Exciton-Electron Interaction in Quantum Wells with a Two Dimensional Electron Gas of Low Density

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II–VI quantum well structures containing a 2DEG of low density have been investigated by means of polarized photoluminescence, photoluminescence excitation and reflectivity in external magnetic fields up to 20 T. The spin splittings of the excition X and the negatively charged exciton X^- are measured as a function of the magnetic field strength. The behavior of the magnetic-field-induced polarization degree of the luminescence line related to X^- demonstrates the formation process of negatively charged excitons from excitons and free carriers polarized by the external magnetic field. We have determined the binding energies of the trion formed either with the heavy-hole or the light-hole exciton. The optically detected magnetic resonance (ODMR) technique was for the first time applied to study the optical transition processes in a nanosecond timescale. The electron ODMR was observed with the detection on either the direct exciton or the negatively charged exciton X. Further evidence for the interaction of excitons with the electrons of the two dimensional gas are demonstrated by a combined exciton-cyclotron resonance line observed in reflectivity and luminescence excitation, shake-up processes observed in photoluminescence as well as inelastic and spin-dependent scattering processes.

Effects resulting from the exciton-electron interaction in the presence of a two-dimensional electron gas (2DEG) of low density at $n_e a_B \ll 1$, where n_e is the electron concentration and a_B is the exciton Bohr radius, became a subject of intensive investigations very recently. This interest has been stimulated by the observation of a negatively charged exciton X^- in CdTe/(Cd,Zn)Te modulation-doped quantum well (QW) structures [1].

In this paper we review several new effects observed in structures where the exciton interacts with a 2DEG of low carrier density. In detail we discuss: the spin splitting and polarization dependence of X and X^- in high magnetic field [2]; the combined exciton-cyclotron resonance [3]; the optically detected magnetic resonance (ODMR) on X^- [4]; the shake-up process [5,6] and the spin-dependent broadening of excitonic states [7].

1. Sample Structures and Experimental Details

In this study we performed measurements on two different types of structures. We used modulation-doped CdTe/(Cd,Mg)Te or ZnSe/(Zn,Mg)(S,Se) QWs with a 2DEG of low density of about $1.5 \cdot 10^{10}\,\mathrm{cm}^{-2}$. The structures were grown by molecular-beam epitaxy on (100) oriented GaAs substrates and are selectively doped with iodine or chlorine separated by a spacer layer from the QW. Furthermore, we investigated specially designed structures, where we varied the carrier concentration by external optical illumination [3]. To achieve this the heterostructures are sandwiched between short period superlattices, where the QWs are separated from the superlattice (SL) by 20 nm thick barriers, which leads to a much higher probability for the electrons to tunnel from the SL to the QW, as compared to holes, due to the difference in the effective masses. Consequently, the electron concentration in the QW can be varied accurately by the intensity of illumination with a radiation energy exceeding the SL band gap. For the experiments with additional microwave illumination we used a back wave oscillator, which frequency can be tuned from 55 GHz to 80 GHz by the application of different dc voltages. For these ODMR experiments the microwaves were chopped at 45 Hz and the synchronous changes of the PL intensities were recorded by a two-channel photoncounter.

2. Magneto-Optical Study of the Trion

2.1. Zeeman splitting and polarization degree. In the first part of this paper we concentrate on the properties of negatively charged exciton states in the magnetic field range extended up to 20 T. PL was excited with a Ti: sapphire laser at energies below the band gap of the barriers and the external magnetic fields were applied perpendecular to the QW layers (Faraday geometry).

In Fig. 1, *a* the PL and PLE spectra detected for a 8 nm-thick CdTe/$Cd_{0.7}Mg_{0.3}$Te QW at a temperature of 1.6 K and at zero magnetic field are shown. The exciton line X dominates in the PLE spectrum but is much weaker than the negatively charged exciton line X^- in the PL spectrum, which reflects the strong probability for excitons to be bound in the X^- complex. This situation is changed under applied magnetic fields when the 2DEG is polarized (Fig. 1, *b*). At 7 T the σ^- polarized PL component of the exciton line increases strongly in intensity and becomes comparable with

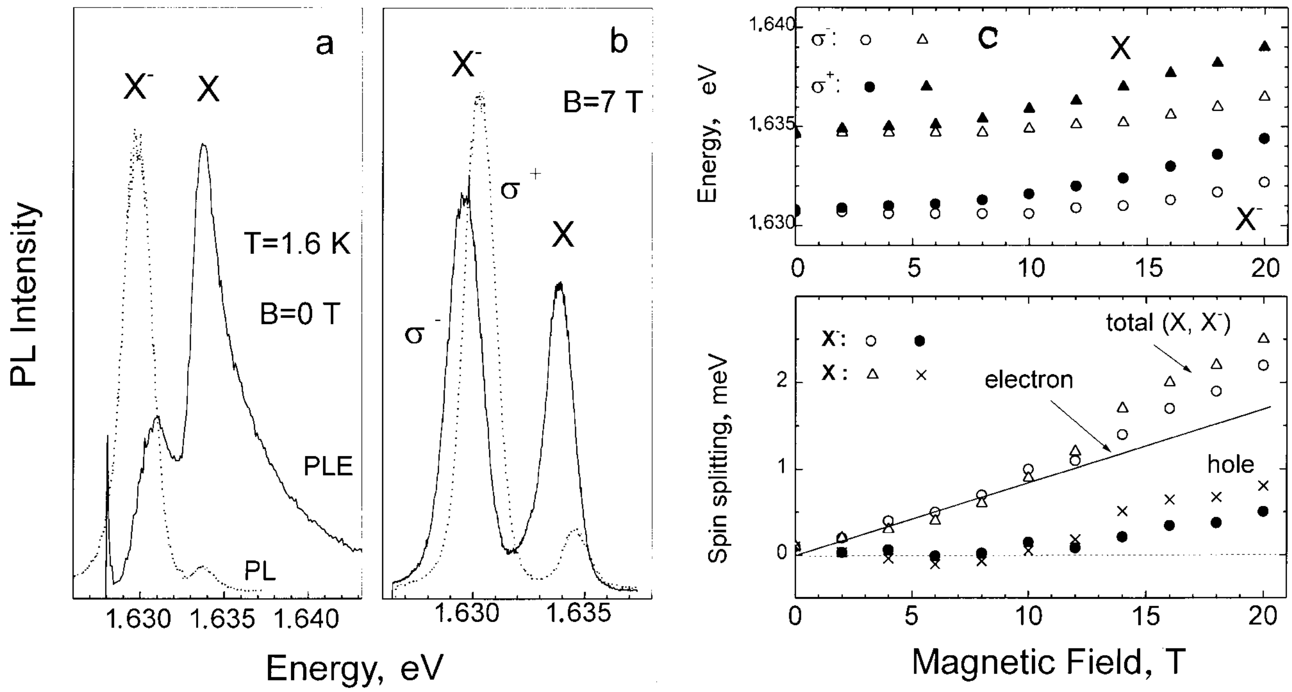

Figure 1. *a* — photoluminescence and PLE spectra of an 8 nm-thick $CdTe/Cd_{0.7}Mg_{0.3}Te$ modulation-doped SQW structure. X and X^- label the heavy-hole exciton and the negatively charged exciton lines; *b* — PL spectra taken in the magnetic field of 7 T are shown for two circular polarizations σ^+ (dashed) and σ^- (solid) lines, respectively; *c* — magnetic field dependence of the PL line positions (upper panel) and the Zeeman splitting (lower panel). The electron spin splitting calculated for $g_e = -1.46$ is plotted by a solid line. The heavy-hole splitting is calculated from the electron and the X and X^- splitting respectively. For details see text.

the X^- line intensity. We also stress here that at 7 T the X and the X^- PL lines are polarized with opposite signs and for the X^- line the high energy component is stronger in intensity than the lower one. Detailed dependencies of X and X^- PL intensities on the magnetic field strength are plotted in the upper panel of Fig. 3 and will be discussed below.

For a detailed analysis of the observed PL polarization a precise knowledge about the spin splitting of the exciton and the free carrier states is essential first. The experimentally determined spin splittings for excitons and X^- are very close to each other (Fig. 1, *c*, upper panel). The excitonic g factor has a positive sign (lower part of Fig. 1). The electron g factor at the bottom of the conduction band in an 8 nm thick $CdTe/Cd_{0.7}Mg_{0.3}Te$ QW ($g_e = -1.46$) is known with high accuracy [4,8]. The heavy-hole spin-splitting was deduced by subtracting the electron splitting from the excitonic one. The hole splitting is zero at magnetic fields below 12 T and increases at higher fields giving rise to a positive g factor value. There is no significant difference observable whether the hole g factor is determined by the splitting of X or X^-. Schematically the spin splitting for 2DEG elecrons, excitons and X^- are presented in Fig. 2. Note that the X^- state is splitted due to the heavy-hole contribution only, but the spin splitting of the X^- optical transitions is also determined by the splitting of the conduction band states (g_e), as the 2D electron is the final state after X^- recombination (for details see referance [9]).

The magnetic-field-induced polarazation degree for X and X^- PL lines in QWs with and without 2DEG is displayed in lower part of Fig. 3. In undoped QWs the X^- line is unpolarized and the X line polarization increases linearly with a slope of $0.04T^{-1}$ only, which is considerably smaller than the thermal equilibrium value of $0.3T^{-1}$. In modulation-doped QWs the polarization degree of the X^- line is nonmonotonic and alters its sign at 12 T and the exciton line polarization is strongly enhanced. It is obvious that the presence of a strongly polarized 2DEG induces such strong modification. One can see in Fig. 2 that in

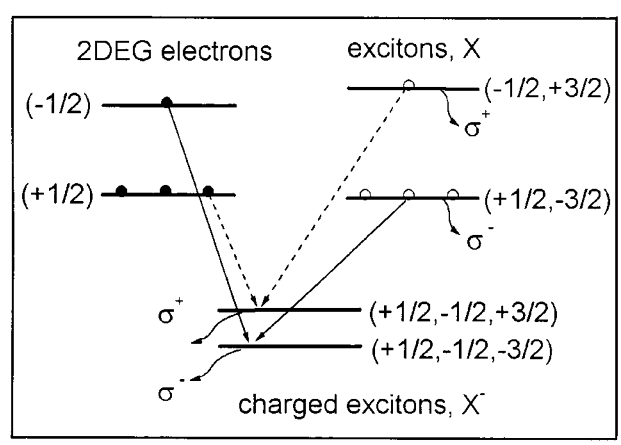

Figure 2. Schematical presentation of an X^- formation process in external magnetic fields from excitons and 2DEG electrons.

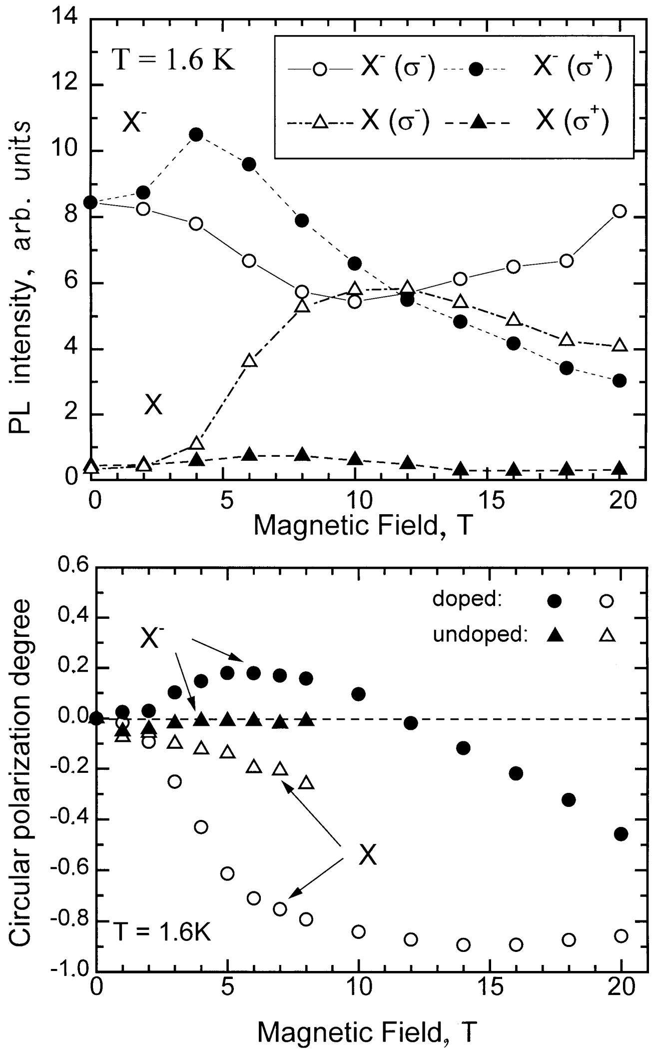

Figure 3. Magnetic field variation of the PL line intensities detected for different circular polarizations for an 8 nm-thick $CdTe/Cd_{0.7}Mg_{0.3}Te$ modulation-doped QW (upper panel). Magnetic-field-induced circular polarization degree of the exciton and negatively charged exciton PL lines with (circles) and without (triangles) modulation doping (lower pannel).

magnetic fields the X^- formation process is limited to the creation of a $(+1/2, -1/2, +3/2)$ state constructed from a $(+1/2)$ electron and a $(-1/2, +3/2)$ exciton. The $(+1/2, -1/2, -3/2)$ X^- state is populated by relaxation from the $(+1/2, -1/2, +3/2)$ state. In magnetic fields below 12 T these states are not splitted and the state which contribute to the σ^+ polarized transition is preferably populated. At higher fields, when the state contributing to the σ^- polarized transition becomes the lowest one, its thermal occupation leads to the rise of the negative polarization. The exciton polarization under these conditions is determined by the X^- formation process: $(-1/2, +3/2)$ excitons are washed out for X^-, but $(+1/2, -3/2)$ excitons are preserved as the $(-1/2)$ electron states are empty. As a result the exciton PL increases strongly for the σ^- polarized component (see Fig. 3, upper panel).

2.2. The effect of microwave radiation on the trion. The formation process of the trion is also reflected in the PL spectra which are taken with and without additional microwave illumination. These experiments have been performed on an 8 nm-thick QW structures with optical tuning of the 2DEG density [3,4]. At $B = 0$ T the microwaves decrease the X^- emission and increase the X emission, whereas at magnetic fields above 3 T the microwave radiation increase the X^- emission and decrease the X emission in σ^- polarization and have no obvious influence on X^- and X emission in σ^+ polarization.

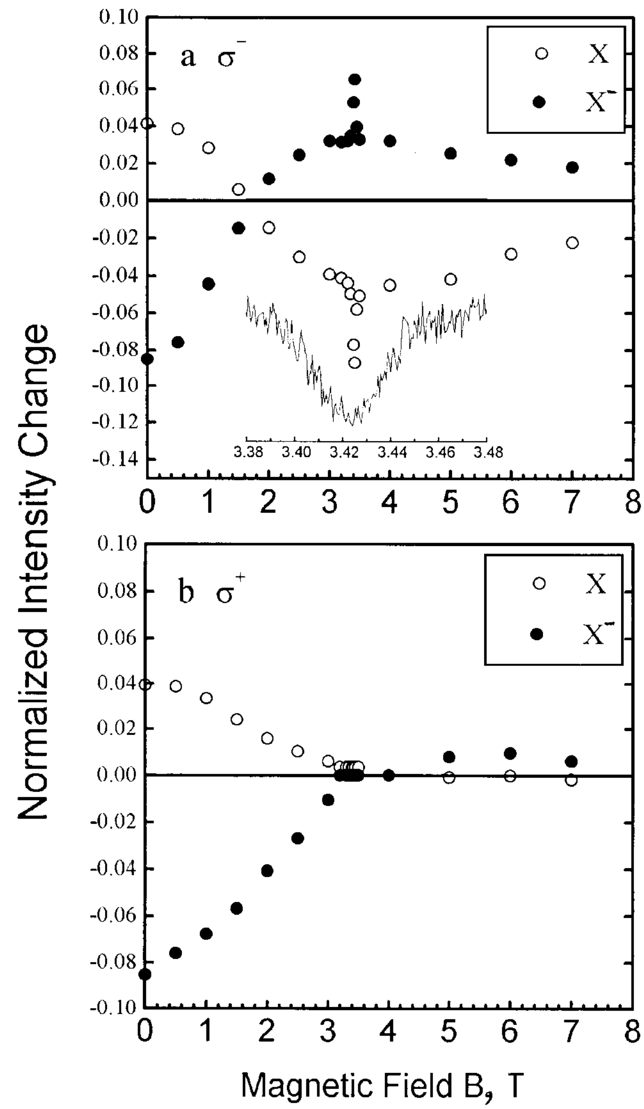

Figure 4. ODMR (70 GHz) signals detected from an 8 nm thick $CdTe/Cd_{0.7}Mg_{0.3}Te$ QW with optical tuning of 2DEG density on X emission and X^- emission for (a) σ^- and (b) σ^+ polarization. The changes are normalized by the respective total intensities of X and X^- emissions. In σ^- polarization sharp ODMR lines were observed on the broad ODMR background signals. In the inset the sharp electron ODMR (70 GHz) line detected on X emission in σ^- polarization is shown. The resonance position lies at $B_{res} = 3.424$ T with a linewidth $\Delta B = 39$ mT.

The ODMR (70 GHz) spectrum detected on X and X^- emission are shown in Fig. 4, *a* for σ^- polarization and Fig. 4, *b* for σ^+ polarization. The changes are normalized by their respective total intensities of X and X^- emission, and the positive and negative signs represent the increase and decrease of PL intensity, respectively. The maximal change is about 8% of the total intensities of X and X^- emission. With the increase of the magnetic field strength, the σ^- ODMR signals decline fast to zero and then change their signs, whereas the σ^+ ODMR signals decline slowly to zero. In σ^- polarization a sharp positive and negative ODMR line (at $B \approx 3.42$ T) appears respectively on the broad ODMR background signals detected on X^- and X emission. However, no sharp ODMR lines was observed in σ^+ polarization.

The details of the sharp ODMR lines are shown in the inset of Fig. 4, *a*. For 70 GHz microwaves the resonant lines lie at $B_{res} = 3.424$ T with the linewidth $\Delta B = 39$ mT. From the resonant magnetic field strength for different microwave frequencies $g^* = -1.461 \pm 0.002$ was obtained very precisely. We identify the sharp lines as the ODMR of the electrons of the 2D gas, whereas the broad background and the polarization dependence is determined by the formation process of X^-.

The scheme of the formation of X^- under magnetic field is shown in Fig. 2 where the g factor of the electron is negative and that of heavy hole is very small for low fields and positive for higher field strength. The formation and the recombination of X^- from the optically active exciton can be written as

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

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

Under the applied excitation conditions in this structure the electron density is estimated to be less than $4 \cdot 10^{10}$ cm^{-2}. Therefore, at the resonant magnetic field $B_{res} = 3.424$ T and $T = 1.6$ K all electrons lie in the $N = 0$ Landau level and most of them populate the lower spin state $(+1/2)$ by thermalization because of the long lifetime ($\tau = 2.7\,\mu$s) of the excess electrons [3]. In addition, the measured large exciton polarization (see Fig. 3, *b*) shows that most excitons are populating the (-1) state.

For the σ^- polarized luminescence of the trion, there is a small electron population in the $(-1/2)$ state and a strong exciton population in the (-1) state. Microwave resonant absorption increases the electron population in the $(-1/2)$ state and enhances the formation probability of the $(-3/2)$ X^-. Therefore, we observed a positive electron ODMR signal detected on the X^- emission and simultaneously a negative electron ODMR signal detected on the X emission since the enhanced formation of X^- is in expense of X.

In σ^+ polarization, there exists a strong electron population in the $(+1/2)$ state and a small exciton population in the $(+1)$ state. The formation probability of the $(+3/2)$ X^- therefore in not sensitive to changes of the $(+1/2)$ electron population. Thus, the sharp electron ODMR line can not be observed for the $(-3/2)$ X^- in σ^+ polarization although the microwave resonant absorption induces a decrease of the $(+1/2)$ electron population.

The microwave radiation pumps electrons from the low spin state $(+1/2)$ to the upper spin state $(-1/2)$ continuously until the electrons reach a new steady population. The change of the electron population by the magnetic resonant absorption results in the change of the formation probability of X^-. This is the reason why the magnetic resonance of electrons can be detected on the X^- or the X emission.

From the descussion above it is obvious that electron spin-dependent and the electron spin-conserving formation and recombination processes of X^- makes the electron ODMR detectable. This formation mechanism of X^- can be further supported by the broad ODMR background (at $B > 3$ T) shown in Fig. 4, *a, b*. Besides the microwave resonant absorption, the heating of electrons in the microwave field [10,11] also induced an increase of the $(-1/2)$ electron population and a decrease of the $(+1/2)$ electron population due to the raising of the excess electron temperature [12]. This enhances the formation probability of the $(-3/2)$ X^- just as the resonance case discussed above. The broad background signals (at $B > 3$ T) decrease with the magnetic field strength due to the suppression of the microwave heating by magnetic field [13].

We could say the above microwave heating effect is polarised since the background (at $B > 3$ T) occur only in σ^- polarization. At low magnetic fields ($B < 1$ T) where the two electron spin states have nearly the same population, another microwave heating effect occurs and we call it non-polarized because the background occur in both circular polarizations. The increase of the electron kinetic energy by microwave heating decreases the probability for an exciton trapping an electron. This is in accord with the temperature experiments on X^- [14]. So we observed a decrease of X^- emission and an increase of X emission in both circular polarization at $B < 1$ T shown in Fig. 4, *a, b*.

In σ^- polarization, the polarized effect of the microwave heating plays the main role at high magnetic fields ($B > 3$ T) and the non-polarized effect at low magnetic fields ($B < 1$ T). For $1 < B < 3$ T, the two effects compete against each other, so the ODMR background signals decline fast to zero and change their signs with the increase of magnetic field strength. In σ^+ polarization, only the non-polarized effect plays a role and the ODMR background signals decline slowly to zero due to the suppression of the microwave heating. Since the polarized ODMR background (at $B > 3$ T) is due to the microwave heating of excess electrons and electrons which are bound to impurities can not be heated by microwaves, the ODMR measurements prove clearly the attribution of the PL line to X^- and not to an impurity bound exciton.

2.3. Trions formed with the light hole exciton. A fingerprint of X^- is the strong polarization of its resonance in external magnetic fields. When the excess electrons are strongly polarized by the external magnetic field, excitation of the X^- singlet state is allowed

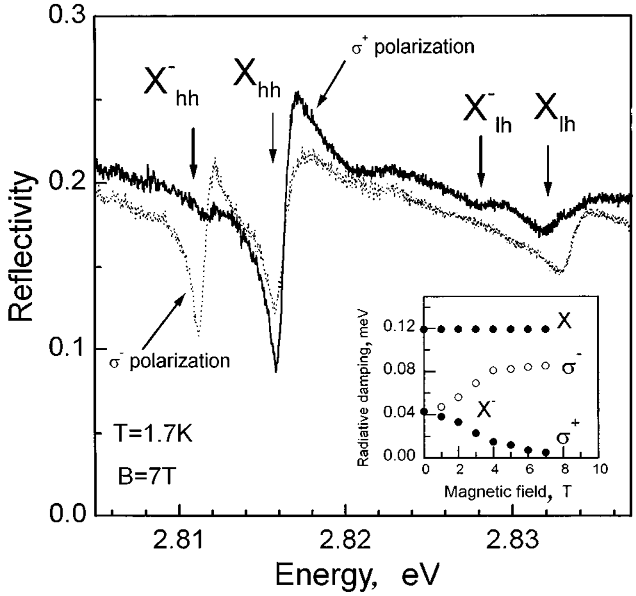

Figure 5. Reflectivity spectra of a 10 nm $ZnSe/Zn_{0.89}Mg_{0.11}S_{0.18}Se_{0.82}$ quantum well at 1.6 K and 7 T for σ^+ (solid) and σ^- (dashed) lines. The insert shows the radiative damping (oscillator strength) for the exciton and the trion.

for one defined polarization only. In contrast to the CdTe/(Cd,Mg)Te structures discussed so far the g factor of the electron in ZnSe/(Zn,Mg)(S,Se) QWs is positive ($g_e = 1.1$). Therefore, the allowed transition for X^- formed with the heavy-hole exciton in these structures is the σ^- polarization, whereas σ^+ is the allowed polarization for the trion formed with the light hole exciton. Such a behavior is demonstrated for a 10 nm $ZnSe/Zn_{0.89}Mg_{0.11}S_{0.18}Se_{0.82}$ QW with $n_e \approx 4 \cdot 10^{10}\,\mathrm{cm}^{-2}$ in Fig. 5. The resonance indeed appear in opposite circular polarization of the reflected light. Furthermore, from Fig. 5 the binding energy of the trion can be determined. At $B = 7$ T the binding energy of X^- formed with the heavy hole exciton is 4.6 meV, and 3.8 meV for X^- formed with the light hole exciton.

We have fitted all experimentally observed resonances in the frame of a model of a nonlocal dielectric response [15]. In the insert of Fig. 5 we have plotted the dependencies of the exciton and trion radiative damping as a function of the magnetic field. It is obvious that the exciton radiative damping constant Γ_0 (i. e. the exciton oscillator strength) shows no dependence on the magnetic field strength for both circular polarizations. The trion oscillator strength, in contrary to the excitonic one, decrease with the magnetic field for the σ^+ polarization and increases for the σ^- polarization. This different behavior of Γ_0 for σ^+ and σ^- polarizations is due to the singlet structure of the trion ground state, where the two electrons involved have opposite spins. In the presence of a magnetic field the background electrons are polarized and the trions could be created by photons of one polarization only.

3. Combined Exciton-Cyclotron Resonance

In external magnetic fields, besides the trion line, another new line appears in the PLE and reflectivity spectra, which can not be attributed to the normal magneto-exciton peaks. This line is attributed to a combined exciton-cyclotron resonance (ExCR) and is a supplementary example for new features observable in semiconductor quantum wells containing an electron gas of low density [3]. An incident photon creates an exciton in the ground state and simultaneously excites one of the resident electrons from the lowest to first (ExCR1) or to the second (ExCR2) Landau level. As can be seen from Fig. 6 the energetic positions of these ExCR lines are in the range of the Coulomb bound states, but they behave very differently. For instance, the intensity of the ExCR line increase strongly for larger n_e, whereas the magnetoexciton lines are insensitive to this parameter. Furthermore, Fig. 6 shows that the ExCR lines shift linearly in magnetic fields with a slope of 1.2 meV/T (ExCR1) or 2.9 meV/T (ExCR2), respectively, which is comparable to the electron cyclotron energies in CdTe/(Cd,Mg)Te QWs. An extrapolation of these shifts to zero field meets approximately the energy of the 1s state of the heavy-hole exciton ($1s{-}hh$). This linear shift of the ExCR lines, as opposed to the quadratic one of the heavy-hole excitons ($1s$, $2s$, and $3s$ states also displayed in Fig. 6), shows that the free electrons contribute to the observed process. Theoretically the shift of the ExCR lines in an external magnetic field behave like $N \cdot \hbar\omega_{c,e}(1+m_e/M)$, where m_e is the electron and $M = m_e + m_h$ is the exciton

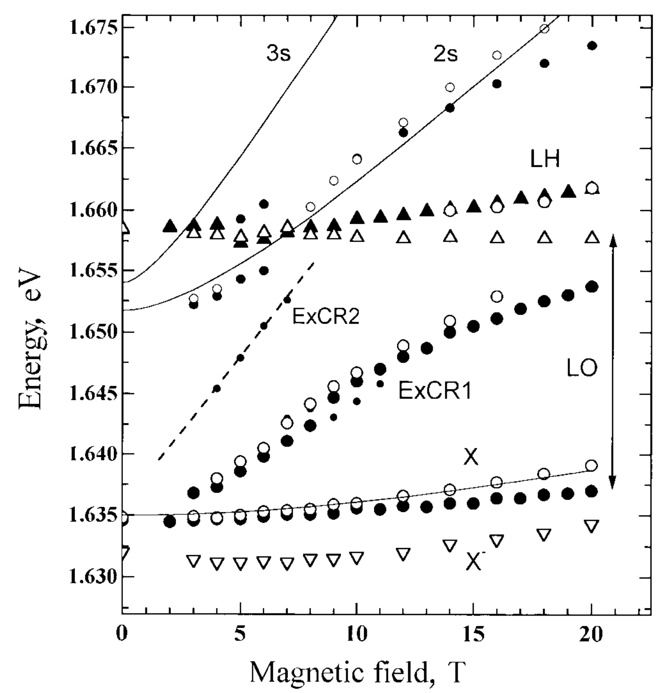

Figure 6. Fan chart of an 8 nm $CdTe/Cd_{0.7}Mg_{0.3}Te$ quantum well. Open symbols represent σ^+ and closed symbols σ^- polarization respectively. The lines represent calculation for the exciton 1 s, 2 s and 3 state applying the model described in ref. [18].

mass, $\hbar\omega_{c,e}$ is the cyclotron energy and N an integer (details of the theoretical consideration are in ref. [3]). Applying the experimentally obtained mass values $m_e = 0.11m_0$ and $m_{hh} = 0.48m_0$ gives 1.24 meV/T very close to the experimental value.

The ExCR line is strongly σ^- polarized, when the spin of the free electron gas is parallel to the free electron gas polarization in the external field direction. As a σ^- photon creates an electron with the same spin orientation as the background electron. In this case the exciton angular momentum in the final state is (-1), and the recombination of such a photon is dipole allowed. On the other hand, a σ^+ polarized photon leads to a final state exciton with a magnetic moment of 2, whose recombination is dipole forbidden. Such excitons can recombine only after a spin flip of either the electron or the hole caused by scattering processes, and therefore their contribution to the PL intensity is much weaker.

In addition, it can be seen in Fig. 6 that for magnetic field above 11 T the ExCR1 line exhibits a strong bowing and splitting into two components. In this field range the *LO*-phonon energy equals the cyclotron energy resulting in a resonant polaron coupling [16], which can be established from the typical anticrossing behavior caused by the mixing of electron and phonon states. The observation of a resonant polaron coupling via the ExCR line also evidences the participation of 2*D* electrons in the ExCR process.

4. Additional Features Correlated with the 2DEG

Besides the linearly blue shift of the ExCR line observed in PLE and reflectivity the PL spectra of QWs with an electron gas of low density exhibit lines that are correlated with shake-up processes [17]. We observe a series of low energy satellites related to the excitation of the 2DEG [5]. In emission the transition energy of the photon is lowered by energy conservation. The shake-up process excites, similar to the ExCR mechanism, inter Landau level transitions giving rise to a red shifted PL lines with an energy separation from the trion energy of about $-N\hbar\omega_{c,e}$.

Besides all of these above discussed elastic scattering processes we would like to mention inelastic and spin-dependent scattering processes, where the photo-generated excitons loses their energy by scattering to an ortho-exciton state and the simultaneous excitation to an upper Zeeman sublevel. Details of these scattering processes are published elsewhere [7].

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