Optical Properties of a ZnO-MgZnO Quantum-Well

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Abstract—**The optical gain and the luminescence of a ZnO quantum well with MgZnO barriers is studied theoretically. We calculated the non-Markovian optical gain and the luminescence for the strained-layer wurtzite quantum well taking into account of the excitonic effects. It is predicted that both optical gain and luminescence are enhanced for the ZnO quantum well when compared with those of InGaN-AlGaN quantum well structure due to the significant reduction of the piezoelectric effects in the ZnO-MgZnO systems.**

Index Terms—**ZnO-MgZnO quantum well, optical gain, luminescence, non-Markovian**

ZnO and related wide bandgap wurtzite oxides are drawing considerable attention recently for their potential applications to optoelectronic devices such as high power light emitting diodes (LEDs) and laser diodes [1~5]. This oxide system may have an advantage over conventional III-V nitride systems for which the built-in electric fields caused by strain induced piezoelectric effects and spontaneous polarization [6] are known to reduce the optical gain or the luminescence quite significantly [7-8]. The piezoelectric field for ZnO on MgZnO are expected to be much smaller than that of the III-V nitride systems [2]. From physics point of view, there is another interesting feature of ZnO-MgZnO structure. It is found that due to a large exciton biding energy, excitonic effects on the optical transitions are quite pronounced even at temperatures well above the room temperature [1-2]. So far, there has been very little theoretical work on optical gain and luminescence of these II-VI wurtzite oxide systems. It seems that any reasonable theoretical model should include the strain effects as well as the many-body phenomena including the excitonic effects.

In this paper, we study the optical gain and the luminescence of a ZnO quantum well with MgZnO barriers theoretically. Non-Markovian model of the optical gain and the luminescence for the strained-layer wurtzite quantum well is employed taking into account of many-body effects within the time-dependent Hartree-Fock approximation [10-11]. Plasma screening, bandgap renormalization, and excitonic (or Coulomb) enhancement of optical transitions are included in the model. Non-Markovian lineshape is related to the memory effects in the system-reservoir interaction. The band structure of wurtzite quantum well is calculated within the 6x6 multiband effective mass theory [11-13] which also takes into account the biaxial strain, spontaneous polarization, and the piezoelectric effects.

We consider a quantum well of width L_z grown along the z-axis with biaxial strain. Biaxial strain is assumed to be adjustable by changing alloy compositions of both quantum well and barrier. The growth directions are along the (0001) (z-axis) for the wurtzite quantum well. In order to calculate the wave functions and the energy band structure, we used the multiband effective mass theory [11-13]. The self-consistent screening potential is obtained from the Poisson equation and the spontaneous polarization and the piezoelectric potential are included in the model. The self-consistent band structures and wave functions are obtained by solving the Schrodinger equation for electrons, the block diagonalized 3x3 Hamiltonian for holes, and Poisson equation iteratively until the solutions converge [7, 8].

The non-Markovian optical gain with many-body effect is given by [9, 10]

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$$g(\omega)=\frac{\omega\mu c}{n_r V}\sum_{\sigma\eta lm}\sum_{\vec{k}_{\|}}|\hat{\varepsilon}\cdot M_{lm}^{\eta\sigma}(\vec{k}_{\|})|^2\left(f_c^l-f_{h\sigma}^m\right)C_{lm}^{\eta\sigma}(\vec{k}_{\|}), \quad (1)$$

where ω is the angular frequency; μ is the permeability; n_r is the refractive index; c is the speed of light in free space; V is the volume; f_c^l and $f_{h\sigma}^m$ are the Fermi functions for the l th subband in the conduction band and the m the subband in the valence band of H^σ, respectively; $M_{lm}^{\eta\sigma}(\vec{k}_{\|})$ is the dipole matrix element between the l th conduction subband with a spin state η and the m th valence subband of the 3x3 block Hamiltonian H^σ ; and $\hat{\varepsilon}$ is the unit vector in the direction of the photon polarization.

The renormalized lineshape function $C_{lm}^{\eta\sigma}(\vec{k}_{\|})$ is obtained by [10]

$$\begin{aligned}C_{lm}^{\eta\sigma}(\vec{k}_{\|})=&\frac{1}{\left(1-\mathrm{Re}\,q_{lm}^{\eta\sigma}(\vec{k}_{\|})\right)^2+\left(\mathrm{Im}\,q_{lm}^{\eta\sigma}(\vec{k}_{\|})\right)^2}\\&\times\left\{\mathrm{Re}\,\Xi_{lm}^{\eta\sigma}(0,\Delta_{lm}^{\eta\sigma}(\vec{k}_{\|}))\left(1-\mathrm{Re}\,q_{lm}^{\eta\sigma}(\vec{k}_{\|})\right)\right.\\&\left.-\mathrm{Im}\,\Xi_{lm}^{\eta\sigma}(0,\Delta_{lm}^{\eta\sigma}(\vec{k}_{\|}))\,\mathrm{Im}\,q_{lm}^{\eta\sigma}(\vec{k}_{\|})\right\}\end{aligned} \quad (2)$$

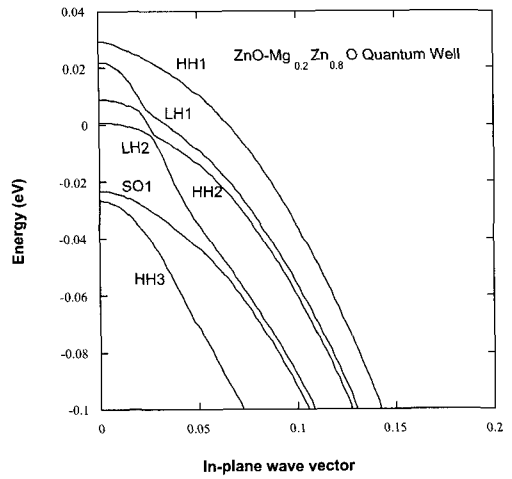
where $\mathrm{Re}\,q_{lm}^{\eta\sigma}(\vec{k}_{\|})$ and $\mathrm{Im}\,q_{lm}^{\eta\sigma}(\vec{k}_{\|})$ are the real and the imaginary part of the Coulomb interaction between the electron in the l th conduction subband with a spin state η and the hole in the m th valence subband of the 3x3 block Hamiltonian H^σ in the presence of photon fields, respectively; $\mathrm{Re}\,\Xi_{lm}^{\eta\sigma}(0,\Delta_{lm}^{\eta\sigma}(\vec{k}_{\|}))$ and $\mathrm{Im}\,\Xi_{lm}^{\eta\sigma}(0,\Delta_{lm}^{\eta\sigma}(\vec{k}_{\|}))$ are the real and the imaginary part of the non-Markovian lineshape; $\Delta_{lm}^{\eta\sigma}(\vec{k}_{\|})=E_l^c(\vec{k}_{\|})-E_m^{h\sigma}(\vec{k}_{\|})+E_G+\Delta E_{SX}+\Delta E_{CH}-\hbar\omega$; $E_l^c(\vec{k}_{\|})$ and $E_m^{h\sigma}(\vec{k}_{\|})$ are the l th subband energy in the conduction band and the m the subband energy in the valence band of H^σ at $\vec{k}_{\|}$, respectively; E_G is the bandgap energy; ΔE_{SX} is the screened exchange energy change; ΔE_{CH} is the Coulomb-hole contribution to the bandgap renormalization; and $\hbar$ is the Plank constant. Especially, $\mathrm{Re}\,q_{lm}^{\eta\sigma}(\vec{k}_{\|})$ describes an excitonic enhancement of optical transitions. Non-Markovian optical gain with many-body effects is derived from the time-convolutionless quantum kinetic equations for the system of interacting electron-hole pairs under an external optical field using the projection-operator formalism [9]. In this model, the exact calculation of the Coulomb vertex correction to the optical susceptibility within the two band model and the consistent merging of non-Markovian relaxation with Coulomb effects are carried out [10].

The spontaneous emission rate $R_{sp}(\omega)$, the number of emitted photons per second per unit volume per unit energy interval, is related to the optical gain $g(\omega)$ by [14-16]

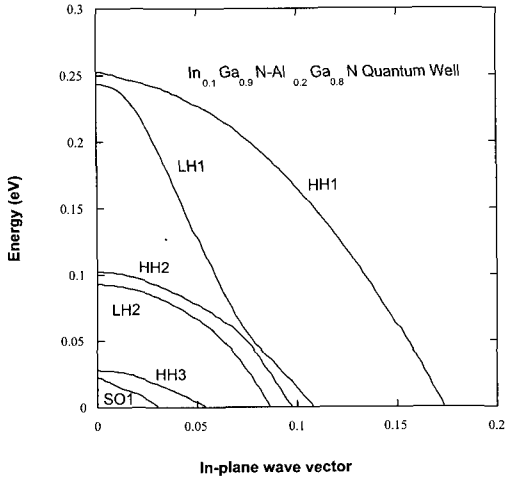
$$g(\omega)=\hbar\left(\frac{\pi c}{n_r\omega}\right)^2\left\{1-\exp\left(\frac{\hbar\omega-\Delta\mu}{k_BT}\right)\right\}R_{sp}(\omega), \quad (3)$$

where $\Delta\mu=E_G+\Delta E_{SX}+\Delta E_{CH}+\mu_n-\mu_p$; μ_n and μ_p are renormalized chemical potentials for the electron and the hole, respectively, such that $g(\Delta\mu)=0$; k_B is the Boltzmann constant; and T is the temperature. We note that at $\hbar\omega=\Delta\mu$, the definition of $R_{sp}(\omega)$breaks down, so we interpolated $R_{sp}(\omega)$ at $\omega_{trans}=\Delta\mu/\hbar$ from the values of $R_{sp}(\omega)$ at $\omega_{trans}\pm\delta$. The importance of using the non-Markovian lineshape functions would be pronounced in the above relation between the spontaneous emission rate and the optical gain. One of the remarkable features of this relation is that there is a transparency point in the gain spectra which coincide with the chemical potential separation that suggests the carriers and the photons are in equilibrium or in quasi-equilibrium. The optical gain spectra calculated with the Lorentzian line shape function have two errors: unnatural absorption region below the renormalized bandgap energy and mismatch of the transparency point of the gain with the chemical potential separation. It was shown [17] that these two anomalies associated with the Lorentzian lineshape are removed in the non-Markovian model with many-body effects.

As a numerical example, we considered 4 nm ZnO – $Mg_xZn_{1-x}O$ quantum well with the barrier width of 7 nm. In Fig. 1, we plot the valence band structures of (a) ZnO –$Mg_{0.2}Zn_{0.8}O$ quantum well and (b) $In_{0.1}Ga_{0.9}N$-$Al_{0.2}Ga_{0.8}N$ quantum well, respectively. The valence band structures are obtained from the self-consistent solution [18] of the 6x6 Hamiltonian, which also takes into account the biaxial strain, spontaneous polarization, and the piezoelectric effects for the carrier density of



(a)



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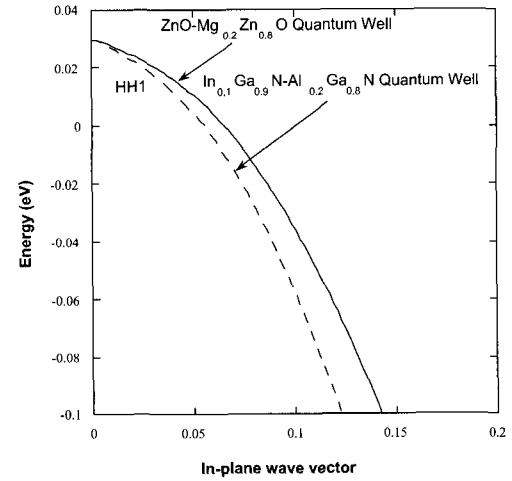

(c)

Fig. 1. (a) Valence band structure of a 4 nm ZnO-$Mg_{0.2}Zn_{0.8}O$ quantum well (b) Valence band structure of a 4 nm $In_{0.1}Ga_{0.9}N$- $Al_{0.2}Ga_{0.8}N$ quantum well (c) HH1 hole energy dispersions are compared for a 4 nm ZnO-$Mg_{0.2}Zn_{0.8}O$ quantum well and a 4 nm $In_{0.1}Ga_{0.9}N$- $Al_{0.2}Ga_{0.8}N$ quantum well.

$3x10^{19}$ cm^{-3}. Physical parameters for the $k \bullet p$ model are taken from references 19 and 20. It is interesting to note that the energy levels are more closely spaced in the case of the ZnO quantum well than the InGaN system, which would imply that the zone center effective mass of the heavy hall for the former is heavier than the latter. This speculation can be confirmed in Fig. 1(c) where we have plotted the HH1 states for both ZnO (solid line) and InGaN (dashed line) quantum wells. We have moved the position of HH1 of InGaN quantum well to make the comparison easy.

From eq. (1), one can expect that the optical gain is roughly proportional to the dipole moment $M_{lm}^{\eta\sigma}(\bar{k}_{\|})$, which is defined by [9]

$$M_{lm}^{\eta\sigma}(\bar{k}_{\|}) = \begin{cases} \sum_{\nu=1,2,3} \langle g_m^{(\nu)} \mid \phi_l \rangle \langle \nu \mid \hat{e} \cdot e\bar{r} \mid S, \eta \rangle \text{ for } \sigma = U \\ \sum_{\nu=4,5,6} \langle g_m^{(\nu)} \mid \phi_l \rangle \langle \nu \mid \hat{e} \cdot e\bar{r} \mid S, \eta \rangle \text{ for } \sigma = L \end{cases}, \quad (4)$$

where $g_m^{(\nu)}(\bar{k}_{\|}, z)$ is the hole envelope function; $\{\mid \nu \rangle\}$ denotes the transformed Bloch basis at the zone center; m is the quantum well subband index; $\bar{k}_{\|} = k_x \hat{x} + k_y \hat{y}, \bar{r}_{\|} = x\hat{x} + y\hat{y}$, and $\sigma = U$ (or L) refers to the upper (or lower) blocks; respectively; $\phi_l(z)$ is the electron envelope function for the l th conduction subband with a spin state η; and $\hat{e}$ is the unit vector in the direction of the photon polarization. In the case of $In_{0.1}Ga_{0.9}N$-$Al_{0.2}Ga_{0.8}N$ quantum well, the unscreened piezo electric field is 2.7 MV/cm while the internal field of ZnO quantum well is negligible. As a result, the calculated square of the dipole moment for the $In_{0.1}Ga_{0.9}N$-$Al_{0.2}Ga_{0.8}N$ is $2.81x10^{-22}$ m^2, while that of ZnO –$Mg_{0.2}Zn_{0.8}O$ quantum well is $1.19x10^{-20}$ m^2 at the zone center thus making the latter about 50 times larger than the former.

In Fig. 2, non-Markovian optical gain spectra with many-body effects are plotted for a 4 nm ZnO –$Mg_xZn_{1-x}O$ quantum well for (i) x=0.1; (ii) x=0.2; (iii) x=0.3; and (iv) an $In_{0.1}Ga_{0.9}N$-$Al_{0.2}Ga_{0.8}N$ quantum well, versus photon energy for carrier densities of 3 x 10^{19} cm^{-3}. The optical gain is expected to increase with increasing confinement potential heights. Confinement potential enhancement can be achieved by adding magnesium to the barrier. In this calculation, we have used $\tau_c = 10\, fs$

and the dephasing time of 20 *fs* as input parameters for the ZnO quantum well and $\tau_c = 10\,fs$ and the dephasing time of 25 *fs* for the InGaN system [21-22]. As expected, the optical gain of ZnO-MgZnO quantum well is larger than that of InGaN-AlGaN quantum well due to the reduction of the piezoelectric field effects in the ZnO systems.

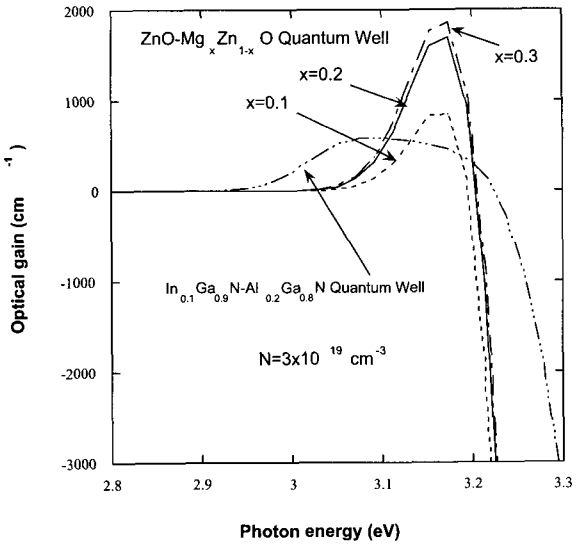

Fig. 2. Optical gain of a 4 nm ZnO-$Mg_xZn_{1-x}O$ quantum well for (i) x=0.1; (ii) x=0.2; (iii) x=0.3 and 4 nm $In_{0.1}Ga_{0.9}N$-$Al_{0.2}Ga_{0.8}N$ versus photon energy for carrier densities of 3 x 10^{19} cm^{-3} at room temperature.

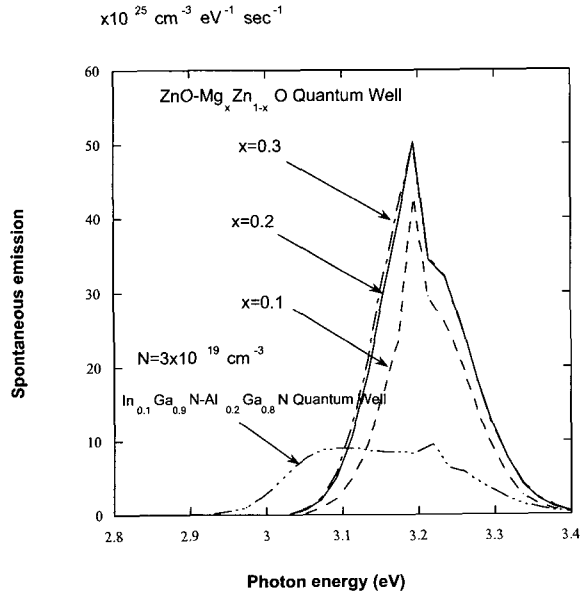

Fig. 3. Spontaneous emission spectra of a 4 nm ZnO-$Mg_xZn_{1-x}O$ quantum well for (i) x=0.1; (ii) x=0.2; (iii) x=0.3 and 4 nm $In_{0.1}Ga_{0.9}N$- $Al_{0.2}Ga_{0.8}N$ versus photon energy for carrier densities of 3 x 10^{19} cm^{-3} at room temperature.

In Fig. 3, we show the luminescence spectra which are the spontaneous emission rates calculated from the non-Markovian optical gain with many-body effects for for a 4 nm ZnO –$Mg_xZn_{1-x}O$ quantum well for (i) x=0.1; (ii) x=0.2; (iii) x=0.3; and (iv) a $In_{0.1}Ga_{0.9}N$-$Al_{0.2}Ga_{0.8}N$ quantum well, versus photon energy for carrier densities of 3 x 10^{19} cm^{-3}. We would like to note that the peak luminescence of ZnO quantum well is expected to be quite larger than that of the InGaN quantum well.

In summary, we studied the optical gain and the luminescence of a ZnO quantum well with ternary MgZnO barriers theoretically. Non-Markovian model of the optical gain and the luminescence for the strained-layer wurtzite quantum well is employed taking into account of many-body effects within the time-dependent Hartree-Fock approximation. The optical gain and the luminescence are expected to be enhanced significantly when compare with those of InGaN-AlGaN quantum well due to the reduction of the internal field caused by spontaneous polarization and piezoelectric effects for the ZnO-MgZnO systems.

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