Experimental measurement of microwave-induced electron spin-flip time

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(Received 21 August 2000; accepted for publication 8 November 2000)

The electron spin resonance (ESR) is optically detected by monitoring the microwave-induced changes in the circular polarization of the neutral exciton (X) and the negatively charged exciton (X^-) emission in CdTe quantum wells with low density of excess electrons. We find that the circular polarization of the X and X^- emission is a mapping of the spin polarization of excess electrons. By analyzing the ESR-induced decrease in the circular polarization degree of the X emission, we deduce the microwave-induced electron spin-flip time $>0.1\ \mu$s, which is much longer than the recombination time of X and X^-. This demonstrates that the optically detected ESR in type I quantum wells with low density of excess electrons does not obey the prerequisite for the conventional optically detected magnetic resonance.
[DOI: 10.1063/1.1338961]

The optically detected magnetic resonance (ODMR) technique is a powerful tool to study recombination processes in semiconductors,[1] as well as to determine the effective Landé g factor. According to previous views,[2–4] a prerequisite to realize ODMR is that the microwave-induced spin-flip time should be shorter than or at least comparable to the optical transition time. This is the reason why ODMR is mostly applied to study the slow optical transitions on the microsecond timescale, such as the indirect transitions in type II quantum wells (QWs).[2,3] Up to now, there is no report on the measurement of the microwave-induced spin-flip time, and thus the prerequisite for ODMR was never checked experimentally. Recently, electron spin resonance (ESR) was successfully detected by monitoring the microwave-induced intensity changes of the neutral exciton (X) and the negatively charged exciton (X^-) emissions (fast optical transitions on the subnanosecond timescale) in type I CdTe QWs with low density of excess electrons.[5] The negatively charged exciton X^- is a three-particle complex, i.e., two electrons bound to a hole.[6–10] In this letter we report on an experimental measurement of the microwave-induced electron spin-flip time by analyzing the microwave-induced changes in the circular polarization degree of the X emission at the resonant magnetic field. We demonstrate experimentally that the optical detection of ESR occurs with the microwave-induced spin-flip time much longer than the optical transition time, and therefore the prerequisite for the conventional ODMR is not satisfied in our case.

The sample studied in this work was type I CdTe/$Cd_{0.7}Mg_{0.3}Te$ multiple QWs grown by molecular beam epitaxy on (100) GaAs/CdTe hybrid substrates.[10] This structure consists of six QW units, each of which is 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 200-Å-thick $Cd_{0.7}Mg_{0.3}Te$ barriers. Under the excitation above the superlattice miniband gap (1.8 eV) by an argon ion laser (514 nm), the different tunneling probabilities for electrons and holes from superlattice miniband into QW through the 200-Å-thick barriers provide the low-density excess electrons in the QWs. The excess electrons combine with excitons to form X^-.

The sample was placed in an optical cryostat with a split magnet system in Faraday geometry where the magnetic field is perpendicular to the QW plane. The excitation light from the argon ion laser is linearly polarized and the excitation intensity is kept at a low level of 0.1 W/cm^2. 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, a back wave oscillator was used as the microwave source. The microwave frequency was set to 70 GHz and the maximal power output of 200 mW was used. Microwaves modulated at a frequency of 45 Hz were irradiated onto the sample through a rectangular waveguide. The luminescence intensities with microwave irradiation on and off were simultaneously recorded by a two-channel photon counter.

Figure 1 shows the low-temperature photoluminescence spectra at $B=0$ T and 4 T. The peak at high energy side is assigned to neutral heavy-hole excitons X and another peak, 4.1 meV low in energy, is assigned to X^-. At $B=4$ T, the σ^- and σ^+ components of the photoluminescence spectra are very different. The X emission intensity in σ^- polarization is much stronger than that in σ^+ polarization, whereas the X^- emission intensity in σ^- polarization is weaker than that in σ^+ polarization. This phenomenon is also observed at other magnetic fields (not shown here).

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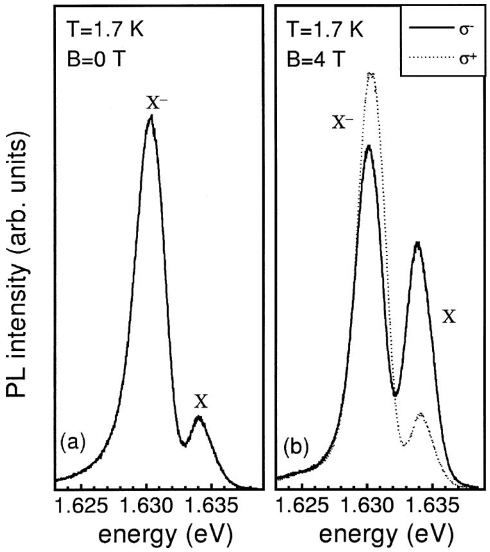

FIG. 1. Photoluminescence spectra of 80-Å-CdTe-200-Å-$Cd_{0.7}Mg_{0.3}Te$ QWs under the excitation of an argon ion laser (514 nm) at (a) $B=0$ T and (b) $B=4$ T. The excitation intensity is kept at a low level of 0.1 W/cm^2 and the density of excess electrons introduced by optical injection is around 10^{10} cm^{-2}.

Figure 2 shows the measured circular polarization degree of the X and X^- emissions as a function of magnetic field (solid curves) with and without microwave irradiation. The circular polarization degree of the luminescence is defined by

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

where $I_+(I_-)$ is the luminescence intensity in $\sigma^+(\sigma^-)$ circular polarization, respectively. With increasing magnetic field, the circular polarization degree of the X and X^- emissions increases. The microwave irradiation results in a small decrease of the circular polarization degree for the X and X^- emissions at each magnetic field. Furthermore a resonant decrease of the circular polarization degree of the X and X^- emissions is observed at $B=3.424$ T where ESR occurs.[5] $|g_e^*|\mu_B B=h\nu$ yields the electron Landé factor $|g_e^*|=1.461$.

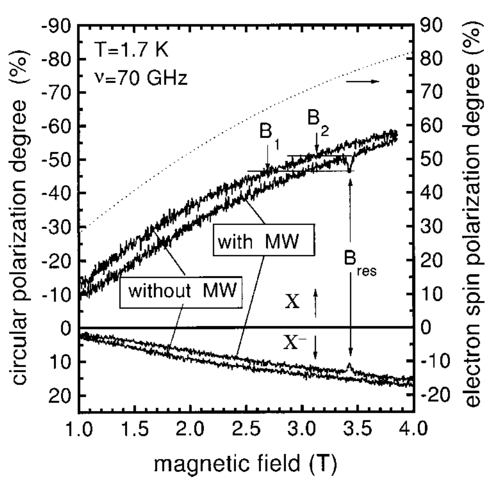

FIG. 2. Circular polarization degree of X and X^- emissions as a function of magnetic field (solid curves) with and without microwave irradiation. ESR occurs at $B_{res}=3.424$ T with microwave frequency at 70 GHz. For comparison, the electron spin polarization degree calculated by using the formula $P_s^e=\tanh(|g_e^*|\mu_B B/2k_BT_e)$ is plotted as a function of magnetic field at $T_e=1.7$ K (dotted curve).

The observation of ESR in the circular polarization of the X and X^- emissions can be explained by the spin dependent formation of X^- described as[5]

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

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

At high magnetic fields, excess electrons are more populated in the $|+\frac{1}{2}\rangle$ state. Both ESR and the microwave heating induce a population transfer from the $|+\frac{1}{2}\rangle$ to $|-\frac{1}{2}\rangle$ state. This enhances the $X^-_{-3/2}$ emission (in σ^-) and decreases the X_{-1} emission (in σ^-), whereas they have no obvious influence on the $X^-_{+3/2}$ or X_{+1} emissions (in σ^+). A detailed discussion is presented in Ref. 5. As a result, both ESR and the microwave heating result in a decrease of the circular polarization degree of the X and X^- emissions.

Microwave irradiation can be regarded as a perturbation to the spin polarization of excess electrons. Since a decrease of the electron spin polarization manifests as a decrease of the circular polarization degree of the X and X^- emissions, we see a correlation between the circular polarization of the X and X^- emissions and the spin polarization of excess electrons. The calculated spin polarization degree of excess electrons as a function of magnetic field is plotted in Fig. 2 (dotted curve). With increasing magnetic field, the circular polarization degree of the X and X^- emissions increases monotonically, just following the spin polarization degree of excess electrons. Therefore, we can conclude that the spin polarization of excess electrons causes the circular polarization of the X and X^- emissions, in other words, the circular polarization degree of the X and X^- emissions is a mapping of the spin polarization degree of excess electrons. Based on this conclusion we can deduce the microwave-induced electron spin-flip time under ESR condition by solving the rate equations as presented in the following.

The rate equations of excess electrons under the microwave irradiation at the resonant magnetic field are

$$\frac{dn_-}{dt}=G(n_+-n_-)-\frac{n_--n_-^0}{T_1}, \tag{4}$$

$$\frac{dn_+}{dt}=-G(n_+-n_-)-\frac{n_+-n_+^0}{T_1}, \tag{5}$$

where n_- and n_+ are the electron population in the $|-\frac{1}{2}\rangle$ (upper) and $|+\frac{1}{2}\rangle$ (lower) spin state respectively, and T_1 is the electron spin relaxation time. The first term on the right-hand side of Eqs. (4) and (5) is the ESR-induced population transfer where G describes the spin-transition (spin-flip) rate. The second term accounts for the loss of electron population through the spin relaxation of electrons. To reach Boltzmann equilibrium, $n_-\rightarrow n_-^0$ and $n_+\rightarrow n_+^0$ with $n_-^0/n_+^0=\exp(-|g_e^*|\mu_B B/k_BT_e')$ where T_e' is the electron temperature under microwave irradiation. Note that $n_++n_-=n_+^0+n_-^0$. The microwave heating manifests as a rise of the electron temperature.

The electron spin polarization degree is defined as $P_s^e=(n_+-n_-)/(n_++n_-)$. By solving Eqs. (4) and (5) in steady state, we can deduce the electron spin polarization degree under the microwave irradiation at the resonant magnetic field

$$P_s^e = \frac{1}{1+2GT_1} \tanh \frac{|g_e^*|\mu_B B_{\mathrm{res}}}{2k_B T_e'}, \tag{6}$$

where T_e' is the electron temperature with microwave irradiation. Without microwave irradiation ($G=0$, $T_e'=T_e$), the electron spin polarization degree P_s^e at magnetic field B is reduced to

$$P_s^e = \tanh \frac{|g_e^*|\mu_B B}{2k_B T_e}, \tag{7}$$

where T_e is the electron temperature without microwave irradiation.

From Fig. 2, we see that the circular polarization degree of the X emission at $B_{\mathrm{res}}=3.424\,\mathrm{T}$ under ESR and the microwave heating is equal to that at $B_1=2.70\,\mathrm{T}$ without microwave irradiation and the circular polarization degree of the X emission at $B_{\mathrm{res}}=3.424\,\mathrm{T}$ under the microwave heating but without ESR is equal to that at $B_2=3.14\,\mathrm{T}$ without microwave irradiation. Because the circular polarization degree P_c is a mapping of P_s^e, we obtain the following two equations for the spin polarization degree of excess electrons:

$$\tanh \frac{|g_e^*|\mu_B B_{\mathrm{res}}}{2k_B T_e'} = \tanh \frac{|g_e^*|\mu_B B_2}{2k_B T_e}, \tag{8}$$

$$\frac{1}{1+2GT_1} \tanh \frac{|g_e^*|\mu_B B_{\mathrm{res}}}{2k_B T_e'} = \tanh \frac{|g_e^*|\mu_B B_1}{2k_B T_e}. \tag{9}$$

We take T_e equal to the lattice temperature, i.e., $T_e=1.7\,\mathrm{K}$ as the excitation intensity is kept at a low level of $0.1\,\mathrm{W/cm^2}$ and the heating effect of photoexcitation can be neglected. In the above discussions, we have adapted the circular polarization of the X emission, not that of the X^- emission, as the mapping of the electron spin polarization in order to reduce errors because the circular polarization degree of the X emission is much larger than that of the X^- emission.

By placing $B_2=3.14\,\mathrm{T}$ into Eq. (8), we obtain $T_e'=1.85\,\mathrm{K}$. The microwave-heating induced rise of electron temperature is $T_e'-T_e=0.15\,\mathrm{K}$. This indicates that the spin polarization degree of excess electrons is very sensitive to the electron temperature because the thermal energy $k_B T=0.15\,\mathrm{meV}$ at $T=1.7\,\mathrm{K}$ is close to the spin splitting $\Delta E=|g_e^*|\mu_B B=0.29\,\mathrm{meV}$ at $B=3.424\,\mathrm{T}$.

Placing $B_1=2.70\,\mathrm{T}$, $T_e'=1.85\,\mathrm{K}$, and $T_e=1.7\,\mathrm{K}$ into Eq. (9), we obtain $GT_1=0.05$. In CdTe the electron spin relaxation time is measured to be $T_1>6$ ns by the Hanle effect at very low magnetic fields.[11] Therefore we get the microwave-induced electron spin-flip time $\frac{1}{G}>0.1\,\mu\mathrm{s}$, much longer than the recombination time of X and X^- ($\sim 0.2\,\mathrm{ns}$) in CdTe QWs.[12] This shows that the prerequisite for the conventional ODMR which demands that the microwave-induced electron spin-flip time should be shorter than or at least comparable to the recombination time[2–4] is not satisfied for optically detected ESR. The successful realization of optically detected ESR in type I QW is attributed to two reasons:[5] (1) the long-living excess electrons; (2) the electron-spin dependent formation and the electron–spin conserving recombination of X^- [see Eqs. (2) and (3)]. Although an excess electron takes part in the formation of X^-, it does not participate in the recombination of X^- and furthermore its spin is left unchanged after the recombination of X^-.[13] Therefore, the creation of X^- does not influence the spin lifetime of excess electrons and ESR can induce a decrease of the electron spin polarization degree which manifests as a decrease of the circular polarization degree of the X and X^- emission. In fact, optically detected ESR observed is an effect of magnetic resonance in a long-lived electron population on the circularly polarized components of the emission of a rapidly decaying exciton population, and thus optically detected ESR does not need to obey the prerequisite for the conventional ODMR.

In summary, we have observed ESR by monitoring the microwave-induced changes in the circular polarization of X and X^- emission in CdTe quantum wells with excess electrons of low density. We identify that the circular polarization of X and X^- emission is a mapping of the spin polarization of excess electrons. The microwave-induced electron spin-flip time is determined to be $>0.1\,\mu\mathrm{s}$, which is much longer than the recombination time of X and X^- lifetime. Optically detected ESR does not need to obey the prerequisite for the conventional ODMR and is feasible in type I QWs by introducing low density of long-living excess electrons.

The authors gratefully acknowledge Professor Hou-Zhi Zheng and Professor F. H. Yang for useful discussions. The experiments were performed in Würzburg. C. Y. was supported by the Volkswagen Foundation. The work was partially supported by the Deutsche Forschungsgemeinschaft via Grant No. Os98-5 and the Volkswagen Foundation.

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