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Magnetic polarons in a single diluted magnetic semiconductor quantum dot

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Quasi-zero-dimensional magnetic polarons in single $Cd_{0.93}Mn_{0.07}Te/Cd_{0.6}Mg_{0.4}Te$ quantum dots have been studied by photoluminescence spectroscopy. By comparing the experimental data with model calculations, the energy, the internal magnetic field, and the volume of the magnetic polarons are obtained. Moreover, the magnetic environment of the recombining electron hole pair causes a distinct broadening of the emission line (~ 4 meV) of one diluted magnetic single quantum dot. The alignment of the Mn-spins in high magnetic fields results in a linewidth narrowing of almost one order of magnitude and the linewidth becomes comparable to that of a nonmagnetic $Cd_{0.93}Mg_{0.07}Te/Cd_{0.6}Mg_{0.4}Te$ reference sample.

A variety of electronic or magnetic properties of a solid can be artificially tailored by including magnetic ions in a semiconductor crystal matrix. This has been shown, e.g., in diluted magnetic semiconductors (DMSs), where the strong *sp-d* exchange interaction between the charge carriers and the magnetic Mn^{2+} ions results in a giant Zeeman splitting of the valence- and conduction-band states, a large Faraday rotation, and the formation of magnetic polarons (MPs).[1–3] The MP is a small region of the crystal with strongly correlated spins of localized carriers and magnetic ions. The spin ordering decreases the carrier energy and the optical properties of such MPs have been widely investigated in three-dimensional (3D) and 2D DMS.[1,3–6] In a DMS quantum dot (QD) the MP formation should be much more pronounced due to the strong, three-dimensional confinement of the charge carriers.[7] However, investigations on Mn-containing QDs are quite scarce up to now and to our knowledge limited to QD ensembles.[8,9] In order to get quantitative access to zero-dimensional MPs, inhomogeneous broadening effects due to size and/or composition fluctuations have to be avoided completely, i.e., experiments on a single DMS QD are required. Whereas a variety of experimental techniques with a high spatial resolution has been developed to select single QDs,[10–12] these approaches have only been applied to nonmagnetic semiconductor nanostructures up to now.

In this paper we present photoluminescence (PL) studies on single QDs in the $Cd_{0.93}Mn_{0.07}Te/Cd_{0.6}Mg_{0.4}Te$ system in direct comparison to its electronic, but nonmagnetic, analog $Cd_{0.93}Mg_{0.07}Te/Cd_{0.6}Mg_{0.4}Te$. This allows a detailed study of the interaction of single electron-hole pair with its magnetic environment. Special emphasis will be laid on the MP formation and on the emission linewidth in DMS SQDs. Besides a detailed investigation of (i) the temperature and magnetic field dependence of the ground MP state energy and (ii) the degree of circular polarization of its emission at weak magnetic fields, we have been able to resolve the emission line from the first excited (*metastable*) MP state and determine its energy. A quantitative description of the experimental data allows an extraction of MP parameters like the effective internal exchange magnetic field B_{MP} and therefore the MP localization radius in the SQD.

The structures consist of three monolayers of $Cd_{0.93}Mn_{0.07}Te$ (or $Cd_{0.93}Mg_{0.07}Te$, respectively) embedded in nonmagnetic $Cd_{0.6}Mg_{0.4}Te$ barriers grown by molecular beam epitaxy. So-called ''natural'' QDs are expected to form due to spatial variations of size and composition.[11,13] To achieve the high spatial resolution required for measuring SQDs, Al masks with apertures down to 150 nm in diameter have been prepared by electron beam lithography and lift-off technique.[11] The PL spectra from such small areas usually consist of individual lines (typically between 10 and 20) of SQDs distributed over a spectral range of about 100 meV. For detailed investigations we have studied SQDs with PL lines energetically well separated from the others. Magneto-PL studies were carried out in an optical cryostat with a split-coil solenoid in Faraday configuration at $T = 2$ K. An optical cryostat with a highly accurate ($\lesssim 0.1$ K) temperature control in the range of $T = 2–300$ K was used to study the temperature behavior of the PL spectra. The samples were excited by the 514.5 nm line of an Ar^+ laser and the PL signal was detected with a liquid-nitrogen-cooled charge-coupled device (CCD) camera. To avoid the heating of Mn spin subsystem, low excitation power densities of about 2 W/cm^2 have been used.[14] Increasing the excitation density by a factor of 3 does not affect the PL energy within our experimental resolution of 0.1 meV, while for higher power, a clear blueshift of the signal is observed, mainly attributed to the heating of the spin subsystem.

Figure 1 compares typical PL spectra at 2 K in σ^+ polarization from nonmagnetic $Cd_{0.93}Mg_{0.07}Te$ and DMS $Cd_{0.93}Mn_{0.07}Te$ SQDs at different external magnetic fields B_{ext}. The PL spectrum of the nonmagnetic SQD consists of a narrow line with a halfwidth $\Gamma \lesssim 0.5$ meV, which is limited by the spectral resolution of our system. With increasing B_{ext}, the line undergoes a small redshift of up to ~ 0.4 meV at $B_{ext} = 8$ T while no change of the linewidth is obtained. In contrast, the behavior of the DMS SQD is quite different. First, the line shows a rather large redshift characteristic for

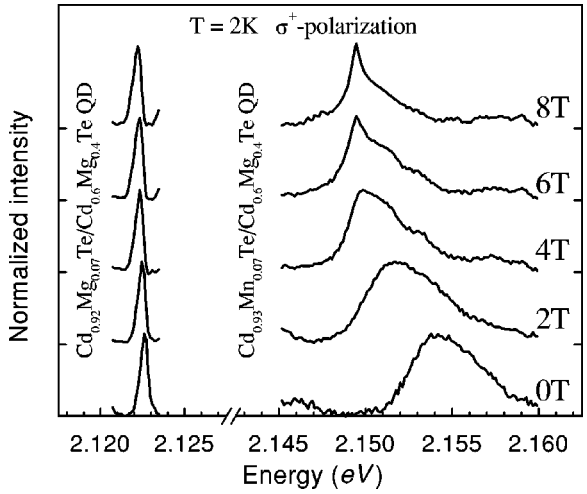

FIG. 1. PL spectra of a DMS $Cd_{0.93}Mn_{0.07}Te/Cd_{0.6}Mg_{0.4}Te$ single QD (right) and a nonmagnetic $Cd_{0.93}Mg_{0.07}Te/Cd_{0.6}Mg_{0.4}Te$ single QD (left) in σ^+ polarization at different magnetic fields.

DMS (~ 5 meV at $B_{ext}=8$ T). Second, and even more important, the emission line is rather broad ($\Gamma \approx 4$ meV) at low B_{ext} and decreases strongly by applying a magnetic field, being comparable to that in nonmagnetic QDs at $B_{ext}=8$ T. This is quite surprising and is a direct consequence of the magnetic environment of the recombining electron-hole pair in $Cd_{0.93}Mn_{0.07}Te$ SQDs, as will be shown in the following.

In Fig. 2, the magnetic field dependence of the $Cd_{0.93}Mn_{0.07}Te$ SQD PL peak energy ($\hbar\omega_{PL}^{\pm}$) is shown for σ^+ and σ^- polarization, respectively. Due to the very strong PL polarization, the σ^- line could only be measured in rather weak (<1 T) fields. The energy of the σ^+ peak decreases with magnetic field and saturates at $B_{ext} \gtrsim 5$ T. In order to explain the magnetic field dependent PL spectra of DMS SQDs, the formation of MPs due to an alignment of magnetic ion spins in the strong exchange field of the band carriers has to be taken into account. In $Cd_{1-x}Mn_xTe$ the

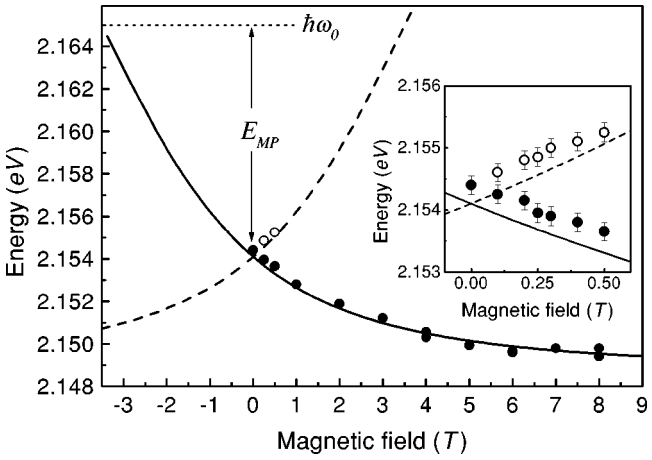

FIG. 2. Measured (symbols) and calculated (lines) magnetic field dependences of the PL transition energies in a $Cd_{0.93}Mn_{0.07}Te/Cd_{0.6}Mg_{0.4}Te$ single QD for σ^+ (solid symbols and solid line) and σ^- polarization (open symbols and dashed line). In the inset, a magnified representation in the range of low magnetic fields is given.

exchange interaction constant β for holes is four times larger than for electrons (α) and therefore the holes play the main role in the interaction of excitons with Mn ions.[15] The hole induced exchange field inside a MP, B_{MP}, is proportional to the squared wave function $|\Psi(\vec{r})|^2$ and the relation between B_{MP} and the localization volume of the hole, V, can be written as[16]

$$B_{MP} \approx \frac{\gamma}{3\mu g_{Mn}} \beta J \frac{1}{V}, \tag{1}$$

where $J=3/2$ is the hole spin, μ the Bohr magneton, and $g_{Mn}=2$ is the g factor of the Mn^{2+} ion. The parameter γ takes into account that only a part of the hole wave function is inside the DMS QD. For the investigated 3 ML thick $Cd_{1-x}Mn_xTe$ QDs, where the extension of the QD in growth z direction is much smaller than in lateral direction, a value of $\gamma = 0.5 \pm 0.05$ was estimated.

The formation of exciton MPs includes several important relaxation processes.[17,18] The first, very fast, process is the relaxation of the exciton spin resulting in a strong correlation between its direction and the direction of the instantaneous total magnetic moment $\vec{M}$ of the Mn^{2+} ions in the QD. As a result, the exciton spin will be aligned along the z component of $\vec{M}$ because the exchange energy for this state is minimal. The second stage is the alignment of the Mn^{2+} ion spins in the direction of the hole spin. This relaxation process leads to a further decrease of the exciton energy by the formation of MPs with an energy $E_{MP} \approx V\chi B_{MP}^2$ (χ is the magnetic susceptibility). The last relaxation stage should result in an equilibrium distribution of the MP moments. It has a very long characteristic time constant and is usually interrupted by the recombination process.[17,19]

To describe the Zeeman splitting E_Z of the heavy-hole excitonic states in the DMS, a modified Brillouin function $\mathcal{B}_{5/2}(B,T)$ is applied:[15]

$$E_Z = E_{Z0}\mathcal{B}_{5/2}\left\{\frac{5\mu g_{Mn}B}{2k(T+T_0)}\right\}, \tag{2}$$

where $E_{Z0}=f(\alpha-\beta)\gamma N_0 x S_0$, N_0 is the number of units cells per unit volume, $N_0\alpha = 220$ meV and $N_0\beta = -880$ meV. The phenomenological parameters $S_0 < 5/2$ (Mn ion spin) and $T_0 > 0$ take into account the antiferromagnetic interaction of neighboring Mn ions and for $Cd_{0.93}Mn_{0.07}Te$, values of $S_0 \approx 1.32$ and $T_0 \approx 3.1$ K can be taken from the literature.[22] The coefficient f takes into account the influence of carrier confinement on the exchange parameters in DMS and is usually $\lesssim 1$.[6,23]

The PL peak energy ($\hbar\omega_{PL}$) in DMS QDs depends on the ratio τ_f/τ_r. For similar structures as discussed here, a characteristic MP formation time $\tau_f \approx 150$ ps is obtained, which is roughly a factor of 2 smaller than the recombination lifetime $\tau_r \approx 300$ ps.[20,21] Assuming simple exponential laws for the change of both the exciton MP energy and the emission intensity with time it can be shown, that for $\tau_r \approx 2\tau_f$ the maximum of the emission line differs from the equilibrium MP energy by less than $0.05E_{MP}$. This allows us to use the equilibrium limit as a first approximation to extract the MP parameters. In this case the PL peak energy in DMS QDs can be described by the following expression:

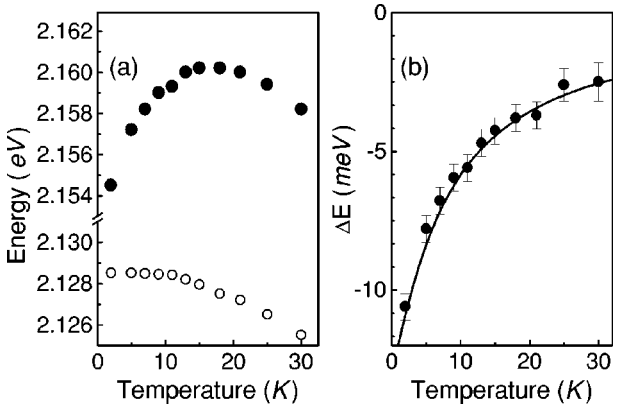

FIG. 3. (a) Temperature dependence of the PL energy for nonmagnetic (open) and DMS (solid circles) single QDs in σ^+ polarization. (b) Experimental (symbols) and calculated (line) difference of the emission energy of a nonmagnetic and DMS single QD vs temperature.

$$\hbar\omega_{PL}^{\pm} = \hbar\omega_0 \mp \tfrac{1}{2}E_Z(B,T), \quad B = B_{MP} \pm B_{ext}, \tag{3}$$

for σ^+ and σ^- polarization, respectively. For σ^+ polarization the field B in (3) is equal to the sum of B_{ext} and B_{MP}. The emission in σ^- polarization originates from the metastable state with the initial MP magnetic moment opposite to the external magnetic field resulting in $B = B_{MP} - B_{ext}$.

As indicated in Fig. 2 by solid (σ^+) and dashed (σ^-) lines, a good description of the experimental dependences is found. We obtain an internal exchange field $B_{MP} \approx 3.5$ T and $fx \approx 0.045$. As the average Mn content in our sample is $x = 0.07$, the difference may be due to a decreased f in QDs[6,23] and/or a smaller concentration of Mn ions in the particular QD under investigation.

Using the value of $B_{MP} = 3.5$ T, an exciton localization volume $V \approx 540 N_0^{-1}$ can be estimated from expression (1). Taking the nominal width of the $Cd_{0.93}Mn_{0.07}Te$ layer (10 Å) as a first approximation of the QD height, this corresponds to an in-plane MP radius of about 30 Å.

The formation of zero-dimensional MPs is further supported by looking at the temperature dependence of the PL peak energy in DMS and nonmagnetic QDs as shown in Fig. 3(a). As expected, the $Cd_{0.93}Mg_{0.07}Te$ QD PL line shows a monotonic shift to smaller energies according to the temperature induced band gap shrinkage. In contrast, the energy of the PL line in the DMS SQD first increases with T (for $T \lesssim 15$ K), which can be attributed to the MP suppression, and only at $T \gtrsim 20$ K the influence of the band gap shrinkage dominates. By subtracting the PL shift of the DMS and the nonmagnetic SQD lines [see symbols in Fig. 3(b)], we can extract $\Delta E(T)$ at $B_{ext} = 0$, which directly reflects the change of the *magnetic* localization within the MP with temperature. This temperature dependence is described by Eqs. (2) and (3) without any additional fitting parameters, i.e., $B_{MP} \approx 3.5$ T and $fx \approx 0.045$ have been used as determined from the fitting of $\hbar\omega_{PL}^{\pm}(B)$. The excellent agreement between experiment and calculations strongly supports the validity of the equilibrium model used. Moreover, a low temperature MP energy of $E_{MP} \approx 10.5$ meV can be directly extracted from the data.

As outlined in Refs. 17 and 18, the degree of circular polarization of the PL peak, $\rho = (I_+ - I_-)/(I_+ + I_-)$, is controlled by the average value of the squared projection of Mn^{2+} ion magnetic moment fluctuations, $\langle \Delta M^2 \rangle$, on the hole spin direction. $\langle \Delta M^2 \rangle$ can be expressed by experimentally measurable values using the well-known result of the fluctuation-dissipation theorem.[24] Such a treatment was done for the 3D and the 2D case (i.e., for isotropic as well as strongly anisotropic hole g factors) by Merkulov *et al.*,[18] leading to a relation between the slope of the degree of circular polarization in small B_{ext} and the exciton MP parameters[16]

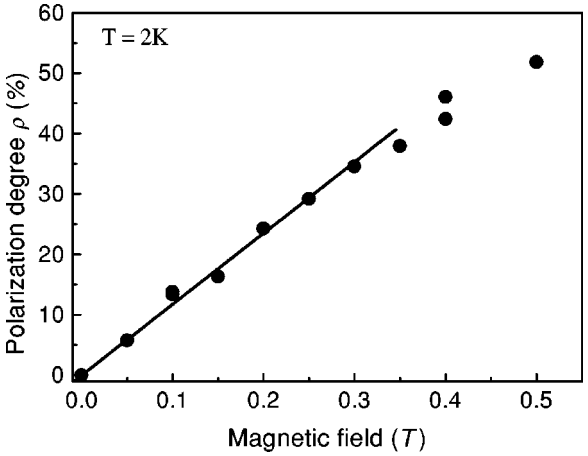

FIG. 4. Measured (symbols) and calculated (line) degree of circular polarization in DMS single QDs at weak magnetic fields.

$$V = \frac{\pi kT}{2\chi}\left(\left.\frac{d\rho(B)}{dB}\right|_{B\approx 0}\right)^2. \tag{4}$$

Figure 4 shows, that this dependence describes the experimental data without any additional fitting parameter quite well, giving one more strong confirmation of the validity of the model used and the parameters found for the exciton MP.

Finally we return to the most striking experimental result, i.e., the large width of the exciton MP emission line and its dependence on magnetic field in DMS SQD (Fig. 1). One obvious broadening mechanism is related to the kinetics of MP formation, i.e., the transient shift of the emission line within the MP formation time. In the case of time-integrated PL measurements, this results in an increase of the PL linewidth due to the overlap of different spectral positions during one period of data acquisition. It was indicated above that for $\tau_r \approx 2\tau_f$ the maximum of the emission line differs from the equilibrium MP energy by less than $0.05E_{MP}$, justifying the usage of Eqs. (2) and (3) for the description of the temperature and magnetic field dependence of the PL energy. In contrast, however, the linewidth Γ is much more sensitive to τ_f/τ_r. In fact, using simple exponential laws for the change of the exciton MP energy and the PL intensity (see above) a linewidth of about ~ 4 meV at $B = 0$ is expected for $\tau_r/\tau_f \approx 2$, in good agreement with our experimental data. It should be noted, however, that the impact of statistical fluctuations of the magnetization[25] and the influence of memory effects, i.e., an incomplete magnetic relaxation, on the emission linewidth cannot be ruled out completely, as the SQD is being probed repeatedly. Further experiments like, e.g., time-resolved studies on a SQD, which will certainly be an experimental challenge, are required to clarify this point.

At high magnetic fields, both the formation of MPs and the statistical magnetic fluctuations are suppressed due to the magnetic field induced alignment of Mn^{2+} ion spins in the external magnetic field B_{ext}. Thus, a pronounced linewidth narrowing is expected, in agreement with our experimental data (see Fig. 1).

In summary, PL spectroscopy of $Cd_{0.93}Mn_{0.07}Te$ SQDs has unambiguously shown the formation of quasi-zero-dimensional exciton MPs. The complete suppression of inhomogeneous broadening effects in the SQD has allowed to obtain the energy, the internal magnetic field and the volume of an exciton MP in a DMS SQD by comparing experimental data with model calculations. In addition, the impact of the magnetic environment on the PL linewidth in SQDs has been emphasized by comparing measurements of $Cd_{0.93}Mn_{0.07}Te$ SQDs and their nonmagnetic $Cd_{0.93}Mg_{0.07}Te$ counterparts.

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