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Temperature dependence of carrier transfer and exciton localization in ZnO/MgZnO heterostructure

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Abstract

MgZnO/ZnO heterostructure was fabricated on sapphire substrate by plasma assistant molecular beam epitaxy. The micro-photoluminescence spectra of sample are reported, which shows that different emission peaks would appear when the laser beam focuses different deepness in the film. A carrier tunneling process from the MgZnO capping layer to ZnO layer was observed by the measured temperature dependence of photoluminescence spectra. This induces the emission intensity of the ZnO grew monotonically from 81 to 103 K.

Keywords: Zno; Heterostructure; Carrier transfer; Exciton localization

1. Introduction

Recently, great interest has been paid to wide band-gap semiconductor materials because short wavelength devices are of intense interest for future opto-electron technical industries [1,2]. ZnO-based semiconductors are recognized as very promising materials due to the potential application as ultra-violet (UV) light-emitting diodes (LED) and laser diodes [2]. Compared with other wide band-gap materials, such as ZnSe, GaN, and SiC, etc., ZnO has many advantages for device applications including higher chemical and thermal stability, higher radiation hardness and lower growth temperature, which are suitable for fabricating long-lifetime devices. Moreover, ZnO has a very large exciton binding energy (60 meV), which allows efficient excitonic emission at high temperature. As well known, a lasing process based on the excitonic effect has a higher gain and a lower

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threshold than that of electron–hole plasma. This makes it desirable to develop optoelectronic devices based on excitonic effect [1,2].

The key technique to obtain the high-performance LED devices is to construct a heterojunction to realize double confinement actions for electrons and photons in optoelectronic devices. A solid solution of MgZnO, of which band gap is larger than ZnO, is a suitable barrier layer for ZnO because its lattice constant is close to that of ZnO. With the advantage in the epitaxial technology of ZnO and related alloys, ZnO/MgZnO multiquantum well (MQW) structures are fabricated on sapphire and $ScAlMgO_4$ substrates successfully [3–5], and the UV lasing based on them with low threshold of 11 KW/cm^2 was reported [5]. However the photoluminescence properties and the carrier injection process from MgZnO layer to ZnO layer have not been fully detected in the simple MgZnO/ZnO/MgZnO heterostructure, although it is important to extensively understand the emission process and crucial to device applications.

In this letter, a micro-photoluminescence spectrum was employed to examine the emission properties of MgZnO/ZnO/MgZnO heterostructure. A carrier transfer and an exciton localization process were observed in temperature-dependent photoluminescence (PL) spectra.

2. Experimental

The growth was carried out using a V80 H molecular-beam epitaxy system equipped with Knudsen-cells for Zn solid source (99.9999%) and Mg solid source (99.999%), and a radio frequency (RF)-plasma source for oxygen. Base pressure was about 1×10^{-10} Torr with a liquid nitrogen supply. The growth temperature was 650 °C. The RF oxygen plasma was generated under conditions of a power of 300 W and a flow rate of 2.0 sccm, which gave rise to a growth pressure of 2×10^{-5} Torr. First a 100 nm thick $Mg_{0.15}Zn_{0.85}O$ buffer layer was grown on a sapphire substrate. Then a 20 nm thick ZnO was sandwiched between the buffer layer and a 30 nm thick $Mg_{0.15}Zn_{0.85}O$ capping layer. The Mg concentration was confirmed by detecting X-ray diffraction and transmittance spectrum corresponding to a band offset of about 0.35 eV above pure ZnO at the heterointerface. In photoluminescence measurement, a continuous wave He–Cd laser of 325 nm (50 mw) was used as an excitation source and a JY Microlaser Raman spectrometer made in France. By adjusting the position of an ultraviolet lens ($\times 10$ UV), equipped with the micro-Raman system, we can change the deepness of the laser beam focus.

3. Results and discussion

Fig. 1 shows the PL spectra of MgZnO/ZnO heterostructure measured at room temperature under different penetration depths of the laser beam. When the laser beam is focused on the surface of the sample an intense UV emission peak located at 3.55 eV (denoted by 1) is observed (curve A), which originates from the radiative recombination of free exciton in the $Mg_{0.15}Zn_{0.85}O$ capping layer [6]. The full-width at half-maximum (FWHM) of peak 1 is 69 meV. There is also an obvious shoulder located at the low energy side of peak 1, which originates from the peak at 3.46 eV with the FWHM of 105 meV. The peak energy of this shoulder is between the energies of the peaks 1 of the $Mg_{0.15}Zn_{0.85}O$ capping layer and pure ZnO, which is assumed to be due to the exciton recombination in the interface. With the focus of the laser beam being removed into the film the peak 1 decreases gradually, and a new emission band (denoted by 2) located at 3.34 eV is dominant in the spectra (curve B) with the FWHM of 95 meV. There is no more emission band in the visible region in both of the spectra. The peak 2 can be assigned as originated from the ZnO layer. Because the capping layer (30 nm) is thicker than ZnO layer, the emission from $Mg_{0.15}Zn_{0.85}O$ layer could still be seen in curve B.

The emission spectra of MgZnO top layer at low temperature are the same as those of other reports when the laser beam is focused on the surface [6,7]. But when the laser focus is moved to the ZnO layer, the spectra are quite different. Fig. 2 shows the temperature-dependent PL spectra, in which

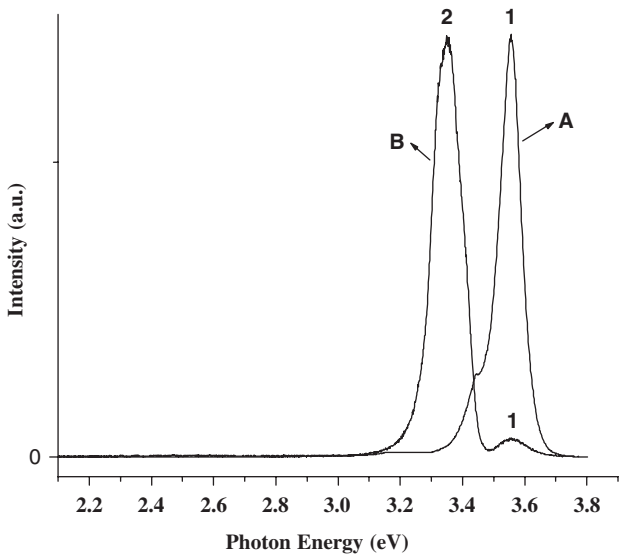

Fig. 1. Room temperature photoluminescence spectra of ZnO/$Mg_{0.15}Zn_{0.85}O$ heterostructure with the excitation laser beam focus adjusted on $Mg_{0.15}Zn_{0.85}O$ capping layer (A) and ZnO layer (B).

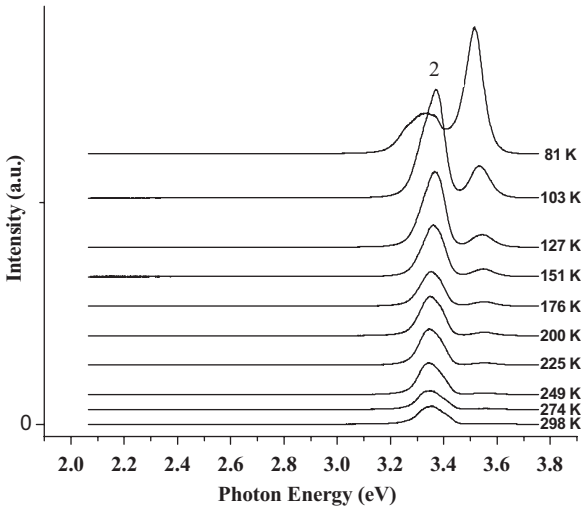

Fig. 2. Temperature dependence of photoluminescence spectra with the excited laser beam focused on ZnO layer of ZnO/$Mg_{0.15}Zn_{0.85}O$ heterostructure.

the laser beam is focused on the ZnO layer. At all temperature range from 81 to 298 K there are two peaks in the spectra (marked as 1 and 2), which originate from $Mg_{0.15}Zn_{0.85}O$ and ZnO, respectively. At 81 K the emission centers of these two peaks are 3.52 and 3.33 eV, respectively, and peak 1 is dominant in the spectra. The peak energies are lower than the photon emission energies at room temperature. With increasing the temperature the intensity of peak 1 decreases sharply. At 103 K peak 2 becomes stronger than peak 1 and finally dominant the spectra when the temperature exceeds 127 K.

The peak energy and emission intensity as a function of temperature are shown in Fig. 3(a) and (b). For the peak 1 the emission energy shows a clear blueshift over all temperature regions. At the same time the photon emission energy of the peak 2 moves to the high-energy side between 81 and 151 K, then decreases monotonically with increasing the temperature. This typical temperature-induced emission energy blueshift can be easily found in InGaN/GaN quantum-well structure, which means that there exists a strong exciton-localization effect [8–10]. In our experiment the interface is assumed to play an important role. Due to the strain effect in the interface, some localization states are formed near the band edge of both $Mg_{0.15}Zn_{0.85}O$ capping layer and ZnO layer.

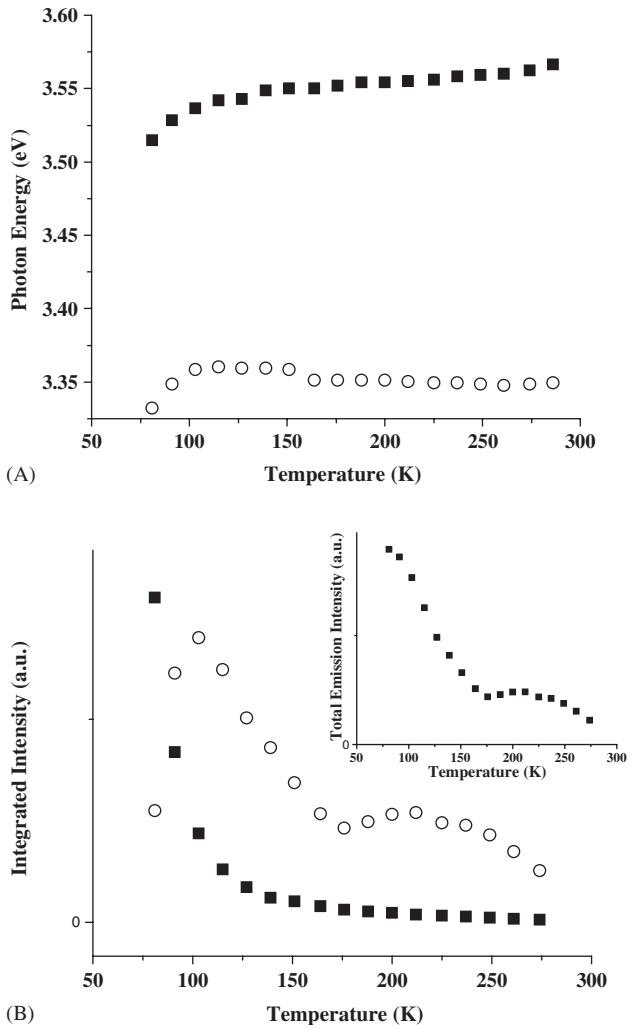

Fig. 3. (A) Peak position of 1 (filled squares) and 2 (open dots) as a function of temperature. (B) Temperature depended emission intensities of band 1 (filled squares) and 2 (open dots). The inset shows the total integrated intensity of both bands 1 and 2.

Fig. 3(b) gives the emission intensity of the peaks 1 and 2 as a function of the temperature. The intensity of the peak 1 decreases exponentially with increasing the temperature due to the thermally activated nonradiative recombination mechanisms. But for the peak 2 there are two intensity increasing regions: one is from 81 to 103 K, the other is from 176 to 211 K. The total integrated intensity of both 1 and 2 versus temperature is shown in the insert of Fig. 3(b). Between 81 and 176 K the intensity decreases almost linearly. When the temperature exceeds 176 K the intensity increases and reaches a maximum value at 212 K.

Based on the above experiment a carrier transfer process is considered from $Mg_{0.15}Zn_{0.85}O$ capping layer to ZnO layer. We have observed that the excitonic nonradiative recombination of ZnO and MgZnO is prominent in this temperature region. If this process is due to the exciton transfer the total emission intensity should decrease with increasing the temperature. Therefore, only the carrier

transfer process is assumed. There are two kinds of carrier transfer processes: tunnelling transfer, which occurs when the carrier energy is lower than the barrier height, and classical activation transfer, due to thermal activation across the barrier, which is negligible at the low temperatures considered in this section. Due to the effect of the different carrier injection processes the emission intensity of ZnO layer could be changed. With raise in temperature, excitons become free carriers. Because the band-gap of ZnO is 0.35 eV lower than that of $Mg_{0.15}Zn_{0.85}O$, these free carriers can be tunneling-injected into ZnO layer. Due to this process the emission intensity increases greatly. For the second increasing process a barrier must exist in the interface. At low temperature the energy of free carriers is low. With increasing the temperature the carriers obtain enough active energy to exceed the barrier and defuse into the ZnO layer. This process results in the increase of the emission intensity of the peak 2 from 176 to 211 K.

In summary, microphotoluminescence spectra and temperature-dependent PL spectra of MgZnO/ZnO heterostructure were reported. This heterostructure was fabricated on sapphire substrate by plasma assistant molecular beam epitaxy (P-MBE). In microphotoluminescence spectra the dominant emission peak was varied depending on the focus position of the laser beam and a carrier injection process from the $Mg_{0.15}Zn_{0.85}O$ layer to the ZnO layer was observed at low temperature region.

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