RT, and the exciton binding energy of the ZnO well layer is 70.8 meV. In the PL spectra at 5 K, P_2 band (one exciton was scattered into an excited state with a quantum number 2) located at 3.371 eV was observed. At excitation densities higher than 76 kW/cm^2, the P band shows a typical super-radiation characteristic. With increasing the excitation density further, the P band converges gradually at the saturation value of 3.354 eV with significant narrowing. It was attributed to the gradually enhanced exciton-exciton scattering.

Keywords: Plasma-assisted molecular beam epitaxy; Photoluminescence; Exciton–exciton scattering; Super-radiation

1. Introduction

Since 1997, stimulated emission and optically pumped laser action from ZnO thin film were observed at room temperature (RT) [1]. ZnO with a band gap of 3.37 eV at RT and exciton binding energy of 60 meV has attracted much attention as promising candidates for optoelectric applications in visible and ultraviolet (UV) regions [2,3]. The origins of these stimulated emissions were originated from exciton–exciton (Ex–Ex) scattering or electron hole plasma (EHP) under different excitation conditions [4–6]. Recently, excitionc lasing with low thresholds at RT has been obtained in the heterostructure and quantum well (QW) structure of ZnO/ZnMgO. This is due to realizing double confinement actions for electrons and photons in the above structures. In order to accurately design photoelectronic device, it is important to understand the origin of PL spectra under different excitation densities, particularly, to study the formation process of the stimulated emission with increasing excitation density. In this letter, optical properties of ZnO /$Mg_{0.1}Zn_{0.9}O$ SQW under higher excitation density were studied. A super-radiation process caused by Ex–Ex scattering in ZnO/$Mg_{0.10}Zn_{0.90}O$ SQW was observed. We discussed the effect of excitation density on the behaving of the photoluminescence (PL).

2. Experiment

A V80 H molecular-beam epitaxy (MBE) system was employed for growing the $Mg_{0.10}Zn_{0.90}O$/ZnO SQW, where Knudsen effusion cells were used to evaporate elemental zinc with 99.9999% purity and Mg with 99.999% purity. Atomic oxygen was generated from ultra-pure O_2 gas (99.999%) activated by an rf atomic source with an electrostatic ion trap operating at 500 V during growth.

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Sapphire substrates were pretreated at 800 °C in ultrahigh vacuum ($<1\times10^{-8}$ mbar) for 30 min to remove the surface contaminants. The growth temperature is 550 °C. A 100-nm-thick $Mg_{0.10}Zn_{0.90}O$ buffer layer was grown on the substrates, and a ZnO well layer with 1.5 nm was sandwiched between the buffer layer and a 60 nm $Mg_{0.10}Zn_{0.90}O$ capping layer, which was labeled as sample C.

Photoluminescence spectra were measured at RT. The 325 nm line of a He–Cd laser with the power of 50 mW was used as the excitation source and a JY63 Raman spectrometer was employed to detect the luminescence signals. A pulse laser worked at 345 nm lines was used as the excitation source in high pumping density in low-temperature spectra measurements. The laser pulses were generated by an optical parametric amplifier (OPA) pumped by a Ti: sapphire regenerative amplifier. The OPA delivers at 1-kHz repetition rate with duration of 130 fs, and pulse energy of 0.11 μJ at 345 nm.

3. Results and discussion

Fig. 1 gives RT PL and absorption spectra of the grown sample. From Fig. 1, the absorption of ZnO /$Mg_{0.1}Zn_{0.9}O$ SQW mainly comes from MgZnO barrier layer. The PL spectrum shows that a strong exciton emission at 3.392 eV from the ZnO well layer and a weak emission at 3.531 eV from the MgZnO barrier layer. Compared with ZnO film sample, the exciton emission band of ZnO well layer shows an obvious blue shift due to quantum confinement effect. The insert of Fig. 1 shows the temperature dependence of the integrated intensity of the exciton emission band from 81 to 300 K. The experimental data (○) with increasing temperature is theoretically fitted using the following equation:

$$I(T) = I_0/[1 + A\ \exp(-E/k_B T)], \quad (1)$$

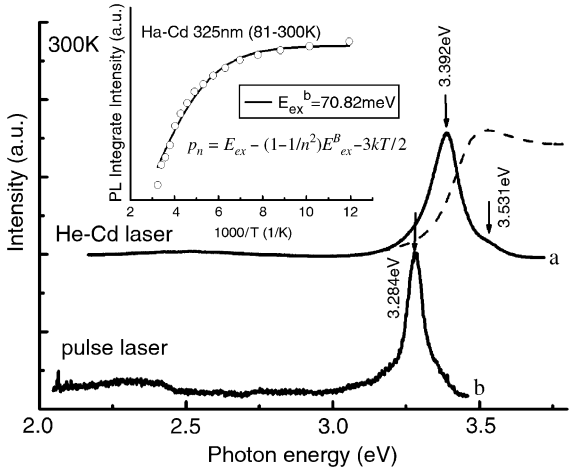

Fig. 1. The PL spectra (solid curve) of SQW sample at RT excited using (a) a continuous wave He–Cd laser, and (b) a pulsed Ti-sapphire laser. The RT absorption spectrum was given by dashed curve .The inset shows the integrated intensities of exciton emission band as a function of temperatures ranging from 81 to 300 K.

where E is the activation energy of the thermal quenching process, k_B is Boltzman constant, I_0 is the emission intensity at 0 K, T is a thermodynamic temperature and A is a constant [7]. The solid line gives the result of theoretical fitting to the experimental data and we obtain binding energy of exciton confined in two dimensions is 70.8 meV. It is consistent with the value reported by Gruber et al. [8] in which the exciton binding energy of QW is higher than that of ZnO bulk crystal (59 meV [1]). The above result confirms further the formation of the QW structure.

Fig. 1 also exhibits the RT PL spectrum of the sample under high excitation density of 140 kW/cm^2 using the pulse laser. Note that the emission peak energy of 3.284 eV is lower than that of 3.392 eV excited by He–Cd laser. This emission peak under high excitation density is considered to be result from inelastic Ex–Ex collision [8]. One exciton was scattered into an excited state with a quantum number n, while another one was recombined by emitting a photon. The photon energies formed in this process were given by

$$p_n = E_{ex} - (1 - 1/n^2)E_{ex}^B - 3kT/2, \quad n = (2, 3, 4, \ldots), \quad (2)$$

where p_n is the emitted photon energy, E_{ex} is the exciton emission energy, E_{ex}^B is the binding energy of the exciton, n is the quantum number of the envelope function, and the thermal energy $3kT/2$ is about 38.7 meV at 300 K [1,8].

The calculated value $(1-1/n^2)\ E_{ex}^B$ according to formula (1) is 69.3 meV, which is close to the $E_{ex}^B = 70.8$ meV.This means that Ex–Ex scattering leads to one exciton scattered into higher exciton state near continuous states. Therefore, the RT emission band at 3.284 eV under high pumping density (140 kW/cm^2) is attributed to P_μ-band emission (one exciton scattered into continuum states) caused by Ex–Ex collision.

To investigate further the process of the exciton scattering, the PL spectra at 5 K were measured with different excitation densities, as shown in Fig. 2. At the lower pumping intensity, two spontaneous emission bands were observed. The emission at low-energy side was attributed to the P_2 band due to Ex–Ex collision, where one exciton emitted a photon by the radiative recombination and another one was scattered into $n = 2$ state [3]. When the excitation intensity increased from 76 to 140 kW/cm^2, the P_2 band at 3.371 eV shifts gradually to a saturation value of 3.352 eV.The energy shift of 19 meV is close to the maximum peak shift ($E_{ex}^n - E_{ex}^2$) of 17.8 meV, which suggests the P-band excited under 140 kW/cm^2 is dominated by P_∞-band. The inset of Fig. 2 gives the integrated emission intensity (I) versus excitation density (I_{ex}). From Fig. 3, we found the superlinear increase of the emission intensity as the excitation density is increased above 76 kW/cm^2, which shows clearly I follow the relation of $I_{ex}^{3.13}$. This superlinear increase indicates the typical properties of a super-radiation recombination. The energy position and line width of P band at different excitation density are shown in Fig. 3. The significant red shift and

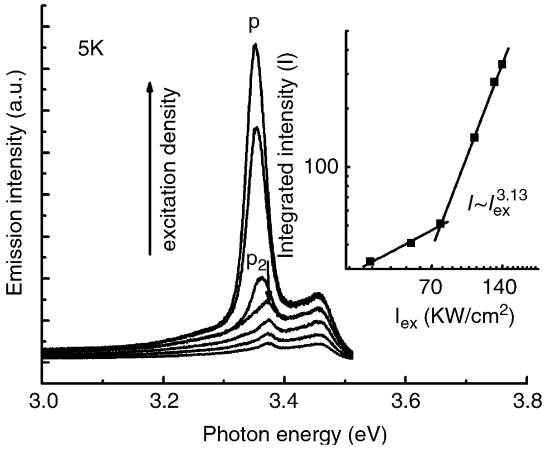

Fig. 2. The evolution of P band emission as the excitation intensity increases from 38 to 140KW/cm^2 at 5 K. The inset shows the integrated intensities of P bands as a function of the excitation densities.

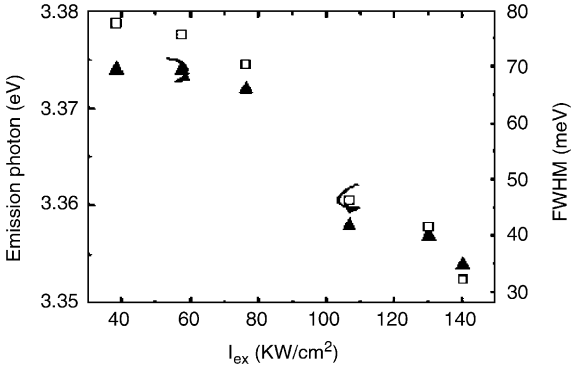

Fig. 3. The FWHMs and positions of P bands in Fig. 2 as a function of the excitation densities.

the narrowing are attributed to enhancement of Ex–Ex scattering process with higher excited state as increases the excitation density. Fig. 3 shows that the energy of the band at high-energy side is larger than that of the free exciton emission, the detailed reason is not clear now and need to be investigated in further.

4. Conclusions

In summary, ZnO/ZnMgO SQW sample with well width of 1.5 nm was grown on sapphire substrate by P-MBE. At RT, a sharp absorption edge from MgZnO barrier layer and a stronger exciton emission at 3.392 eV from the ZnO well layer indicates that the sample has high optical quality. Under high excitation density the Ex–Ex scattering dominantly contributes to the PL spectrum, P_2 band caused by Ex-Ex scattering were observed at lower excitation density. Under higher excitation density above 76 kW/cm^2, the P_∞ band gradually plays main role in the spectrum and shows a superradiational characteristic.

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