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Magneto-optical study of *n*-type modulation-doped ZnO/$Mg_xZn_{1-x}O$ single quantum well structures

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We report on the effect of electron injection on the magneto-optical properties of MgZnO/ZnO(5 nm)/MgZnO single quantum wells (SQWs) by embedding a Ga delta-doped layer in one of the MgZnO barriers. Approaching the delta-doped layer to the SQW resulted in a photoluminescence (PL) blueshift due to the screening of internal polarization field by the injected electrons. Circular polarization in PL was enhanced by a magnetic field up to 50 T with a Faraday configuration. The observed Zeeman splitting and the degree of circular polarization in the magnetic field revealed that the PL originates from the negatively charged excitons X^-.

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I. INTRODUCTION

Recent studies on the quantum wells (QWs) based on polar hexagonal semiconductors [e.g., group III nitrides[1] and ZnO (Refs. 2–4)] have revealed that the polarization gives a significant influence on the optical properties of excitons. Due to the spatial separation of the electron and hole wave functions under a polarization field, the oscillator strength of excitons becomes smaller than under field-free conditions.[5,6] The electric field within the well layer also gives rise to the quantum-confined Stark effect on the luminescence, which results in redshifts of the fundamental optical transitions.

Doping has been proven to be an efficient way to reduce the effects of the internal electric field on the optical properties in III-nitride QWs. Enhanced optical emission efficiency has been observed in these modulation-doped QWs, in which flat band conditions are established.[5–10] It is important to study on the interplay between the quantum-confined Stark and modulation-doping effects because it provides insight into the electronic structures of the quantized states in the QWs.

The electron injection from the barrier layers in unstrained zinc-blende II-VI or III-V QWs is known to result in an observation of optical transitions where the background electrons are involved, such as charged excitons.[11,12] Modulation-doped III-nitride QWs have recently been produced and their physical properties have been studied in a number of experimental papers.[7–10,13–15] To date, optical properties of modulation-doped ZnO QWs have not been studied.[16] Polarization-sensitive magneto-optical study provides us with powerful information concerning the origin of photoluminescence (PL) because the doping-related transitions are known to be the issues of spin-sensitive photoexcitation.[11,12,17,18]

In this paper, we present the results of magneto-PL and magnetic-circular dichroism (MCD) studies for *n*-type modulation-doped QWs. When the delta-doped layer is located far away from the QW, the PL was quenched by a magnetic field. Approaching the delta-doped layer to the QW gave rise to the detection of circularly polarized photoluminescence and anomalous MCD under a magnetic field, suggesting the presence of charged excitons (X^-).

II. EXPERIMENTAL PROCEDURE

The samples were one-sided modulation-doped $Mg_{0.12}Zn_{0.88}O/ZnO/Mg_{0.12}Zn_{0.88}O$ single quantum wells (SQWs) grown on $ScAlMgO_4$ substrates.[4] The width of the quantum wells (L_w) was 5 nm. Top side of the barriers was delta doped with Ga (5×10^{10} cm^{-2}). The distance between the delta-doped layer and QW (defined as L_s) was 6 or 14 nm. We employed a pulsed-field magnet generating a field of up to ~50 T for PL experiments. A He-Cd laser (325 nm) was used as the excitation light source (≈ 5 mW/cm^2).[4] MCD was measured at a static magnetic field of 6 T. Circular polarization was produced by combination of a linear polarizer and a photoelastic modulator operating at 50 kHz. Using the lock-in technique, the measured signal intensity is proportional to the difference between the left- and right-circular polarized reflectance intensities divided by their sum. The measurement temperatures were 4.2 K for the PL and 12 K for the MCD.

III. RESULTS AND DISCUSSION

Magneto-PL spectra taken at 4.2 K are shown in Fig. 1 for the modulation-doped QWs [$L_s=$ (a) 6 nm and (b) 14 nm] and (c) for a bulk single crystal. The PL peak energy of the $L_s=14$ nm sample is approximately 3.27 eV at $B=0$ T. It is similar to the calculated transition energy of exciton in undoped ZnO/$Mg_{0.16}Zn_{0.84}O$ QW having $L_w=5$ nm,[3] whereas this energy is lower than its bulk excitonic gap.[19] The inter-

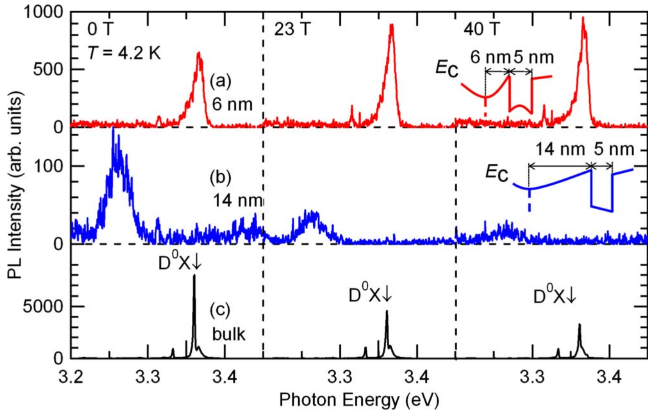

FIG. 1. (Color online) Photoluminescence spectra of modulation-doped ZnO/$Mg_{0.12}Zn_{0.88}O$ QWs (L_w=5 nm) with two different Ga delta-doped layer positions [L_s= (a) 6 and (b) 14 nm] and (c) those of a bulk single crystal. Data were taken at T =4.2 K under magnetic fields of B=0, 23, and 40 T. Also shown are sketches of the conduction band potential profile for respective SQWs.

nal electric field separates in the growth direction the electrons and holes to induce the redshift in PL. The doped layer is located too far from the QW to dope electrons with enough high density for screening the internal electric field. As L_s decreased from 14 to 6 nm, the PL energy at B=0 T shifted to higher energy side (3.366 eV). This energy is similar to that of bulk excitonic gap. Furthermore, the PL intensity was significantly higher for L_s=6 nm QW than that for L_s =14 nm QW. Both the disappearances of Stokes-like shift and PL enhancement by approaching the delta-doped layer suggest that the injected high-density free electrons screen the internal electric field in ZnO SQW. The similar screen-related blueshift has been confirmed by the excitation-intensity dependence of PL in undoped ZnO QWs.[4] Schematic conduction band profiles are given in inset, taking into account the polarization direction with the O-face surface configuration.[20] Sheet electron concentration for similar samples to those used in this study was measured by Hall effect at room temperature. The values for L_s=6 nm samples were in the same order to the nominal Ga concentration. Increasing L_s resulted in a steep decrease in electron concentration and that of modulation-doped QW with L_s=14 nm was lower than its detection limit. The blueshift in PL is consistent with the previous observation in modulation-doped InGaN/GaN quantum wells.[5,7,8]

Increasing the magnetic field applied normal to the heterointerfaces induced a suppression of PL intensity for the case of L_s=14 nm [Fig. 1(b)]. Such PL quenching has been observed so far for spatially indirect excitons in type-II or coupled quantum wells.[21] It is thought that, also in the case of L_s=14 nm, the internal electric field gives rise to the spatial separation of the electron and hole. The observed quenching should be similarly explained for the case of spatially indirect excitons in terms of the magnetic-field-induced coupling of the exciton internal structure and center-of-mass motion.[21,22] It is noted that reduced Coulomb energy can be regarded as a perturbation to the cyclotron energy. Application of magnetic field induces a transition from a hydrogen-like exciton valid in weak magnetic-field regime to a magnetoexciton in intense field regime. According to the theory of Lozovik *et al.*,[22] an electron-hole pair of such an indirect *magnetoexciton* tends to be further separated in the *lateral* direction at a high magnetic field because the magnetoexciton is exposed to Lorentz force. Therefore, the oscillator strength of excitons is reduced to result in the additional quenching of the PL with increasing magnetic field.

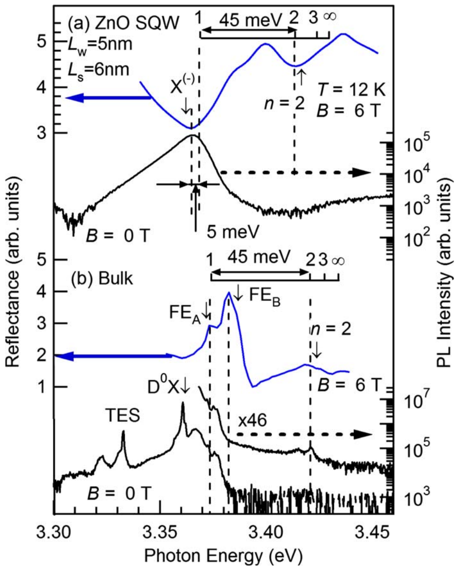

FIG. 2. (Color online) (a) Reflectance (blue, B=6 T) and photoluminescence (black, B=0 T) spectra taken at T=12 K for the ZnO/$Mg_{0.12}Zn_{0.88}O$ modulation-doped SQW with L_s=6 nm. The excitonic structures are denoted by arrows. (b) Also shown for comparison are the data for a bulk single crystal.

On the other hand, the PL intensity of the L_s=6 nm QW was slightly enhanced with increasing the magnetic fields. It seems difficult to explain the tendency in terms of excitons because the excitonic PL intensity tends to decrease or remain almost unchanged in cases of other semiconductors. An increase of intensity was reported for the *charged* exciton PL in ZnSe QWs.[18] Therefore, the PL peak should be rather assigned to charged excitons, the speculation of which will be later discussed in detail.

From the energy separation between ground and excited states of free excitons in reflectance spectra for a modulation-doped QW, we obtained the results suggesting the contribution of charged exciton to the reflectance anomaly. We show reflectance (B=6 T, blue lines) and PL (B=0 T, black lines) spectra for a modulation-doped SQW with L_s=6 nm [Fig. 2(a)] and for a single crystal [Fig. 2(b)]. There can be seen two reflectivity anomalies in the vicinity of the excitonic resonance. The weaker ones at higher energies are assignable to the n=2 states of the excitonic transitions.[23] The stronger transitions at lower energies for a single crystal can be apparently assigned as FE_A and FE_B (n=1 states). That for the modulation-doped QW is denoted as $X^{(-)}$ to indicate the overlap of free (X) and charged (X^-) exciton transitions as discussed below. The energy separation between n=1 and n=2 states is 45 meV in the crystal (3/4 of the free exciton binding energy of 60 meV)[19,23] and that was measured to be approximately 50 meV for the modulation-doped QW. The energy distance between the Rydberg series is appeared to be slightly larger than that of bulk, indicating the absence of internal electric field effects in this QW. On the other hand, the energy at the $X^{(-)}$ dip was 3.364 eV, which is significantly lower than the bulk excitonic gap (3.374 eV). This is even lower than the absorption energy of 3.382 eV in an undoped multiple QW ($L_w=5$ nm) with negligible internal electric field effects.[24] This redshift for the $L_s=6$ nm QW seems unable to be compatible with the suggested enhancement in the binding energy of excitons. The assumption of the contribution from charged excitons to the $X^{(-)}$ dip is thought to compromise these observations that are, at a glance, inconsistent with each other. Measured from the $n=2$ dip energy (3.414 eV), the resonance energy of free exciton is 3.369 eV for the QW if the binding energy remains unchanged from its bulk value (60 meV). The assumption is supported by the fact that this QW ($L_w=5$ nm) is too wide to consider the quantum size effects in ZnO.[24] The energy difference from the dip then provides a lower bound for binding energy of the charged exciton of a ZnO QW, which is estimated to be approximately 5 meV. In other words, the X^{-} contribution to the reflectance anomaly ($n=1$) is suggested based on the energy distance of the excitonic Rydberg series.

Similar to the reflectance spectrum observed in the modulation-doped QW, the PL band could also include both contributions from the charged and free excitons. In order to distinguish the respective components, we conducted polarization-sensitive magneto-PL study on the SQW relying on the selection rule for the charged exciton different from that for free exciton. It is well known that the emission from the charged exciton states tends to be strongly polarized under magnetic fields.[18] The polarized PL spectra taken at 4.2 K under various magnetic fields are shown in Figs. 3(a) and 3(b) for σ^+ and σ^- circular polarizations, respectively. It is evident that the PL is strongly polarized to σ^+ direction under a high magnetic field.[11,25] The degree of circular polarization, $\rho=(I^+-I^-)/(I^++I^-)$, and the peak energies were plotted in Figs. 4(a) and 4(b), respectively. The PL peak energies for both polarization components are almost coincident and independent of the magnetic field. At almost zero splitting of the $\sigma^{\pm}$, PL [Fig. 4(b)] and sizable ρ [Fig. 4(a)] suggest the cancellation of the Zeeman splitting between the initial and final states of optical transition.[17] If these PL transitions arise from the neutral excitons, of which ground state is the vacuum, sizable ρ should be accompanied by a finite Zeeman splitting. Therefore, the charged exciton (X^-) transition is the most plausible candidate for this radiative recombination as schematically depicted in Fig. 5. If the emission band is entirely assigned to the radiative recombination of the charged exciton, the emission must be almost fully polarized at $B \geq 40$ T. As shown in Fig. 5, the final (ground) states for the relevant emissive transition is an electron spin doublet, while the initial state is X^-, having the spin state originating from the hole $M=\pm 1/2$ (where M denotes spin degeneracy). The hole spin splitting is $|g_h|\mu_B \cong 0.07$ meV/T (g_h and μ_B being hole g factor and Bohr magneton) using the reported hole g factor.[25] At 50 T, 4.2 K, nearly all the X^-'s are in the $M=-1/2$ state, giving rise to the emission is polarized σ^+. The polarization property of the PL band is, however, not fully polarized even at 50 T. A curve of $\rho=\tanh(|g_h|\mu_B B/2k_B T)$ using $|g_h|=1.2$ is appended for comparison in Fig. 4(a), where k_B and T represent the Boltzmann constant and temperature. This suggests the possibility of the superimposed luminescence from the neutral excitons, the emission of which is apt to exhibit smaller degree of magnetic-circular dichroism. This is due to a very small effective g factor of excitons ($g_{exc}=g_e-|g_h| \cong 0.2$). Indeed, for some modulation-doped QWs with different growth parameters, the spectra are dominated by the neutral exciton only, the emission of which is almost unpolarized even at 50 T. In the unpolarized case, the neutral exciton component contributes at maximum to the emission spectra with the σ^+ direction because the intensity of neutral exciton is independent of the polarization states. Even in this case, it can be safely said that the difference between the σ^+ and σ^- components at $B \geq 40$ T is dominantly reflected the line shape of the trion radiative recombination [which is shown in Fig. 3(c)]. We performed the line-shape analysis for the "subtracted" spectra. An asymmetric line shape of the PL peak with a tail toward the lower-energy side is rather typical of charged exciton transition, obeying an inverse Boltzmann function convoluted with a Gaussian function representing the fluctuations.[26] This is because even a negatively charged exciton of large wave vector (k vector) may recombine in its decay process, the mismatch in wave vector between the charged exciton and the photon being given to the electron. The relevant equation should take the form[26]

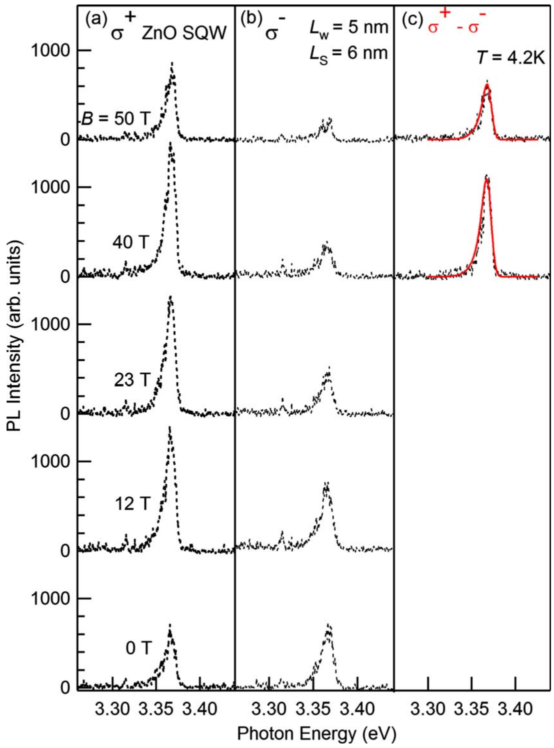

FIG. 3. (Color online) Evolution of circularly polarized [(a) σ^+ and (b) σ^-] PL spectra taken at $T=4.2$ K with a magnetic field applied normal to the QW plane ($B=0-50$ T) for the modulation-doped ZnO/$Mg_{0.12}Zn_{0.88}O$ SQW with $L_s=6$ nm. (c) Black broken lines are experimental data and solid lines are the results of fit for the differential $\sigma^+-\sigma^-$ spectra using Eq. (1).

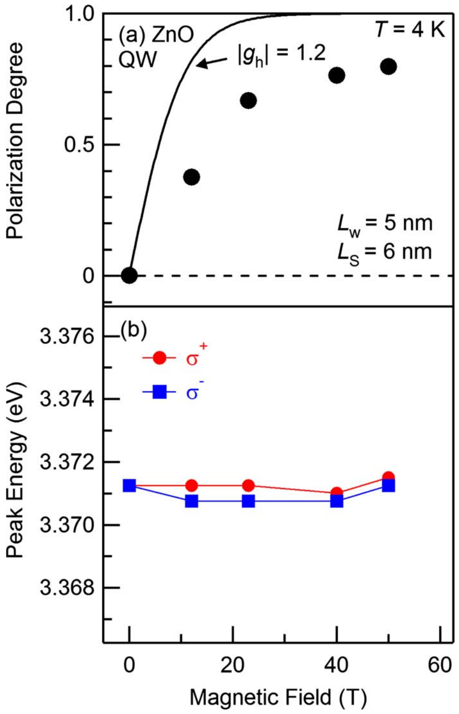

FIG. 4. (Color online) (a) Degree of circular polarization (ρ) and (b) the peak energies of the PL spectra in both polarizations for modulation-doped ZnO/$Mg_{0.12}Zn_{0.88}O$ SQW. The result of calculation using $|g_h|=1.2$ is also shown by a solid line. Circles (squares) refer to σ^+ (σ^-) polarization. The spacer thickness (L_s) is 6 nm.

$$F(\hbar\omega) = I_{X-}\int \exp[(E_{X-} - \hbar\omega - \varepsilon)/kT]\theta(E_{X-} - \hbar\omega - \varepsilon) \times \exp[-(\varepsilon/\Gamma)^2]d\varepsilon. \quad (1)$$

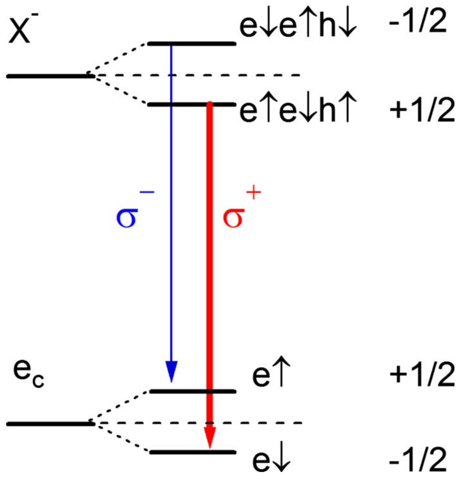

FIG. 5. (Color online) An optical transition model that illustrates the radiative transitions with a negatively charged exciton (X^-) as an initial state.

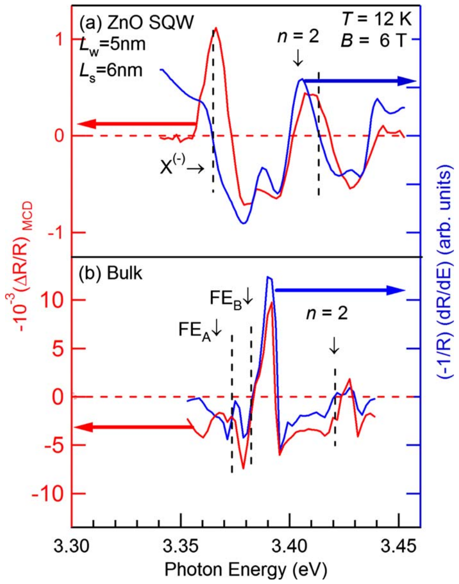

FIG. 6. (Color online) (a) MCD (red, $B=6$ T) and logarithmic derivative (blue) spectra of the modulation-doped ZnO$\|$$Mg_{0.12}Zn_{0.88}O$ QW with $L_s=6$ nm at $T=12$ K. (b) Also shown for comparison are the data for a bulk single crystal.

Here, I_{X-} is the PL intensity, E_{X-} is energy of charged exciton transition, Γ is an inhomogeneous broadening, and $\hbar\omega$ is photon energy. The subtracted spectra were well reproduced by Eq. (1) and the results of the fit are presented by solid lines in Fig. 3(c). The effective temperature (T_e) deduced from this fitting is typically $T_e=90$ K. It is significantly higher than the lattice temperature (4.2 K), suggesting the nonequilibrium nature of the charged excitons as well as the broadening effects due to the well-width fluctuation. These results of magnetic-circular dichroism give supporting evidence for the existence of charged excitons in the modulation-doped ZnO QWs. The successful description of PL line shape based on k-vector characteristics rules out the assignment to the exciton bound to neutral donors (D^0X). The asymmetrical and broad luminescence observed here is completely different from very narrow and symmetric luminescence (typically 1 meV) from D^0X as a result of no translational degree of freedom.

In case of ZnO, the free excitons involving the s-like Γ_7 conduction band and the three valence bands are labeled as A, B, and C in order of increasing exciton energy. The lowest-energy A excitons have the hole symmetry of Γ_7 and $M=\pm 1/2$, which gives rise to σ^+ polarized emission consistent with the experimental observation. It is considered that the A exciton is involved in a charged exciton complex for this modulation-doped QW.[19,25,27]

Figures 6(a) and 6(b) show the reflectance MCD spectra (red) taken for modulation-doped QW and a single crystal at $B=6$ T, respectively.[28] In the MCD spectrum of the single crystal [Fig. 6(b)], each of the neutral excitonic transitions (including $n=2$) has a derivativelike structure.[28] This is typical for the exciton-related MCD spectrum, which has been explained in terms of the rigid-shift model for neutral excitons.[28] The reflectivities for both left- and right-circular polarized light are described by the same function of energy, $R_+=f(E-E_+)$ and $R_-=f(E-E_-)$, where E_+ and E_- are the energies of the excitonic Zeeman levels. Thus, MCD should be proportional to the logarithmic energy derivative of the reflectance (blue curves in Fig. 6). We can, thus, directly determine the effective g factor for the excitons ($g_{exc} \cong 0.2$) (Ref. 28) from the relation of $(\Delta R/R)_{\mathrm{MCD}} = -2g_{ex}\mu_B B(1/R)(dR/dE)$. Previous reports suggested such a small g factor of ZnO.[25,29] The modulation technique allows the precise determination of g factor.

In the spectrum of the modulation-doped QW with $L_s = 6$ nm [Fig. 6(a)], the transition *higher* than 3.37 eV ($n = 2$) also has a derivativelike structure as observed in a single crystal. On the other hand, deviation of $(\Delta R/R)_{\mathrm{MCD}}$ from the derivative reflectance (dR/dE) becomes significant in the lower-energy side; the structure seems *unable to be* interpreted in the framework of simple neutral excitons, suggesting that the charged exciton component is overlapped to the $n = 1$ reflectance anomaly.

IV. CONCLUSION

The effects of carrier injection into ZnO SQWs were studied on its magneto-optical properties. Magnetoluminescence spectroscopy allowed us to probe the electron-hole separation of the modulation-doped QWs, dependent on the spacer layer thickness (L_s). Approaching the delta-doped layer to the QW gave rise to more efficient screening of the internal electric field. The circular magnetoluminescence with the absence of Zeeman splitting suggests the charged excitons as a result of carrier injection from the barrier layer.

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