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Low-threshold lasing action in an asymmetric double ZnO/ZnMgO quantum well structure

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$ZnO/Zn_{0.85}Mg_{0.15}O$ asymmetric double quantum well (ADQW) and multiple quantum well (MQW) were fabricated with plasma assisted molecular epitaxy on c-plane sapphire, with their optical properties and optical pumped lasing characteristics studied. Due to the good crystalline quality, the lasing threshold of the MQW is $\sim 20\,\mathrm{kW\,cm^{-2}}$. The widths of the narrow well (NW) and the wide well (WW) of the ADQW were chosen to fascinate rapid LO phonon assisted carrier tunneling from NW to WW, so as to enhance the exciton density at the WW. Very low lasing threshold of $6\,\mathrm{kW\,cm^{-2}}$ has been achieved. © *2013 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4822265]

Because of its wide band gap energy (3.37 eV at room temperature), large exciton binding energy (60 meV), and high optical gain ($300\,\mathrm{cm^{-1}}$), ZnO has attracted extensive studies in its application on ultra-violet (UV) laser diode.[1,2] Multiple quantum well (MQW) structure can be used as the active region for the semiconductor laser device. Reducing the lasing threshold is crucial for the advancement of wide bandgap semiconductor laser device. Previous optically pumped stimulated emission studies performed on ZnO/ZnMgO MQW show that the lasing threshold is dependent on the QW quality and has typical value of $\sim 100\,\mathrm{kW\,cm^{-2}}$ to $\sim 600\,\mathrm{kW\,cm^{-2}}$.[3–6] A low threshold of $11\,\mathrm{kW\,cm^{-2}}$ was achieved while the ZnO/ZnMgO QW structure was grown on the lattice matching substrate $ScAlMgO_4$.[7]

The basic structure of asymmetric double quantum well (ADQW) consists of two wells with distinctive widths (a wide well (WW) and a narrow well (NW)) separated by a thin barrier (Figure 1(a)). Theoretical and experimental studies on exciton tunneling in II-VI ADQW[8,9] showed that exciton tunneling as a whole entity through the LO phonon assisted tunneling was slow. However, exciton tunneling through indirect state in a two-step process would have high efficiency. LO assisted electron tunneling from NW to WW is rapid if the energy separation between the lowest electron subbands in NW (N_{e1}) and WW (W_{e1}) is larger than the LO-phonon energy (i.e., $\Delta E_{e1} = E(N_{e1}) - E(W_{e1}) > \hbar\omega_{LO}$ as shown in Figure 1(b)). The electron-tunneling rate increases with ΔE_{e1}, reaching the maximum while $\Delta E_{e1} \approx \hbar\omega_{LO}$, and further increasing ΔE_{e1} would decrease the electron-tunneling rate.[10] For the case of the ZnCdSe/ZnSe ADQW with $\Delta E_{e1} = 47\,\mathrm{meV}$ and $\hbar\omega_{LO} = 31.7\,\mathrm{meV}$, Ten *et al*.[9] reported that LO phonon assisted electron tunneling could occur through two possible transitions, namely $(N_e,N_{HH}) \rightarrow (W_e,N_{HH})$ and $(N_e,W_{HH}) \rightarrow (W_e,W_{HH})$, which had short tunneling time as small as $\sim$300 fs. Hole tunneling from NW to WW is not possible in a single particle picture because of the large effective hole mass. However, because of the Coulomb enhancement originated from the strong interaction between the electron and hole in the ZnSe based materials having a large exciton binding energy, LO-phonon assisted tunneling of hole from NW to WW is feasible if the electron is at the WW. The tunneling time for the process $(W_e,N_{HH}) \rightarrow (W_e,W_{HH})$ was found to be $\sim$1 ps. With the carriers being excited, majority of the excited carriers in the NW tunnel to the WW before the electron-hole recombination occurring at the NW because the carrier tunneling time is smaller than the carrier recombination time. This implies that majority of the excited electrons and holes will accumulate and recombine at the WW, and thus, would lead to the lowering of the lasing threshold as compared to the symmetric QW.[11]

The present study aims to fabricate low lasing threshold ZnO/ZnMgO ADQW structure through: (1) improving the crystalline quality of the film, and (2) designing the ADQW structure so as to achieve the criteria for the rapid exciton tunneling from the NW to the WW. With the growing parameters being optimized, high quality ZnO/ZnMgO ADQW was grown on c-plane sapphire substrate using the plasma-assisted molecular beam epitaxy (PAMBE) method. The ADQW structure was designed so that $\Delta E_{e1} = 80\,\mathrm{meV}$, which is larger than and close to the phonon energy of $ZnO \sim 70\,\mathrm{meV}$ to fascinate the LO phonon assisted tunneling of electron and hole. The thus fabricated ADQW structure has a low lasing threshold of $6\,\mathrm{kW\,cm^{-2}}$ measured at the room temperature in an optical pumping experiment.

The ADQW structure was grown on the c-plane Al_2O_3 substrates by PAMBE at 650 °C. The background pressure was 10^{-7} Pa. Zn (6N) and Mg (5N) were evaporated from the conventional effusion cells. Using 5N oxygen as source, oxygen plasma was generated by a radio frequency (rf) activated radical cell at the rf power of 300 W. The substrate was annealed at 800 °C for 30 min in the ultra-high-vacuum

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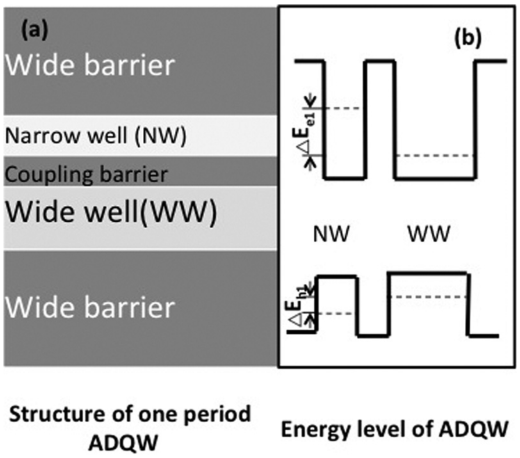

FIG. 1. (a) The schematic diagram of one period of the ADQW; (b) the energy band diagram of the corresponding ADQW in (a).

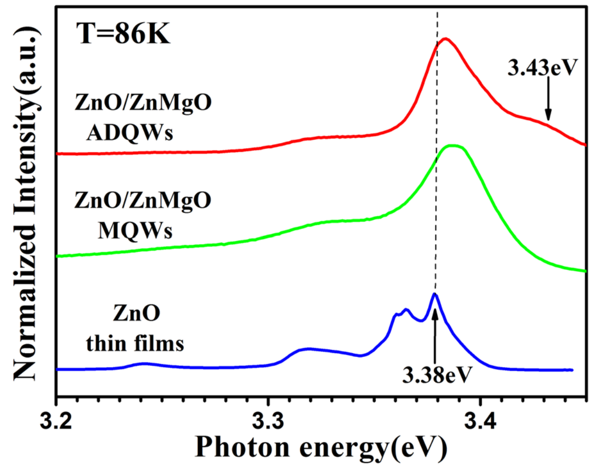

FIG. 2. The PL spectra of the ZnO thin film, the ZnO MQW, and the ADQW samples measured at 86 K.

(UHV) chamber before the growth to remove the surface contamination. The growth rates of ZnO and $Zn_{0.85}Mg_{0.15}O$ were 0.10 nm s^{-1} and 0.08 nm s^{-1}, respectively. Each period of the $ZnO/Zn_{0.85}Mg_{0.15}O$ ADQWs includes one narrow ZnO well, one thin $Zn_{0.85}Mg_{0.15}O$ barrier, and one wide ZnO well (Figure 1(a)), which are denoted 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. Totally five periods of ADQW structures were grown. Single layer ZnO film and ZnO/ZnMgO MQWs were also fabricated on the c-plane Al_2O_3 substrate using PAMBE for the purpose of comparison. Each of the ZnO/ZnMgO MQW periods consists of a ZnO layer and a ZnMgO barrier layer with their widths both equal to 7 nm, and a total of 10 periods were grown. The 325 nm line of a 15 mW He-Cd was used as the excitation source of the PL measurements. Energy dispersive spectroscopy (EDS) was used to determine the Mg contents in the ZnMgO barrier. The room temperature stimulated emission experiment was performed using a quadruplet Q-switched Nd:YAG laser (at 266 nm) at pulsed operation (6 ns, 10 Hz). The laser power was calibrated using the Newport 818-UV. A circular pump spot of diameter 8 mm was collimated onto the surface of the samples. The laser emission was collected from the edge of the sample through an objective lens and analyzed by a monochromator. A tunable Ti:sapphire femtosecond-pulsed laser at the wavelength of 266 nm was used as the excitation source of the time-resloved photoluminescence (TRPL) measurement. The incident light intensity was 100 mW/cm^2. A Hamamatsu C5680-04 streak camera was used for TRPL measurement.

X-ray diffraction (XRD) measurements (Ω-2θ scan) on the single layer film sample shows only the (0002) peak at 34.42° having a narrow full width half maximum (fwhm) of 0.05°. The reflection high energy electron diffraction (RHEED) study carried out during the growth shows streaky pattern of the RHEED image,[12] implying the growth is of two dimensional. These results reveal the very high crystalline quality of the film grown.

The PL spectra of the ZnO thin film, the ZnO/ZnMgO MQW, and the $ZnO/Zn_{0.85}Mg_{0.15}O$ ADQW with $L_N/L_b/L_W = 2$ nm/2nm/7nm structures measured at 86 K are shown in Figure 2. The 3.38 eV emission peak of the ZnO film is attributed to the free-exciton (FX) emission. Similar peaks were also found in the MQW and the ADQW samples but with a small blue shift associated to the quantum confinement effect in the MQW and the WW of the ADQW (both with widths of 7 nm). A 3.43 eV peak is only observed in the ADQW sample and is attributed to the blued shifted FX emission originated from the NW ($L_N = 2$ nm) of the ADQW.

Figure 3 shows the room temperature emission spectra with different pumping intensities for the ZnO thin film, ZnO MQW, and ZnO ADQW samples, while the integral emission intensity against the excitation power are shown in Figure 4. Emission peak is at ~400 nm for the film sample as shown in Figure 3. Typical lasing actions of a series of sharp lasing modes with widths less than 0.1 nm on the broad spontaneous emission band and an abrupt increase of the integrated emission intensity at the excitation power of 51 kW cm^{-2} were clearly shown in Figures 3(a) and 4(a), respectively. Consider no artificial optical oscillator cavity and the lasing behavior as observed in the ZnO film, the mechanism of lasing action can be explained by the coherent random lasing[13,14] and the lasing threshold is also close to the previously reported values.[15,16] Lasing actions of the ZnO/ZnMgO MQW and ADQW structures were also studied at room temperature. As shown in Figures 3(b) and 3(c), serious sharp lasing emissions with widths <0.1 nm at around 393 nm and 385 nm, respectively, for the MQW and ADQW samples are clearly observed. Plotting the integral emission intensity against the excitation energy (Figures 4(b) and 4(c)), obvious kinks at the lasing thresholds of the ZnO MQW and the ZnO ADQW at about 20 kW cm^{-2} and 6 kW cm^{-2}, respectively, are found. The adoption of the ADQW structure has significantly reduced the threshold as compared to that of the MQW sample.

TRPL measurements at 20 K were performed on the spontaneous exciton emission peaks of the film (3.37 eV), MQW (3.39 eV), and the ADQW (3.39 eV) samples. The TRPL spectrum of the ZnO film (figure not shown) was well fitted by a single exponent having the lifetime of 90 ps.[17] For the 3.385 eV spontaneous emission peaks of the ZnO MQW and ZnO ADQW samples, the TRPL spectra were well fitted by the biexponential function, which yielded lifetimes of

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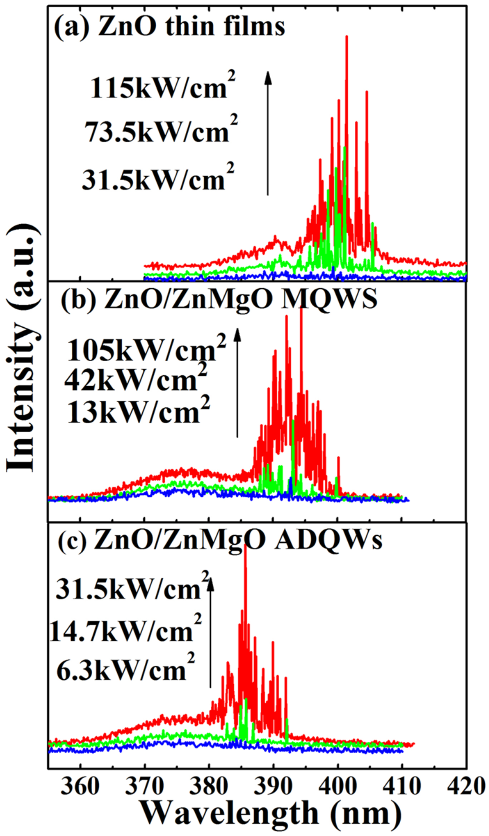

FIG. 3. Room temperature lasing spectra of the (a) ZnO thin film, (b) ZnO/MgZnO MQW, and (c) ADQW samples under the excitation of the 266 nm optical pumping with different intensity.

$\tau_1 = 238$ ps and $\tau_2 = 544$ ps for the MQW sample, and $\tau_1 = 245$ ps and $\tau_2 = 634$ ps for the ADQW sample. The lifetimes as found in the QW samples are larger than that of the ZnO film, which is due to the stability enhanced by the exciton confinement of the QW.

The optical gains of the samples were also studied by the variable stripe-length (VSL) measurements and the measured effective optical gain g_{eff} as a function of the pump density were shown in Figure 5. The solid lines were fitted according to the equation: $g_{eff} = g_0 \ln(P/P_{tr})$, where g_0 is the gain coefficient, P and P_{tr} are the pump density and the transparency pump density, respectively. The results in Figure 5 clearly demonstrated that the MgZnO ADQW's effective optical gain was enhanced as compared to that of the MQW. The measured optical gain given in Figure 5 is in fact the effective optical gain, g_{eff}, of the epi-wafer. g_{eff} is defined as $g_{\mathrm{eff}} = \Gamma \times g$, where g is the optical gain of the optical material (i.e., active region) and Γ is the confinement factor given by

$$\Gamma = \frac{\int_{\text{gain region}} |E(z)|^2}{\int_{\text{all}} |E(z)|^2}, \tag{1}$$

where E is the transverse field. With E calculated by the simple effective index method[18] and the corresponding heterojunctions geometries, the confinements of the ADQW and MQW were roughly equal to $\Gamma_{ADQW} = 0.12$ and $\Gamma_{MQW} = 0.15$. From the data in Figure 5, the optical gain of the ADQW is about 2.5 times larger than that of MQW at the pump density of 20 kW cm^{-2}.

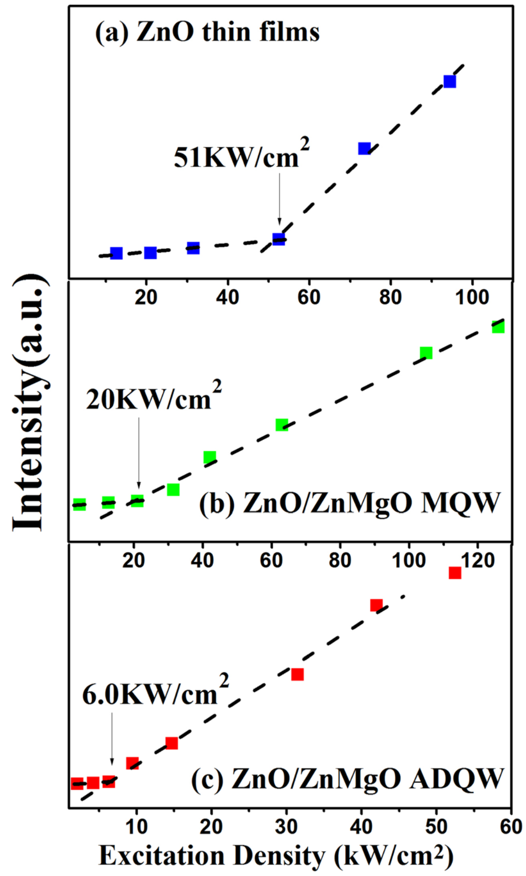

FIG. 4. The integrated intensities of the emission peaks from (a) the ZnO thin film; (b) the ZnO MQW; and (c) the ADQW samples as a function of the excitation density. The measurements were taken at the room temperature.

The low threshold 20 kW cm^{-2} of the MQW sample is associated with the very good crystalline quality of the sample. The lasing threshold of the ADQW sample (6 kW cm^{-2}) is even lower than that of the MQW sample (20 kW cm^{-2}), and it is worthy to have a discussion hereby. The energy of the n-th conduction band E_n in a QW with length of L is given by: $Lh^{-1}(2m^* E_n)^{1/2} = \arctan\ [(V_0 - E_n/E_n)^{1/2}] + n\pi$, where m^* is the effective mass, and V_0 is the depth of the

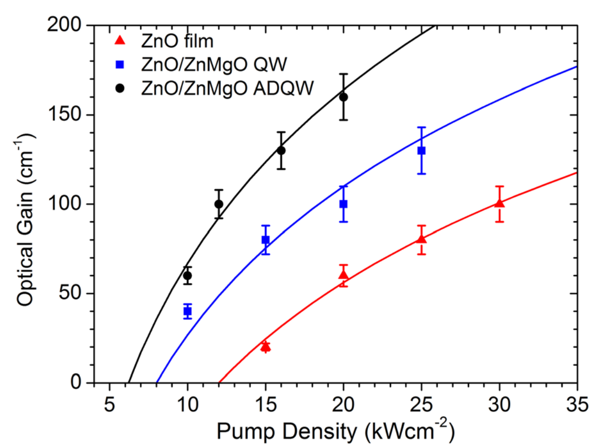

FIG. 5. The optical gains against the pump density for the ZnO thin film, the MQW, and the ADQW structures.

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QW. For the present case, V_0 is taken to be the conduction band offset of $ZnO/Zn_{0.85}Mg_{0.15}O$, i.e., $\Delta E_C = 180$ meV.[19] The effective mass of electron for ZnO has been reported to have values $m^* = 0.24\ m_0$.[20,21] The lowest electron subbands of the NW and the WW were obtained by solving the equation with $n = 1$, $L_N = 2$ nm, and $L_W = 7$ nm, and their separation was found to be $\Delta E_{e1} = 80$ meV, which is larger than and close to the LO phonon energy of ZnO ($\hbar\omega_{LO} \sim 70$ meV). This implies that the rapid electron tunneling from the NW to the WW through the LO phonon assisted tunneling would be feasible. Although such electron tunneling rate for the present structure is not known, that for the ZnCdSe/ZnSe ADQW (with $\Delta E_e = 47$ meV and $\hbar\omega_{LO} = 31.7$ meV) was reported to be as fast as $\sim$300 fs.[9] While taking the valence band offset of $ZnO/Zn_{0.85}Mg_{0.15}O$ and the effective hole mass to be $\Delta E_V = 120$ meV (Ref. 19) and 0.59 m_0,[22] the separation between the first hole subbands in the NW and the WW was calculated according to Eq. (1) and was equal to $\Delta E_{h1} = 40$ meV, which was smaller than the ZnO phonon energy. Single particle hole tunneling from the NW to the WW is thus very slow due to its heavy effective mass. However, because of the strong Columbic electron-hole entanglement in ZnO (and also ZnSe), the rate of hole tunneling through LO phonon emission would be enhanced if the electron is at the WW (i.e., $(W_e, N_{HH}) \rightarrow (W_e, W_{HH})$) and for the case of ZnSe having value of $\sim$1 ps. As the exciton binding energy of ZnO is even larger than that of ZnSe, it is plausible to say that the corresponding hole tunneling rate in the present ZnO/ZnMgO ADQW would not be less than that of ZnSe. While the measured carriers lifetimes in the all the QW structures are significantly longer than the lifetimes of tunneling, effectively majority of the excited carriers would be accumulated at the WW. The higher optical gain for the ADQW sample is also shown in Figure 5. In this case, the population density in the upper level will be larger than those of the ZnO film and MgZnO MQW, so the population inversion can be realized with a lower pump density.

In conclusion, optical pump lasing threshold as low as 6 kW cm^{-2} has been achieved in the ZnO/ZnMgO ADQW structure, which is lower than that of the MQW grown with the similar condition. The optical gain of the ADQW structure is enhanced relative to the MQW structure. The long decay time was realized in ADQWs. The low threshold is attributed to the enhanced free carriers' densities at the WW accumulated by the tunneling of electron and holes from the NW to the WW. The present result shows that ADQW could be a promising structure for the realization of low-threshold exciton-based laser device.

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