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The optical properties of ZnO/ZnMgO single quantum well grown by P-MBE

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ABSTRACT

In this paper, ZnO/$Zn_{0.9}Mg_{0.1}O$ single quantum well (SQW) structures were fabricated on *c*-plane sapphire (Al_2O_3) substrate by plasma-assisted molecular beam epitaxy (P-MBE). The photoluminescence (PL) peak of the SQW shifted from 3.31 to 3.37 eV as the well layer thickness was decreased from 6 to 2 nm. The spectral linewidth increases with temperature due to the scattering of excitons with acoustic and optical phonons. The transition energy of the localized exciton in the ZnO/$Mg_{0.1}Zn_{0.9}O$ SQW with well width of 3 nm was found to be about 3.407 eV at 80 K, consistent with theoretical calculation. The first subband energies in the conduction and valence band were calculated to be 49 and 11 meV, respectively.

1. Introduction

ZnO is one of the most attractive materials for application to optical devices operating in blue and ultraviolet region due to its direct wide band gap (3.37 eV at room temperature) and large exciton binding energy (60 meV) [1,2]. In order to construct optical and electrical confinement structures, a barrier material is needed to combine with ZnO. ZnMgO is one of the most accessed materials because its lattice constant is very close to that of ZnO. Although the crystal structures of ZnO and MgO are different (ZnO: wurtzite structure, MgO: rocksalt structure), hexagonal $Zn_{1-x}Mg_xO$ films have been reported with *x* values up to 0.5 [3]. Much effort has been made to characterize these structures. Makino et al. [4] reported the temperature dependence of time-integrated and time-resolved photoluminescence (PL) spectra of $Zn_{0.88}Mg_{0.12}O$/ZnO and $Zn_{0.73}Mg_{0.27}O$/ZnO multi-quantum wells (MQWs) grown by laser molecular beam epitaxy (MBE). Recently, ZnO/MgZnO single quantum wells (SQWs) have been grown by metal–organic chemical vapor deposition (MOCVD) on GaN templates and they observed carrier confinement [5]. The ZnO/MgZnO SQWs also have been grown by the plasma-assisted molecular beam epitaxy (P-MBE) on Al_2O_3 [6–8] and on Si(1 1 1) substrates [9].The optical properties of the ZnMgO MQWs have been discussed [4,10]. But the detailed works on the origin of PL in ZnO/ZnMgO single quantum wells are very scarce.

In this work, ZnO/$Zn_{0.9}Mg_{0.1}O$ single quantum wells with different well widths (L_W) were fabricated on *c*-plane sapphire (Al_2O_3) substrates by P-MBE. The room temperature (RT) PL of the single quantum well structures was discussed. Efficient excitonic emission from these SQWs can be observed up to room temperature. We have also investigated the temperature dependence of PL peak position and linewidth in a temperature range from 80 K to RT. The first subband energies in the conduction and valance band of the ZnO/ZnMgO SQWs are calculated.

2. Experimental

ZnO and ZnMgO films were grown on Al_2O_3 substrates by P-MBE. Elemental Zn (6*N*) and Mg (5*N*) were evaporated using conventional effusion cells. Pure oxygen (5*N*) was used for oxygen source and oxygen plasma was generated through a radio frequency (rf) activated radical cell. The rf power of 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 of the quantum well was shown in Fig. 1. A high quality ZnO buffer layer about 50 nm is grown at 800 °C. Then,

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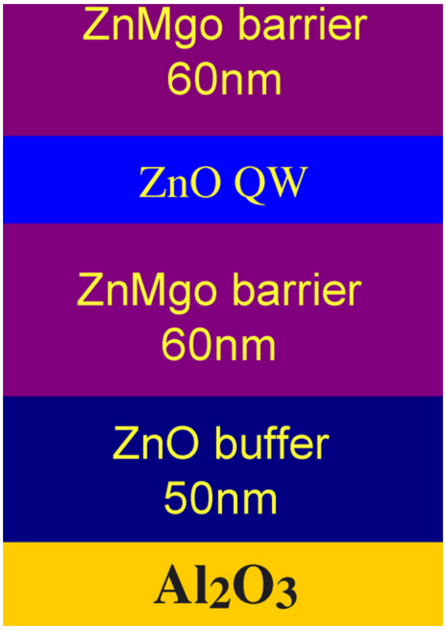

Fig. 1. $ZnO/Zn_{0.9}Mg_{0.1}O$ single quantum well structure grown on Al_2O_3 substrate by using ZnO buffer layer.

ZnMgO/ZnO/ZnMgO single quantum well structure is grown at 600 °C in the growth pressure of 1.0×10^{-6} mbar. A ZnO layer was sandwiched between the buffer layer and a 60-nm thick $Mg_{0.1}Zn_{0.9}O$ cap layer. Here, the thickness of ZnO layer was varied from 2 to 20 nm. The barrier and well layer thicknesses were determined by prescribed deposition time, since the growth rate under this condition was found to be 0.1 nm/s for ZnO and 0.08 nm/s for $Mg_{0.1}Zn_{0.9}O$ in separate experiments.

The Mg concentration in the MgZnO barrier layer was detected by energy dispersive X-ray spectroscopy (EDX). The PL measurements were performed on a JY-630 micro Raman spectrometer at temperatures from 80 to 290 K, and a He–Cd laser operating at 325 nm was used as the excitation.

3. Results and discussion

Fig. 2 shows the PL spectra of the SQWs with different active layer thicknesses from 20 to 2 nm at RT. When the ZnO layer thickness is 20 nm, it is obvious that the samples show two emission peaks: one is a stronger emission from the ZnMgO layer and the other is a weaker one from the ZnO layer. As the ZnO layer thickness decreases, the luminescence from the MgZnO layer becomes weaker and weaker. It can also be seen that as the well layer thickness was decreased from 6 to 2 nm, the PL peak shifted from 3.31 to 3.37 eV. The corresponding emission peaks exhibit a blueshift due to the quantum size effect. The weak broad shoulders at 3.29 and 3.49 eV correspond to the ZnO buffer and the MgZnO barrier layers, respectively.

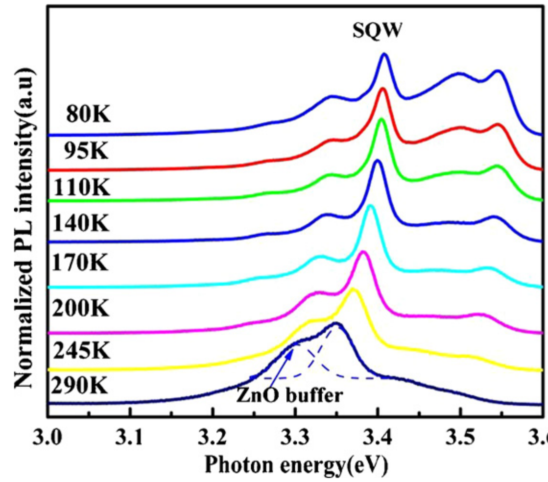

Fig. 3. Temperature-dependent PL spectra of the $ZnO/Zn_{0.9}Mg_{0.1}O$ SQW with well width of 3 nm from 80 to 290 K.

Fig. 3 shows the temperature-dependent PL spectra of the $ZnO/Zn_{0.9}Mg_{0.1}O$ SQW with the well width of 3 nm. As the temperature increases the SQW peak energy shows a redshift from 3.407 to 3.350 eV. It can be seen that the PL peak position shifts towards red and its linewidth increases gradually with increase in temperature up to RT.

The temperature-dependence of PL peak position and linewidth for the SQW with well layer thickness 3 nm are shown in Fig. 4. The linewidth of the PL peak increases monotonically with increasing temperature from 80 to 140 K, and then goes up exponentially at 140 K above. The temperature dependent broadening of the PL peak has been interpreted in terms of exciton–phonon scattering in different temperature regimes using the following theoretical

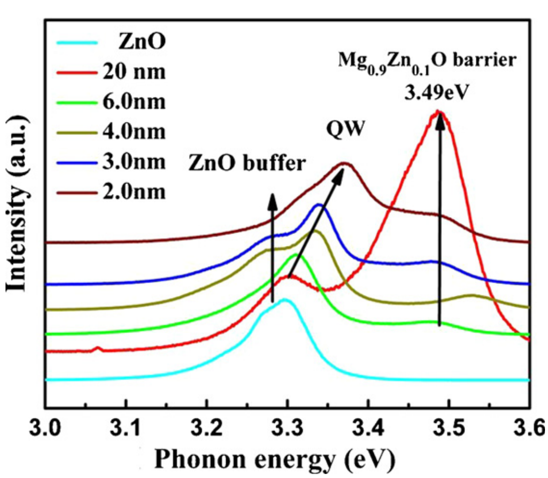

Fig. 2. Room temperature PL spectra of $ZnO/Zn_{0.9}Mg_{0.1}O$ SQW with different well width.

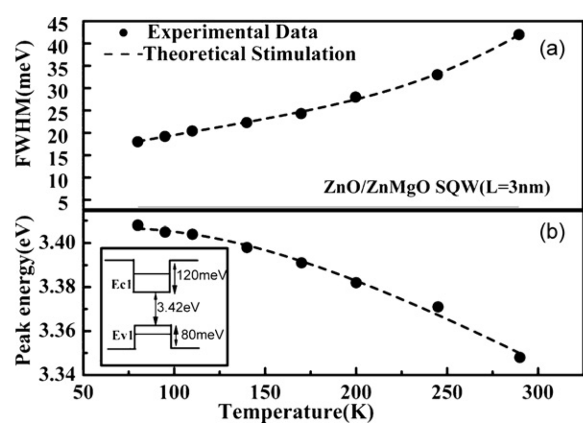

Fig. 4. Variation of the linewidth (a) and PL peak position (b) with temperature for $ZnO/Zn_{0.9}Mg_{0.1}O$ SQW with well width of 3 nm. Dashed lines show the fits to the experimental data. The insert shows the energy band diagram of $ZnO/Mg_{0.1}Zn_{0.9}O$ SQW structure for the well width of 3 nm.

formulation developed by Hellmann et al. [11] and O'Neill et al. [12]:

$$\Gamma(T) = \Gamma\text{inh} + \gamma\text{ph}T + \Gamma\text{LO}/[\exp(h\omega\text{LO}/KT) - 1] \quad (1)$$

where $h\omega$LO is the LO phonon energy, Γ_{inh} is the inhomogeneous linewidth at 0 K, γ_{ph} is the coupling strength of the exciton–acoustic phonon interaction [11], Γ_{LO} is the parameter related to the strength of the exciton–LO phonon coupling [12] and $[\exp(h\omega\text{LO}/KT)-1]^{-1}$ is the population of LO phonons of energy $h\omega$LO at temperature T. The dash line in Fig. 4(a) shows the fitting result based on Eq. (1). The best fit yields $\Gamma_{\text{inh}} = 17$ meV, $\Gamma_{\text{LO}} = 281$ meV, LO = 72 meV and $\gamma_{\text{ph}} = 29$ μeV/K, which are very close to the values obtained by Makino and coworkers [13]. According to Hellmann et al., the gradual increase of the linewidth in the low temperature regime can be attributed to the exciton scattering with acoustic phonons while the exponential rise at higher temperature regime to the scattering with LO phonons [13]. The fact that the temperature dependence of the linewidth in our case can be explained using Eq. (1) indicates that all the PL transitions from 80 to 290 K were excitonic in nature [11,12].

As can be seen from Fig. 4(b) the PL peak position has shifted gradually towards low-energy side by an amount of 60 meV with increasing temperature from 80 to 290 K. The variation of the band gap of ZnO SQWs with temperature was studied using the following formula [14]:

$$E(T) = E(0) - \alpha T^2/(\beta + T) \quad (2)$$

where α, β, are the fitting parameters and $E(0)$ is the band gap at 0 K. The dashed lines in Fig. 4(b) are the fitting result to the experimental data using Varshni's relation. The best fitting to the data was obtained for $E(0) = 3.42$ eV, $\alpha = 2.3 \times 10^{-4}$ eV/K and $\beta = 782$ K. The functional forms of the temperature dependence of the band gaps of the 3 nm SQW was found to be nearly the same as that of the ZnO film grown on sapphire [15], indicating the nearly strain free and high quality of the SQW.

The insert in Fig. 4(b) shows a schematics band diagram for the wells and barriers in ZnO/$Mg_{0.1}Zn_{0.9}O$ SQW ($L_w = 3$ nm). The electron/hole effective masses are 0.28 m_0 and 1.8 m_0 (m_0 is the free electron mass) [16], respectively. Here we assume that the electron/hole effective mass is the same for ZnO well layer and $Mg_{0.1}Zn_{0.9}O$ barrier layer. The band discontinuity $E_c/E_v = 6/4$ determined by Coli and Bajaj [17], and applying the Kronig-Penney model the first subband energies in the conduction and valance band are calculated to be 49 and 11 meV, respectively. The transition energy of the localized exciton in a SQW structure is given by [18],

$$E_{n,m} = E_g + E_n + E_m - E^{\text{B}}_{n,m} \quad (3)$$

where E_g is the band gap of ZnO, E_n is the energy of the nth subband in conduction band, E_m is the energy of mth subband in valence band. $E^{\text{B}}_{n,m}$ is the binding energy of the exciton localized in the nth and the mth subbands in the SQWs. Assuming that the E_g at low temperature is 3.420 eV and $E^{\text{B}}_{1,1}$ in our samples is 75 meV similar to that in the ZnO/MgZnO MQWs grown on $ScAlMgO_4$ substrates [19], $E_{1,1}$for ZnO/$Mg_{0.1}Zn_{0.9}O$ SQW (3 nm) is estimated to be 3.405 eV, which is in good agreement with our experimental results. This indicated that the ZnO/$Mg_{0.1}$ $Zn_{0.9}O$ single quantum well was high quality.

4. Conclusion

ZnO/ZnMgO single quantum wells were grown on c-plane sapphire substrates by P-MBE. The PL peak shifted toward high-energy side with decreasing well layer thickness and at constant well width the PL peak shifted towards low-energy side with increasing temperature. The redshift of the PL peak with increasing temperature was found to be due to the band gap shrinkage in accordance with the Varshni's empirical relation. The width of the PL peak increases with temperature due to the scattering of excitons with acoustic and optical phonons in different temperature regimes.

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