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Spontaneous and stimulated emission of $ZnO/Zn_{0.85}Mg_{0.15}O$ asymmetric double quantum wells

S.C. Su [a,b], Y.M. Lu [b,c,*], G.Z. Xing [d], T. Wu [d]

[a] Key Laboratory of Electroluminescent Devices, Department of Education of Guangdong Province, Institute of Opto-electronic Materials and Technology, South China Normal University, Guangzhou 510631, PR China

[b] Key Laboratory of Excited State Processes, Changchun Institute of Optics, Fine Mechanics and Physics, Chinese Academy of Sciences, Changchun 130033, PR China

[c] College of Materials Science and Engineering, Shenzhen University, Shenzhen 518060, PR China

[d] Division of Physics and Applied Physics, School of Physical and Mathematical Sciences, Nanyang Technological University, Singapore

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ABSTRACT

$ZnO/Zn_{0.85}Mg_{0.15}O$ asymmetric double quantum wells (ADQWs) were fabricated on an *m*-plane Al_2O_3 substrate by plasma-assisted molecular beam epitaxy (P-MBE). The ADQW structures were confirmed by comparing the photoluminescence (PL) spectra of the $ZnO/Zn_{0.85}Mg_{0.15}O$ MQWs and $ZnO/Zn_{0.85}Mg_{0.15}O$ ADQWs. The exciton tunnelling properties of the ADQWs were studied by means of temperature-dependent PL spectra. The carrier tunneling through the thin barrier is conducive to stimulated emission in the wide wells (WWs) of the ADQWs. The origin of the stimulated emission is exciton–exciton scattering in the WWs of ADQWs.

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1. Introduction

ZnO and its alloys, such as ZnMgO and BeZnO, have recently attracted great attention due to their potential applications in ultraviolet light emitting diodes (LEDs) and laser diodes (LDs) [1–4]. Due to the toxicity of BeO, much effort has been devoted to the fabrication and understanding of ZnO/MgZnO quantum wells. For normal ZnO/ZnMgO quantum wells including the single quantum wells (SQWs) [5,6] and multiple quantum wells (MQWs) [7–9], the excitonic processes and luminescence have been widely investigated. For special kinds of quantum wells—asymmetric double quantum

* Corresponding author at: Key Laboratory of Excited State Processes, Changchun Institute of Optics, Fine Mechanics and Physics, Chinese Academy of Sciences, Changchun 130033, PR China.
E-mail address: ymlu@szu.edu.cn (Y.M. Lu).

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wells (ADQWs), which consist of wells of two different widths, a wide well (WW) and a narrow well (NW), coupled by a thin barrier—the excitonic processes and luminescence also attract great attention related to the coupling between the two wells [10]. The exciton recombination in ADQWs is more interesting because it is modulated by the exciton tunneling [11,12]. The research on ADQWs has been proceeding for a long time [10,13–15]. However, there are only very few reports concerning the ZnO/ZnMgO ADQWs.

In the work described in this paper, the ZnO/$Zn_{0.85}Mg_{0.15}O$ ADQWs were grown on an *m*-plane $Al2O_3$ substrate by P-MBE. The spontaneous and stimulated emission of the ZnO/$Zn_{0.85}Mg_{0.15}O$ ADQWs was studied.

2. Experimental details

The ZnO/$Zn_{0.85}Mg_{0.15}O$ ADQWs samples were grown on an *m*-plane Al_2O_3 substrate by P-MBE at 650 °C. Elemental Zn (6N) and Mg (5N) were evaporated using conventional effusion cells. Pure oxygen (5N) was used as the oxygen source and oxygen plasma was generated through a radio frequency (rf) activated radical cell. The rf power of the oxygen plasma was 300 W. Before growth, the substrates were inserted into an ultrahigh vacuum chamber and annealed at 800 °C for 30 min, which was expected to remove the surface contaminants. The structure consists of a 50 nm $Zn_{0.85}Mg_{0.15}O$ buffer layer followed by five periods of ZnO/$Zn_{0.85}Mg_{0.15}O$ ADQWs and then a 50 nm $Zn_{0.85}Mg_{0.15}O$ cap layer. Each period of ZnO/$Zn_{0.85}Mg_{0.15}O$ ADQWs includes one narrow ZnO well, one thin $Zn_{0.85}Mg_{0.15}O$ barrier and one wide ZnO well, which will be denoted later as $L_n/L_b/L_w$, where L_n, L_b and L_w are the widths of the narrow well, the thin barrier and the wide well, respectively. Each period of the ADQW was separated by a 40 nm $Zn_{0.85}Mg_{0.15}O$ barrier.

Photoluminescence (PL) spectra were excited by the 325 nm line of a He–Cd laser with output power 50 mW. Energy dispersive spectroscopy (EDS) was used to determine the Mg contents in the ZnMgO barrier. The stimulated emission experiments were performed using the pulse laser output (325 nm) from an optical parametric amplifier (OPA) in an active–passive mode locked femtosecond Ti:sapphire laser operating at a repetition rate of 1 kHz. Emission from the sample edge was collected into a spectrometer (the spectral resolution was approximately 0.5 nm) and detected using an electrically cooled charge-coupled device.

3. Results and discussion

The structure of a single-period ADQW that consists of two wells of different widths coupled by a thin barrier is shown in Fig. 1(a). Fig. 1(b) shows the reflection high energy electron diffraction (RHEED) spectra of the ZnO well layers. The ZnO well layers are produced by two-dimensional growth. The RHEED reveals the high quality of the ZnO/$Zn_{0.85}Mg_{0.15}O$ ADQWs. To provide confirmation of the structure of the ADQWs further, 86 K PL spectra of the ZnO/$Zn_{0.85}Mg_{0.15}O$ MQWs with 3 nm, 6 nm well widths and 3 nm/6 nm/6 nm ZnO/$Zn_{0.85}Mg_{0.15}O$ ADQWs are shown in Fig. 2. The emission peaks of the 3 nm, 6 nm MQWs are consistent with the NWs (3 nm) and WWs (6 nm) of the ADQWs, respectively. Comparison of the PL spectra of MQWs and ADQWs indicated that the ZnO/$Zn_{0.85}Mg_{0.15}O$ ADQW structure was obtained.

In order to further the understanding of the PL properties of the ADQWs, the temperature-dependent PL spectra of the 3 nm/6 nm/6 nm ADQWs are displayed in Fig. 3. The weak emission peak at 3.60 eV is attributed to the luminescence of the $Zn_{0.85}Mg_{0.15}O$ barrier layers. The emission peaks at 3.398 and 3.440 eV are attributed to the 6 nm WWs and 3 nm NWs at 66 K, respectively. It is obvious that the emission from the WWs dominates the spectrum. The main cause for the difference between the emission intensities of NWs and WWs is the exciton tunnelling from the NWs to the WWs. Most of the excitons excited in an NW tunnel through the thin barrier to a WW, which induces the difference between the exciton distributions in the NW and WW. The PL peak of the NW disappeared with increase of temperature. The reason is that increasing the temperature would enhance the exciton tunnelling rate. On the other hand, temperature also influences the stability of excitons. The thermal dissociation of excitons will reduce the PL intensity of the NW. Since the ZnO has a high exciton binding energy (60 meV), the thermal dissociation of excitons can be neglected.

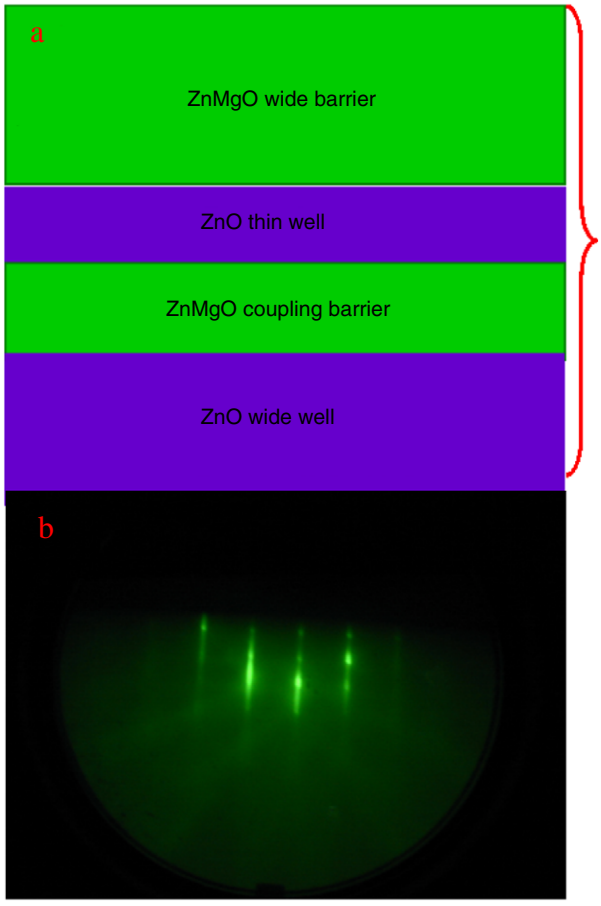

Fig. 1. (a) The structure of single-period $ZnO/Zn_{0.85}Mg_{0.15}O$ ADQWs. (b) The RHEED spectra of the ZnO well layer of the $ZnO/Zn_{0.85}Mg_{0.15}O$ ADQWs.

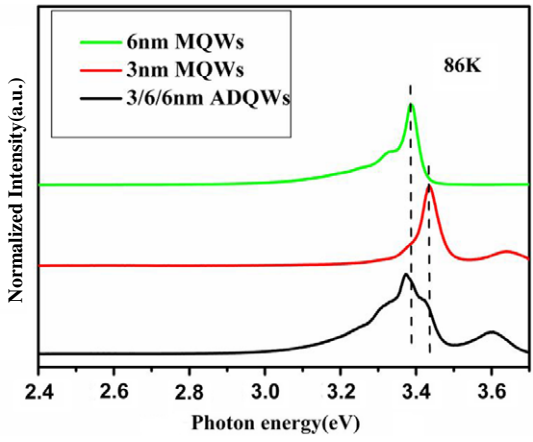

Fig. 2. The 86K PL spectra of the $ZnO/Zn_{0.85}Mg_{0.15}O$ MQWs with 3 nm, 6 nm well widths and 3 nm/6 nm/6 nm $ZnO/Zn_{0.85}Mg_{0.15}O$ ADQWs.

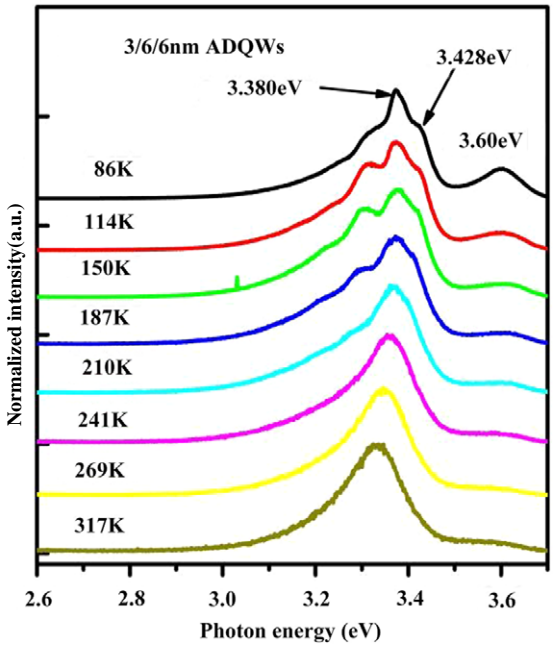

Fig. 3. The temperature-dependent PL spectra of the 3 nm/6 nm/6 nm ADQWs.

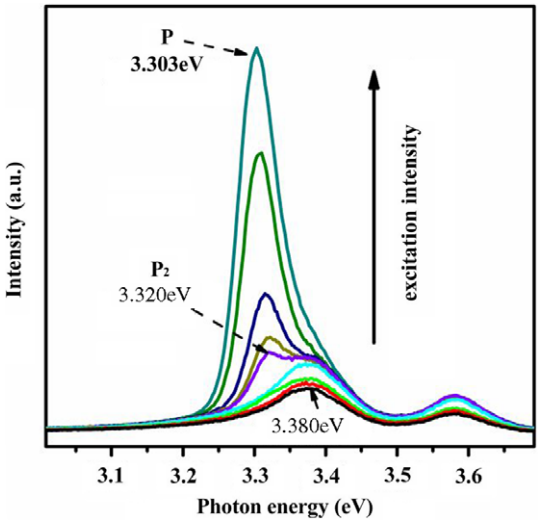

Fig. 4. The evolution of the *P* band emission as the excitation intensity increases from 26 to 90 kW/cm^2 at 66 K.

The exciton tunneling from the NWs to WWs leads to the carrier concentration increasing in the WWs of the ADQWs. It is possible to realize a low threshold stimulated emission in WW. Fig. 4 shows the stimulated emission spectra of 3 nm/6 nm/6 nm ADQWs, measured under the pulsed laser output of the OPA at 66 K. At low pumping intensities, the spontaneous emission bands were observed at 3.380 eV. At the excitation density of 64 kW/cm^2, a new peak (P_2) is observed at 3.320 eV, which is lower than the spontaneous PL peak. As the excitation density increases further to 90 kW/cm^2, a

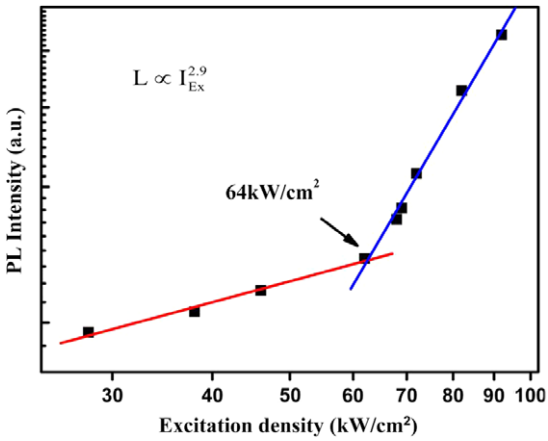

Fig. 5. The integrated intensity of the stimulated emission as a function of the excitation density.

sharp peak (P) at 3.303 eV emerges rapidly from the lower energy side of the P_2 peak and dominates the spectrum. Fig. 5 gives the integrated emission intensity (L) versus the excitation density (I_{ex}). The interdependence of L and I_{ex} can be fitted with $L \propto I_{ex}^{2.9}$. Thus, the superlinear increase of the P peak intensity, as well as the narrow linewidth with the total suppression of the other emissions, clearly indicates that stimulated emissions have occurred in this sample. The threshold is about 64 kW/cm^2. Comparing with previous reports on stimulated emission from ZnO MQWs [3,9], the positions of the P and P_2 peaks are in good agreement with what is expected from an inelastic collision between excitons, in which one of the two excitons obtains energy from the other and scatters into a higher exciton state with a quantum number $n > 1$, while the other recombines radiatively. The photons emitted in this process have the energies of P_n given by

$$P_n = E_{ex} - E_{ex}^b \left(1 - \frac{1}{n^2}\right) - \frac{3}{2}kT \quad (n = 2, 3, \ldots, \infty)\,, \tag{1}$$

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. If the value of E_{ex} is estimated to be 3.380 eV from the spontaneous emission peak, the exciton binding energy is determined as about 69 meV using Eq. (1) with $P_n = 3.303$ eV. Then, the energy difference between P_∞ and P_2 is calculated to be about 17 meV using Eq. (1) with $E_{ex}^b = 69$ meV. This calculated value is in good agreement with the observed peak shift between the P and P_2 emissions in Fig. 4. Thus, the mechanism of the stimulated emission studied in this paper can be reasonably attributed as inelastic exciton–exciton scattering.

Carrier localization is an ordinary and standard technique for achieving a reduction of the stimulated emission threshold. It can be achieved using ZnO/ZnMgO MQWs. Ohtomo realized a low threshold (about 11 kW/cm^2) in 3 nm ZnO/ZnMgO MQWs on $ScMgAlO_4$ [3]. But the most important feature is the match of $ScMgAlO_4$ to ZnO, and that $ScMgAlO_4$ is difficult to obtain. In our early work, we obtained stimulated emission in ZnO/ZnMgO single quantum wells on Al_2O_3, but the threshold is high (about 76 kW/cm^2 at 5 K) [16]. The threshold is 64 kW/cm^2 at 66 K in ADQW structures. The most likely reason for this is that the ADQWs have special structures in comparison with the MQWs. The exciton tunneling from the NWs to WWs of the ADQWs leads to the carrier concentration increasing in WWs of the ADQWs, and realizes low threshold stimulated emission. The large internal electric field expected in ZnO/ZnMgO QWs may provide other mechanisms for lowering thresholds in ADQWs. The exact explanations need further study.

4. Conclusion

In summary, $ZnO/Zn_{0.85}Mg_{0.15}O$ ADQWs were fabricated by P-MBE on m-Al_2O_3. The spontaneous and stimulated emission of the ADQWs was studied. The exciton tunnelling properties of the ADQWs

were studied by means of temperature-dependent PL spectra. The carrier tunneling through the thin barrier is conducive to stimulated emission in WWs of ADQWs. This work would be very important for the design of semiconductor lasers.

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