Circular polarization of excitonic luminescence in CdTe quantum wells with excess electrons of different densities

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The circular polarization of excitonic luminescence is studied in $CdTe/Cd_{1-x}Mg_xTe$ quantum wells with excess electrons of low density in an external magnetic field. It is observed that the circular polarization of X and X^- emissions has opposite signs and is influenced by the excess electron density. If the electron density is relatively high so that the emission intensity of the negatively charged excitons X^- is much stronger than that of the neutral excitons X, a stronger circular polarization degree of both X and X^- emissions is observed. We find that the circular polarization of both X^- and X emissions is caused by the spin polarization of the excess electrons due to the electron-spin-dependent nature of the formation of X^-. If the electron density is relatively low and the emission intensity of X^- is comparable to that of X, the circular polarization degree of X and X^- emissions is considerably smaller. This fact is interpreted as due to a depolarization of the excess electron spins, which is induced by the spin relaxation of X^-.

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I. INTRODUCTION

In semiconductor quantum wells (QW's) with excess electrons of low density ($\sim 10^{10}$ cm^{-2}), some interesting phenomena occur such as negatively charged excitons (X^-),[1–4] the enhanced energy and phase relaxation rates of excitons,[5] combined exciton-cyclotron resonances,[6] and many others,[7,8] as recently have been reported. These phenomena show that an introduction of the excess electrons strongly modifies optical properties of QW's. X^- can be created by resonant excitation in absorption experiments[1] or by a neutral exciton X combining with one of the excess electrons under nonresonant excitation as in photoluminescence (PL) experiments.[2,3] It was reported that the X^- absorption is circularly polarized in magnetic fields due to the spin polarization of the excess electrons,[1,9] and the X absorption has an opposite circular polarization.[1] A possible mechanism for this is the scattering of excitons by spin-polarized excess electrons.[10] Recently, optically detected magnetic resonance (ODMR) experiments[11] have demonstrated that the formation of spin-singlet X^- created by a neutral exciton X combining with one of the excess electrons is in fact electron-spin dependent. In this paper we study the circular polarization of the X^- and X emissions in $CdTe/Cd_{1-x}Mg_xTe$ QW's with the excess electrons of low density ($\sim 10^{10}$ cm^{-2}). The electron density is controlled by optical injection. The circular polarization degree $\mathcal{P}$ of the luminescence is defined by

$$\mathcal{P}=\frac{I_+-I_-}{I_++I_-}, \tag{1}$$

where I_+ (I_-) is the luminescence intensity in the σ^+ (σ^-) circular polarization. Under the excitation by an Ar^+ laser in which case more excess electrons are introduced into the QW, we have observed a considerable degree of circular polarization of X^- and X emissions with mutually opposite signs. We find that the circular polarization of both X^- and X emissions is caused by the spin polarization of the excess electrons due to the electron-spin-dependent formation of X^-. Under the excitation by a Ti:sapphire laser, which produces fewer excess electrons in the QW, the circular polarization degree of X and X^- emissions is much smaller than in the previous case. A model based on the spin relaxation of X^- is proposed that explains this observation satisfactorily.

The paper is organized as follows: In Sec. II the investigated samples and the experimental arrangement are described. The experimental results and discussions are presented in Sec. III and conclusions are drawn in Sec. IV.

II. EXPERIMENTS

The sample studied in this work was grown by molecular beam epitaxy on (100) GaAs/CdTe hybrid substrate.[12] The structure, similar to that studied in Ref. 11, consisted of a nominally undoped 80-Å-thick CdTe single QW separated from 500-Å-thick $CdTe/Cd_{0.7}Mg_{0.3}Te$ superlattices (20 Å/20 Å) by a 200-Å-thick $Cd_{0.7}Mg_{0.3}Te$ barrier.

The density of the excess electrons was controlled by photoexcitation. Under excitation above the superlattice miniband gap, the electron-hole pairs are excited both in the QW's and in the superlattice layers. The different tunneling probabilities for electrons and holes from the superlattice miniband into the QW through the 200-Å-thick barrier lead to a low-density of excess electrons in the QW's.[6] Under excitation below the superlattice miniband gap, no electrons are excited in the superlattice layer and the optical injection of the excess electrons into the QW does not occur. In the latter, however, there exist background electrons in the QW as discussed in Sec. III. The excess (or background) electrons may combine with electron-hole pairs (or excitons) to

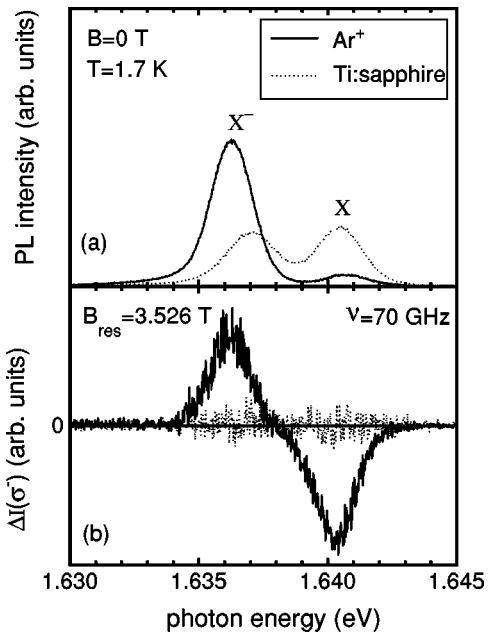

FIG. 1. (a) PL spectra of the 80-Å-wide CdTe QW taken under the excitation by an Ar^+ laser (514 nm, solid curve) and a tunable Ti:sapphire laser (732 nm, dotted curve) at $B=0$ T and at $T=1.7$ K. The excitation energies are, respectively, above and below the superlattice miniband gap energy ($E_g=1.8$ eV). (b) Microwave-induced changes of the X^- and X emission intensity under the two excitations at the resonant magnetic field meeting the condition of the electron spin resonance. The formula $|g_e^*|\mu_B B_{res}=h\nu$ yields the electron effective g factor $|g_e^*|=1.419$. The excitation intensity is kept at a low level of 0.06 W/cm^2 for both (a) and (b).

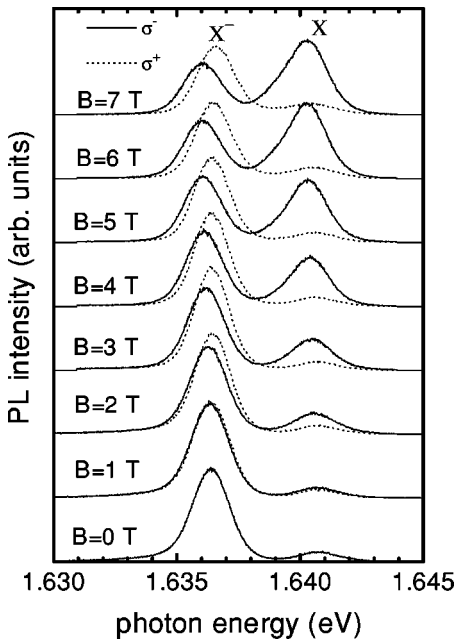

FIG. 2. The σ^- (solid curves) and σ^+ (dotted curves) components of PL spectra at different magnetic fields under excitation by the Ar^+ laser.

form X^-. In our experiments, an argon-ion laser (Ar^+) (514 nm) and a tunable Ti:sapphire laser (732 nm) were used to provide the excitation above and below the superlattice miniband gap ($E_g=1.8$ eV), respectively.

The sample was placed in an optical cryostat with a split magnet system in the Faraday geometry with the magnetic field applied perpendicular to the QW plane. The exciting light of the Ar^+ and Ti:sapphire laser was linearly polarized. The σ^+ and σ^- circularly polarized components of the luminescence were extracted via a $\lambda/4$ wave plate and a linear polarizer. The luminescence was dispersed with a single-grating 1-m spectrometer and detected by a cooled GaAs photomultiplier. For ODMR experiments, microwaves (70 GHz) were irradiated onto the sample through a rectangular waveguide. Microwaves were modulated at a frequency of 45 Hz and the synchronous changes of the luminescence intensity were recorded by a two-channel photon counter.

III. RESULTS AND DISCUSSIONS

Figure 1(a) shows the PL spectra under the excitation by the argon-ion laser (514-nm line, solid curve) and the Ti:sapphire laser tuned to 732 nm (dotted curve) at $B=0$ T and at $T=1.7$ K. The excitation intensity is kept at a low level of 0.06 W/cm^2. Under the excitation by the argon-ion laser, i.e., when the excess electrons are optically injected into the QW's, two peaks are observed in the PL spectrum. The peak at 1.6405 eV is identified as the heavy-hole exciton X and the peak at 1.6363 eV, which is by 4.2 meV lower in energy than X, is identified as the negatively charged exciton X^-.[11] Under the excitation by the Ti:sapphire laser, no electrons are transferred from the superlattice into the QW. However, the X^- peak is still observed. This may be due to that there exist background electrons in the CdTe QW, which originate from residual impurities in the barriers.[13] Since the background electron density is much smaller than that under the excitation by the argon-ion laser, the intensity of the X^- relative to the X emission is smaller as there are only very few excess electrons are available for X^- formation. Note that the energy position of the X^- line under the excitation by the Ti:sapphire laser is 0.7 meV higher than that under the excitation by the argon-ion laser. This result is in accord with the recent observation in absorption spectra that the X-X^- splitting is equal to the Fermi energy plus the X^- binding energy,[14] i.e., the X-X^- splitting is electron-density dependent. The change of the electrostatic potential due to the space-charge effect of different electron densities can be neglected because the energy position of the X line does not shift under the two types of excitations [see Fig. 1(a)].

Figure 2 plots the PL spectra in σ^- (solid curves) and σ^+ (dotted curves) circular polarizations under the excitation by the argon-ion laser at different magnetic fields. With increasing magnetic field, the X emission intensity in σ^- becomes stronger than that in σ^+. In contrast, the X^- emission intensity in σ^- becomes weaker than that in σ^+. The magnetic field dependence of X and X^- emission intensity in the σ^- (solid symbols) and σ^+ (open symbols) polarizations under the excitation by the Ar^+ laser is presented in Fig. 3. With increasing magnetic field, the X emission intensity in σ^- increases drastically, whereas that in σ^+ does not vary. Si-

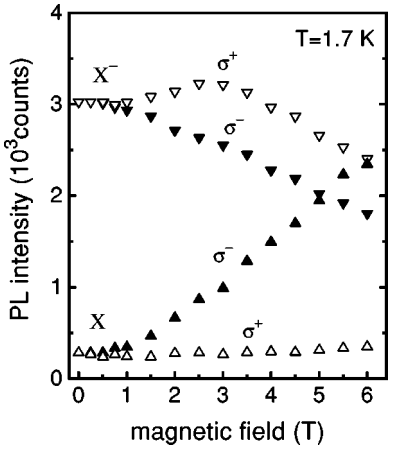

FIG. 3. Magnetic field dependence of the X and X^- emission intensity in σ^- and σ^+ under the excitation by the Ar^+ laser.

multaneously, the X^- emission intensity in σ^- decreases drastically and monotonically from 0 T to 7 T, whereas that in σ^+ shows first a small increase for $B<3$ T and then it decreases drastically for $B>3$ T.

The polarization behavior is also observed under the excitation by the Ti:sapphire laser (not shown here), but the circular polarization degree of the X and X^- emission defined by Eq. (1) is much smaller than that under the excitation by the argon-ion laser. In Fig. 4 we plot the circular polarization degree of the X and X^- emissions as a function of the magnetic field under the two types of excitations.

Obviously, the variation in the circular polarization behavior is due to the introduction of the excess electrons of different densities. Now we discuss the mechanism responsible for the stronger circular polarization of X and X^- emission under the excitation by the Ar^+ laser. As the linearly polarized light with the photon energy far above the band gap energy is used for excitation, the effect of optical pumping in the circular polarization (see Ref. 15) does not occur in our experiments. As a results, the stronger circular polarization of the X and X^- emissions is not caused by the photoexcitation. Instead, we propose that the circular polarization of the X and X^- emissions stems from the spin polarization of the excess electrons since the formation of X^- is electron-spin dependent as described below.

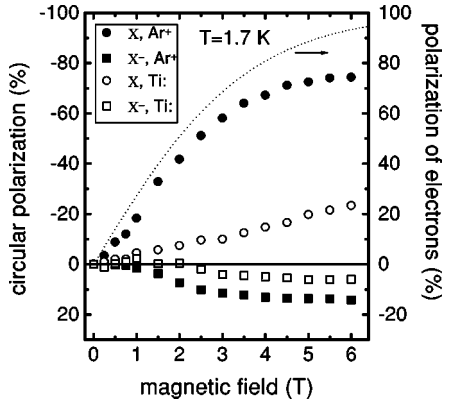

FIG. 4. The circular polarization degree of X and X^- emissions as a function of the magnetic field under the excitation by the Ar^+ laser (solid symbols) and by the Ti:sapphire laser (open symbols). For comparison, the calculated spin polarization degree of the excess electrons by using the formula $P_e=\tanh(|g_e^*|\mu_B B/2kT_e)$ is also plotted as a function of the magnetic field (dotted curve).

The spin states of X and X^- are characterized by their spin projection along the magnetic-field direction z. The optically allowed heavy-hole exciton has two spin states, $|+\frac{1}{2},-\frac{3}{2}\rangle=|-1\rangle$ and $|-\frac{1}{2},+\frac{3}{2}\rangle=|+1\rangle$, which decay by the σ^- and σ^+ emissions, respectively. The spin singlet X^- with two antiparallel electron spins has two spin states as well: $|-\frac{1}{2},+\frac{1}{2},-\frac{3}{2}\rangle$ and $|+\frac{1}{2},-\frac{1}{2},+\frac{3}{2}\rangle$. The former decays by emitting the σ^--polarized photon with the $S_z=-\frac{1}{2}$ electron left, and the later by emitting the σ^+-polarized photon with the $S_z=+\frac{1}{2}$ electron left. The formation and recombination process of X^- can be written as[11]

$$e_{-1/2}+X_{-1}\rightarrow X^-_{-3/2}\rightarrow \text{photon}(\sigma^-)+e_{-1/2}, \tag{2}$$

$$e_{+1/2}+X_{+1}\rightarrow X^-_{+3/2}\rightarrow \text{photon}(\sigma^+)+e_{+1/2}. \tag{3}$$

The above expressions clearly show that the formation of the spin singlet X^- is electron-spin dependent.

With increasing magnetic field, the excess electrons are spin polarized, i.e., the $|+\frac{1}{2}\rangle$ spin state becomes more populated than the $|-\frac{1}{2}\rangle$ spin state. Obviously, the decrease in the population of $|-\frac{1}{2}\rangle$ electron state leads to a corresponding decrease in the formation of $X^-_{-3/2}$ [see Eq. (2)] as compared to that at $B=0$ T. As a result, the X^- emission intensity in σ^- decreases, and the X emission intensity in σ^- increases at the expense of the X^- emission (see Fig. 3, solid symbols). On the other hand, the increase in the population of the $|+\frac{1}{2}\rangle$ electron state causes an increase in the formation of $X^-_{+3/2}$ [see Eq. (3)]. This is the reason why the X^- emission intensity in σ^+ increases with increasing magnetic field for $B<3$ T (see Fig. 3, open downward-pointing triangles). However, the X^- emission intensity in σ^+ has a weaker magnetic-field dependence than that in σ^-. The reason is as follows: the formation of $|+\frac{3}{2}\rangle$ X^- is not very sensitive to the increase of the electron population of $|+\frac{1}{2}\rangle$ states with the increase of the magnetic field because the excitons can already find enough excess electrons to form X^- even at zero magnetic field (X^- has a stronger emission than X as shown in Fig. 1). In fact, the σ^+ component of the PL spectrum in a magnetic field is very similar to the PL spectrum at $B=0$ T (see Fig. 2).

Note that at $B>3$ T the X^- emission intensity in σ^+ decreases drastically as the X^- emission intensity in σ^- with increasing magnetic field (see Fig. 3, downward triangles). This can be explained by the spin relaxation of X^-. As the two electron spins in X^- are antiparallel, the total spin of the X^- is equivalent to the spin of the heavy hole.[16] Thus a spin flip of a hole induces a spin flip of the X^-. As the hole state is not a pure state in semiconductors, the spin relaxation of holes is rather fast,[17] and so is that for X^-. Therefore, the decrease of X^- emission intensity in σ^+ above 3 T just follows the decrease of X^- emission intensity in σ^- with increasing magnetic field.

Note that although the X emission intensity in σ^- increases drastically with increasing magnetic field, the X emission intensity in σ^+ does not vary (see Fig. 3, open upward-pointing triangles). This phenomenon implies that the spin relaxation of X between $|+1\rangle$ and $|-1\rangle$ states is very slow compared to the recombination process of X.

From the above discussion, we see that via the electron-spin-dependent formation of X^-, the spin polarization of the excess electrons results in the X^- emission intensity being weaker in σ^- than in σ^+ and the X emission intensity being stronger in σ^- than in σ^+ in the presence of a magnetic field. As a result, the circular polarization of the X and X^- emission have opposite signs (see Fig. 4). For comparison, in Fig. 4, we plot the calculated spin polarization degree of the excess electrons as a function of the magnetic field by using the formula $P_e=\tanh(|g_e^*|\mu_B B/2kT_e)$ where $|g_e^*|$ is measured to be 1.419 [see Fig. 1(b)]. With increasing magnetic field, the spin polarization of the excess electrons increases and, correspondingly, the circular polarization degree of X and X^- emissions follows such trend. Therefore, we can conclude that the spin polarization degree of the excess electrons determines the circular polarization degree of both X and X^- emissions. We can say that the spin-polarized excess electrons play a role similar to that of ''optical pumping.''[15] In the steady optical pumping experiments the spin relaxation tends to weaken the initial circular polarization of the luminescence. Thus the spin relaxation of X^- reduces the circular polarization degree of the X^- emission. Because the spin relaxation rate of X can be neglected compared to its recombination rate, as discussed above, the circular polarization degree of the X emission is larger than that of the X^- emission (see Fig. 4).

It was earlier reported that the circular polarization of the X and X^- absorptions has an opposite sign,[1] similar to the circular polarization of the X and X^- emissions discussed above. The circular polarization of the X absorption was attributed to the scattering of X by the spin-polarized excess electrons.[10] Obviously, the mechanisms for the circular polarization of the X emission and the X absorption are different.

Now we shall discuss the mechanism of the small circular polarization of X and X^- emissions under the excitation by the Ti:sapphire laser. As the excitation power is kept at a very low level (~ 0.06 W/cm^2), the heating effect of photoexcitation can be neglected, i.e., the electron temperatures are the same under the excitation by either the Ar^+ laser or the Ti:sapphire laser. From Eqs. (2) and (3), we can find that the spin relaxation (or spin-flip) process of X^- will lead to a spin flip of the excess electrons via a recombination of X^-. If the electron density is so low that majority of the electrons are bound to X^- under the continuous photoexcitation, the spin flip of X^- tends to equalize both the X^- population and the electron populations in their two spin states. As a result, a spin depolarization of the excess electrons will take place. If the electron density is so high, on the other hand, that only a small number of the excess electrons are bound to X^- and the other most of the excess electrons are free, the spin depolarization of the small number of the excess electrons that are bound to X^- can be neglected. Because the spin polarization degree of the excess electrons determines the circular polarization degree of the X and X^- emissions, as discussed above, the spin depolarization of the excess electrons results in the circular polarization degree of the X^- (and X) emission under excitation by the Ti:sapphire laser being smaller than that under the excitation by Ar^+ laser (see Fig. 4).

The spin depolarization of the excess electrons can be further verified by ODMR experiments[11] where microwaves are irradiated onto the sample as a perturbation to the electronic system and the microwave-induced changes of the X and X^- emission intensity are monitored. Under the electron-spin-resonance (ESR) condition, i.e., when $|g_e^*|\mu_B B=h\nu$, if the excess electrons are spin polarized, the electrons are resonantly excited from the $|+\frac{1}{2}\rangle$ spin level to the $|-\frac{1}{2}\rangle$ spin level. This should induce an increase of the X^- emission and a decrease of the X emission in the σ^- polarization under the excitation by the Ar^+ laser [see Fig. 1(b), solid curve]. However, the microwaves induce no intensity change under the continuous excitation by Ti:sapphire laser at the resonant magnetic field [see Fig. 1(b), dotted curve], indicating that ESR does not occur in this case. The reason for it can be that the excess electrons become unpolarized due to the relaxation of X^- under under continuous excitation by Ti:sapphire laser.

IV. CONCLUSIONS

We have studied the influence of excess electrons on the circular polarization of excitonic luminescence in a $CdTe/Cd_{1-x}Mg_xTe$ quantum well. It is found that the circular polarization of both X^- and X emissions is caused by the spin polarization of the excess electrons due to the electron-spin-dependent nature of the formation of X^-, and, moreover, it is influenced by the electron density. If the electron density is relatively high so that the emission intensity of the negatively charged exciton X^- is much stronger than that of the neutral exciton X, a stronger circular polarization of both X and X^- emissions is observed. If the electron density is relatively low, so that the emission intensity of X^- is comparable to that of X, the circular polarization degree of X and X^- emission is considerably smaller. This is explained by a spin depolarization of the excess electrons induced by the spin relaxation of X^- under the photoexcitation.

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