

Photoluminescence of “dark” excitons in CdMnTe quantum well, embedded in a microcavity

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Abstract

A diluted magnetic semiconductor (DMS) quantum well (QW) microcavity operating in the limit of the strong coupling regime is studied by magnetooptical experiments. The interest of DMS QW relies on the possibility to vary the excitonic resonance over a wide range of energies by applying an external magnetic field, typically about 30 meV for 5 T in our sample. In particular, the anticrossing between the QW exciton and the cavity mode can be tuned by the external field. We observe the anticrossing and formation of exciton polaritons in magneto-reflectivity experiments. In contrast, magneto-luminescence exhibits purely excitonic character. Under resonant excitation conditions an additional emission line is observed at the energy of the dark exciton. The creation of dark excitons is made possible due to heavy hole–light hole mixing in the QW. The emission at this energy could be due to a combined spin flip of an electron and a bright exciton recombination.

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1. Introduction

The discovery of the giant enhancement of magnetooptical effects in CdMnTe has boosted the research directed towards the understanding of the physics underlying the unusual phenomena

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associated with diluted magnetic semiconductors [1]. The development of the molecular beam epitaxy made possible to study not only the bulk DMS material but also quantum structures. In particular, numerous studies of DMS quantum wells (QW) [2], microcavities [3,4], quantum dots [5] and nanocrystals [6] can be found in the literature. Despite this huge amount of research, CdMnTe QW still reserve surprising phenomena to be discovered.

In this work we study the photoluminescence of $\text{Cd}_{0.95}\text{Mn}_{0.05}\text{Te}$ QW embedded in a $\text{Cd}_{0.4}\text{Mg}_{0.6}\text{Te}$ microcavity. The microcavity is designed to operate in the strong coupling regime, which means that the interaction of excitons and photons in the cavity results in the formation of complex quasiparticles, called exciton–polaritons [7]. In the reflectivity spectra, the formation of polaritons manifests itself by the anticrossing between exciton-like and photon-like polariton branches at the resonance energy [8]. The energy splitting between the two polariton modes is called Rabi energy, in our sample $E_R = 6$ meV. However, in the photoluminescence experiment on the same sample, the presence of polaritons can not be detected, because the PL is mainly due to the localized exciton states while the polariton formation is mainly due to free excitons in the QW. We are concerned here by the exciton PL under magnetic field applied in Faraday geometry. Making use of the giant Zeeman effect in DMS, we can easily control the QW exciton energy by the external magnetic field. In our sample the PL intensity is strongly increased when the exciton energy is approximately one LO phonon below the excitation energy. This general enhancement of the PL intensity allows, in the cross-polarized geometry, to observe an additional peak of the PL 10 meV above the ground exciton state. We tentatively associate the high energy emission with the exciton recombination accompanied by the simultaneous spin flip of an electron from excited to ground state.

2. Experiment

The sample under study was grown by molecular beam epitaxy on a $\text{Cd}_{0.88}\text{Zn}_{0.12}\text{Te}$ [100] oriented substrate. The back and front Bragg mirrors are formed by 20 and 6.5 pairs of $\lambda/4$ -thick $\text{Cd}_{0.4}\text{Mg}_{0.6}\text{Te}/\text{Cd}_{0.75}\text{Mn}_{0.25}\text{Te}$, respectively. The $\text{Cd}_{0.95}\text{Mn}_{0.05}\text{Te}$ QW of 8 nm width is embedded in the middle of $\lambda/2$ $\text{Cd}_{0.4}\text{Mg}_{0.6}\text{Te}$ cavity. The experiments below were done using a spectrograph coupled to a streak camera which allows to obtain both temporal (10 ps) and spectral (0.25 meV) resolution of the signal. A pair of $\lambda/4$ plates is used in the set up. One is used to circularly polarize the Ti-sapphire laser light, the other to resolve the PL polarization. The sample is mounted in the magneto-optical cryostat where its temperature is kept at 2 K.

Fig. 1 shows the normalized polarization resolved PL spectra measured under magnetic field up to 5 T. The position of the spot in the plane of the sample is such that the photon mode energy $E_{\text{ph}} = 1.707$ eV, as measured from the reflectivity spectra [8]. The excitation energy is $E_{\text{exc}} = 1.7125$ eV for $B = 0, 0.5$ T and $E_{\text{exc}} = 1.7184$ eV for higher field experiments. Four different excitation/detection polarization configurations are shown, σ_+/σ_+ , σ_+/σ_- , σ_-/σ_+ , σ_-/σ_- . In σ_-/σ_- configuration the signal could be detected only at low fields. In this configuration as well as in both σ_+/σ_+ and σ_+/σ_- the excitonic emission shows up at exactly the same energy at a given magnetic field and only one PL peak is detected. In contrast, in σ_-/σ_+ geometry an additional emission peak is observed at higher energy under magnetic fields above 2 T. This emission is our main concern in this paper.

Let us mention the strongly nonmonotonic dependence of the total PL intensity on the magnetic field. In the Fig. 2 one can see the PL peaks intensity extracted from Fig. 1 for the four different excitation/detection polarization geometries as a function of the external magnetic field. Overall, the intensity of the emission in all the configurations increases by a factor of 20

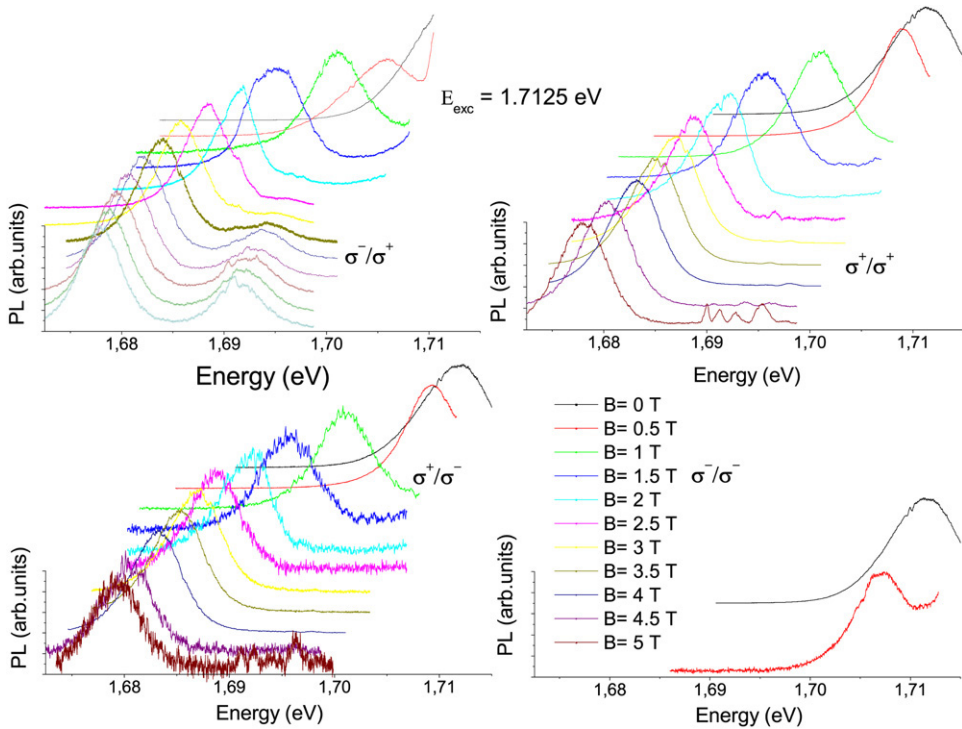


Fig. 1. Normalized photoluminescence spectra in four different circularly polarized excitation/detection geometries.

under magnetic field about 3.5–4 T, and then decreases at higher fields. Such behaviour could result from the interaction of the exciton with the cavity mode. However, the analysis of the PL spectra as a function of in-plane coordinate of the spot (data not shown) as well as the reflectivity experiments where the photon mode is clearly visible, both suggest the presence of another source of the resonance enhancement of the signal.

The symbols in the Fig. 3 show the energies of the two PL peaks which appear in the σ_-/σ_+ configuration. The four solid lines represent the calculated heavy exciton energy levels in the QW taking into account carrier–Mn exchange interaction in both conduction and valence band. The parameters used in the standard Luttinger Hamiltonian are $\gamma_1 = 5.4/m_0$ and $\gamma_2 = -1.8/m_0$, where m_0 is the free electron mass. The exchange integrals for the electron and hole in the CdMnTe are well known [9], the effective Mn concentration in the QW with 5% of Mn ions is $x_{\text{eff}} = 0.03$ [10], the effective temperature $T = 4$ K was the only fitting parameter. The last figure (Fig. 4) illustrates the effect of the excitation energy on the relative PL intensity of the two peaks observed in σ_-/σ_+ configuration. Both the set of the PL spectra for different excitation energies and the intensity of the peaks as a function of the excitation energy are shown. Finally, the time-resolved measurements indicate that the PL lifetimes associated with the two emission peaks are very similar, about 120 ps.

3. Discussion

Let us start from considering the relative intensity of the PL in different configurations. Under σ^+ polarized excitation most of the light is absorbed and reemitted by the fundamental exciton

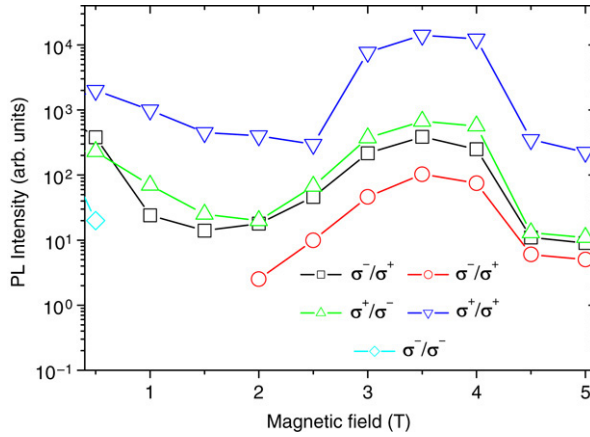


Fig. 2. Intensity of the PL peaks measured for different polarization orientations shown in Fig. 1. Squares and circles stand for the two peaks in the σ^-/σ^+ excitation/detection geometry, diamonds and up- and down triangles stand for σ^-/σ^- , σ^+/σ^- and σ^+/σ^+ , respectively.

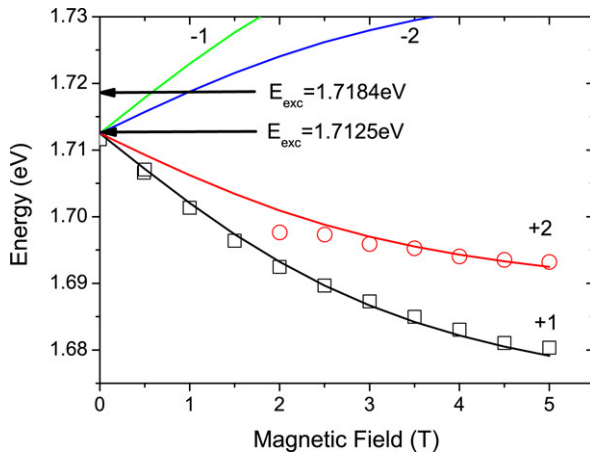


Fig. 3. Energies of the two PL peaks measured in σ^-/σ^+ configuration as a function of the magnetic field in Faraday geometry (symbols). Solid lines are exciton energy levels calculated using the parameters given in the text. The arrow points to the excitation energy.

state with total angular momentum $J_z = +1$, because the excitation energy is chosen below the $J_z = -1$ state (see Fig. 3). A small fraction (5%) is emitted in σ^- polarization (*cf* down and up triangles in Fig. 2). This suggests that the exciton angular momentum is not strictly defined in our experiment, e.g. due to disorder effects in the QW the heavy and light hole states may be mixed. As a result σ^- polarized light can be partly (5%) absorbed by the excitonic states with $J_z > 0$, while no light is absorbed by the $J_z < 0$ states which are above the laser excitation energy. The PL in this case is almost 100% σ^+ polarized. Indeed, as one can see in Fig. 2 in the σ^-/σ^- configuration the same signal shows up on only at 0.5 T.

The main question that arises from these experiments is the origin of the second emission line in the σ^-/σ^+ configuration. The energy of this resonance is exactly that of the dark exciton state with $J_z = +2$. This is shown in Fig. 3, note that there is no fitting parameters to adjust the relative

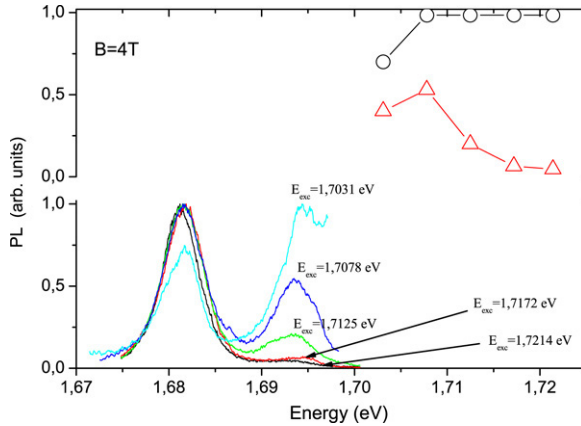


Fig. 4. PL spectra at $B = 4$ T in σ^-/σ^+ configuration measured with different excitation energies (solid lines). Triangles (circles) show the high(low) energy luminescence peak intensity as a function of the excitation energy.

position of the $J_z = +2$ and $J_z = +1$ exciton states. However, similar characteristic times for these two transitions incite us to interpret this line in a different way. One possibility could be the combined exciton–spin flip (XSF) resonance, in analogy with the exciton–cyclotron (XC) resonance in the QWs discovered by Yakovlev and colleagues [11]. In our case the interaction between the excitons may lead to the simultaneous recombination of the $J_z = +1$ exciton and a spin flip of an electron from the spin state $S_z = +1/2$ to the state $S_z = -1/2$. A photon emitted in this process has the energy of the dark exciton state. Such interpretation is corroborated by the PL excitation experiments (Fig. 4), where the intensity of the exciton–spin flip resonance increases under resonant excitation. However, there is an important difference with the mechanism discussed in Ref. [11], because in contrast with XC, XSF resonance is not a spin conserving process.

The last point of the discussion is the enhancement of PL in all polarization configurations under magnetic field about 3.5 T. As mentioned above, it is not related with microcavity structure. For the point on the sample where the experiments were done the photon mode energy is $E_{\text{ph}} = 1.707$ eV, therefore the resonance should be achieved at about 0.5 T, and not at 4 T where the PL is greatly increased. Thus we definitively exclude the hypothesis of the cavity induced resonance from consideration and suppose that it is related with the QW. Indeed, in CdMnTe the LO-phonon energy is about 23 meV [12]. Similar energy separates the PL excitation energy and the emission at 4 T. In this case we can tentatively interpret the enhancement of the luminescence to be due to momentum and energy relaxation by emission of the LO phonons.

In conclusion, we have studied CdMnTe QW embedded in a microcavity. The strong coupling which is clearly visible in magnetorelectivity does not manifest itself in the PL experiments. This is because the PL is essentially due to the localized excitonic states. Indeed, their interaction with the photon mode in the cavity is weaker than the interaction between the free exciton and the photon. In the reflectivity experiments, the latter interaction is probed, while in PL the former one. Therefore, the emission of the “dark” state can be considered as a QW related phenomenon. We suggest that this emission results from the combined exciton–spin flip resonance. This phenomenon could be observed due to the resonant enhancement of the PL under magnetic fields such that the exciton energy in the QW is about 20 meV below the laser energy. We argue that it may be due to exciton energy and momentum relaxation by the LO phonons.

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