

Magnetic-field-induced abrupt spin-state transition in a quantum dot containing magnetic ionsM. Koperski,^{1,2,*} T. Smoleński,¹ M. Goryca,¹ P. Wojnar,³ M. Potemski,² and P. Kossacki¹¹*Institute of Experimental Physics, Faculty of Physics, University of Warsaw, Poland*²*Laboratoire National des Champs Magnétiques Intenses, CNRS-UJF-UPS-INSA, Grenoble, France*³*Institute of Physics, Polish Academy of Sciences, Warsaw, Poland*

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We present the results of a comprehensive magneto-optical characterization of single CdTe quantum dots containing a few Mn^{2+} ions. We find that some quantum dots exhibit an unexpected evolution of excitonic photoluminescence spectrum with the magnetic field. At a certain value of the magnetic field, specific for each quantum dot, each of the broad spectral lines related to the recombination of various excitonic complexes confined inside the dot transforms into a pair of narrow lines split by several meV. We interpret this abrupt change in the character of excitonic emission spectrum as a consequence of a transition from a nonpolarized state of the Mn^{2+} spins in a low field regime to a highly (almost fully) polarized state above the critical magnetic field. Various optical experiments, including polarization-resolved studies, investigation of different excitation regimes and time-resolved measurements corroborate this scenario. However, these measurements indicate also that the observed effect is not related or influenced by the photocreated charge carriers, but it is rather originating from unusual spin configuration in the cluster of Mn^{2+} ions.

DOI: [10.1103/PhysRevB.94.245439](https://doi.org/10.1103/PhysRevB.94.245439)**I. INTRODUCTION**

Semiconductor quantum dots (QDs) doped with magnetic ions constitute a convenient platform for studying magnetism in nanoscale. Both the energy of the system and its spin properties are encoded in the photoluminescence (PL) signal from such QDs. Narrow spectral lines, originating from the atomlike states of the carriers confined in the QDs, ensure high spectral resolution necessary for observing the detailed fine structure of the excitonic complexes [1–10]. The energy spectrum of such QDs is determined predominantly by the exchange interaction between the angular momentum of the carriers and the spin of the magnetic ions. It has been demonstrated that for QDs with a single ion (sometimes even two ions [11,12]) one can distinguish isolated lines related to specific projections of the spin of the magnetic ion (ions) on the quantization axis (given by the anisotropy of the heavy hole [13]). Such direct readout of the magnetic ion spin state has been successfully exploited not only for the studies of the static spin properties [14,15], including the influence of an external magnetic field [16], but also dynamical processes such as relaxation [17] or coherent precession [18,19] of a spin of a magnetic ion incorporated into a semiconductor lattice. Additionally, more application-oriented studies are possible, e.g., a demonstration (as a proof of concept) of a magnetic memory based on a single spin [20–22]. Apart from such extremely diluted concentration of magnetic ions, extensive efforts were devoted to studying the properties of QDs with a large number of ions [23] (of the order of hundreds). The most interesting findings in these QDs are related to the formation of a magnetic polaron [24–27]. In particular, the time resolved experiments revealed a significant redshift of the emission lines during the lifetime of excitonic complexes in QDs with sufficiently high concentration of Mn^{2+} ions (above 3%) [28].

With this work we aim to fill the gap in the research of magnetic QDs by exploring the regime of intermediate concentration of magnetic ions, when on average only a few ions are present in a single QD. For some of such QDs, in which the Mn^{2+} ions appear to be located at the QD's edge, we found an unexpected type of the evolution of the PL spectra with the magnetic field in Faraday configuration, indicating a rapid transition of the spin state of the magnetic ions at a certain, critical value of the magnetic field ranging from 3 to 10 T for various QDs. Such a behavior of the system of spins in a QD comes as a surprise, since it strongly deviates from the typical magneto-PL data for the magnetic QDs studied so far. A firm explanation of the observed transition from a nonmagnetic to a highly (almost fully) polarized spin state of the magnetic ions remains to be established. However, the comprehensive study of this effect by means of various spectroscopic tools presented in details here provides a solid ground for possible interpretations.

II. SAMPLES, EXPERIMENTAL SETUP AND BASIC OPTICAL CHARACTERIZATION

The samples used in our experiments were grown by the molecular beam epitaxy (MBE). They contained a single layer of CdTe QDs embedded in a ZnTe barrier. The doping with Mn atoms was introduced into the QD formation layer and adjusted so that a significant number of QDs would contain a few Mn^{2+} ions. The initial estimation of the Mn^{2+} ions concentration was based on the calibration of the temperature of the Knudsen effusion cell (related to a flux of the Mn atoms in the MBE growth process). In the case of the most intensively investigated sample in this work, the Mn atoms were introduced to one out of six deposited CdTe monolayers, and the estimated Mn concentration in this monolayer yields 0.4%. Therefore the effective concentration of the Mn^{2+} ions in the QDs can be estimated to be 0.07%. This corresponds to a presence of 2–7 Mn^{2+} ions in a single QD (depending on the

*maciej.koperski@fuw.edu.pl

assumed size of a QD within reasonable constraints established, e.g., from the atomic force microscope studies [29]).

The samples were first investigated with magneto-PL experiments, which were followed by comprehensive time-resolved studies. The continuous wave (CW) optical experiments have been performed under the nonresonant excitation conditions using the linearly polarized laser lines at 488 nm or 514 nm or 532 nm. The high magnetic field experiments (up to 29 T) were performed in a polihelix resistive magnet with outer Bitter coil [30,31]. In this setup the samples were placed inside a probe filled with helium exchange gas (cooled down to about 10 K) and equipped with piezostages allowing x - y - z sample positioning with submicrometer precision. A fiber-based optical system was used for the excitation of the sample and for the detection of the spectrally resolved PL signal with a charge-coupled device (CCD) camera. The laser spot diameter in this setup was $\sim 1 \mu\text{m}$.

For time-resolved experiments a split-coil superconducting magnet with a direct optical access was used. The sample was cooled down with pumped liquid helium to 1.7 K. An immersive reflective microscope allowed a focalization of the laser beam down to a submicrometer spot. In this setup, apart from further PL characterization of the samples, the temporal evolution of the PL spectra (with overall resolution of 10 ps) was measured with a streak camera as the detection unit and under the pulsed linearly polarized excitation at 560 nm, supplied by the OPO laser. Also, the effects of an introduction of a dark period in the excitation (tens of microseconds duration) were studied by using an electrically controlled laser with a fast (single nanoseconds of rise/fall times) turn on/off module for excitation and a CCD camera equipped with a gated microchannel plate for detection.

The studies presented here concern the optical properties of single QDs. Although the large concentration of QDs in our samples (of the order of 10^{10} cm^{-2} [29]) results in about ~ 100 QDs under the laser spot, one can easily distinguish sets of separate lines from single QDs in the low-energy tail of the broad PL band. As the concentration of the Mn^{2+} ions in our samples is rather low, it is still possible to recognize the PL signal from single nonmagnetic QDs [see Fig. 1(a)] with a typical pattern of individual narrow lines (linewidth below $100 \mu\text{eV}$) corresponding to the recombination of different excitonic complexes. At the same time, the QDs with a single Mn^{2+} ion [see Fig. 1(b)] may be identified, based on the characteristic splitting of the excitonic lines (sixfold in the case of the neutral exciton). Most frequent in our samples are, however, locations which display the broader spectral lines [see Fig. 1(c)]. As tested with photon correlation experiments [32], these broader emission lines are also attributed to single quantum dots. Moreover, such broader lines appear in the PL spectra in a characteristic sequence of multiple separated peaks originating from the recombination of different excitonic complexes confined in the same QD, in a similar manner as for single nonmagnetic QDs or the dots doped with single Mn^{2+} ions. The origin of the broadening is related to the thermal spin fluctuations in the cluster of Mn^{2+} ions in the QD, which significantly extends the energy space available for a particular excitonic complex. As the large number of possible spin configurations grows exponentially with the number of Mn^{2+} ions, the detailed

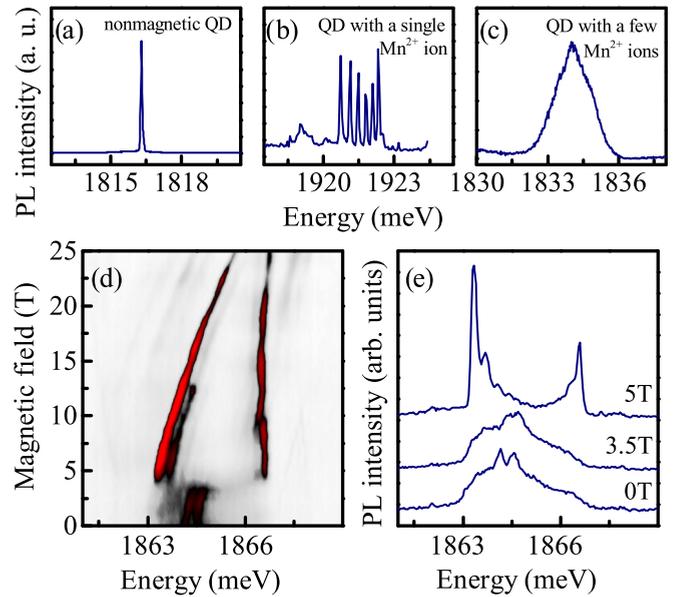


FIG. 1. (a)–(c) Example PL spectra showing a neutral exciton transition without the magnetic field for three selected QDs from the same sample. The spectra correspond to (a) a nonmagnetic QD, (b) a QD with a single Mn^{2+} ion, and (c) a QD1 with a few Mn^{2+} ions. For some QDs with a few Mn^{2+} ions the evolution of the spectrum with the external magnetic field applied in Faraday configuration reveals an abrupt transformation of the PL lines, as seen in a color-scale map of magneto-PL spectra for QD2 presented in (d). From a single broad feature the PL line transforms into two separate narrow lines, which is clearly illustrated by the PL spectra measured at selected values of the magnetic field (e).

fine structure may no longer be resolved in the case of three or more Mn^{2+} ions embedded in the QD. Therefore, in the PL spectra the excitonic transitions are seen as broader features.

III. MAGNETOLUMINESCENCE STUDIES

The magnetoluminescence studies of QDs exhibiting broad emission lines reveal an unexpected behavior of the spin of the Mn^{2+} ions under the influence of an external magnetic field. For a certain number of investigated QDs (roughly about 20–30% for different samples) an abrupt transformation occurs in the character of the PL spectrum: A single broad PL line splits into two narrow components at a certain critical value of the magnetic field, as presented in Figs. 1(d) and 1(e). Above the critical field, the two narrow lines follow the magnetic field evolution similar to the one known from the studies of excitonic transitions in nonmagnetic QDs. In particular, the energies of both narrow lines evolve exclusively due to the linear excitonic Zeeman effect and quadratic diamagnetic shift. Moreover, the linewidth of the two emission lines above the critical field is similar to the one observed for nonmagnetic QDs and remains roughly constant with the further increase of the magnetic field, as one could expect for (Cd,Mn)Te QDs in the regime of high (almost full) spin polarization of the Mn^{2+} ions. The two lines tend to cross at high magnetic field, which remains above the experimental limitation of the present

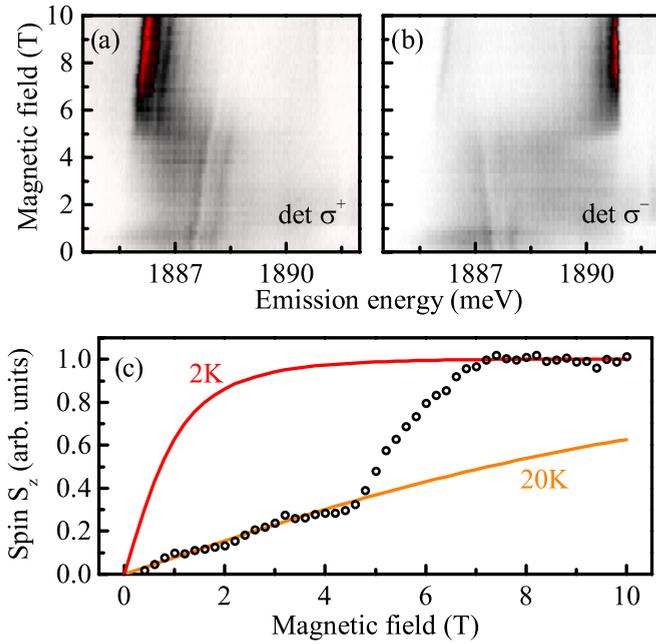


FIG. 2. (a),(b) The polarization-resolved magneto-PL maps of a neutral exciton for a QD3 with a few Mn^{2+} ions (sample temperature of 1.7 K) exhibiting an abrupt transformation. The maps were measured in (a) σ^+ and (b) σ^- polarization of detection under excitation with a linearly polarized 532 nm diode laser. (c) The evolution of the mean spin of the Mn^{2+} ions obtained from these data (see main text). The solid lines represent the Brillouin functions for 5/2 spin at effective temperatures of 2 K and 20 K.

study. That crossing would correspond to the compensation of the excitonic Zeeman effect and the giant Zeeman effect of the Mn^{2+} ions, which shift the energies of excitonic spin states in the opposite directions for (Cd,Mn)Te material. Based on these observations, we interpret the transformation of the PL spectra as a transition from a nonpolarized to a highly polarized spin state of the Mn^{2+} ions. The peculiar behavior of the magneto-optical spectra has been observed in a number of quantum dots. The results presented here refer to experiments performed on eight different dots.

In order to quantitatively characterize the transformation, we turn to the analysis of the polarization-resolved magneto-PL spectra of the neutral exciton, which are presented in Figs. 2(a) and 2(b). We tentatively assume that the PL signal measured in a given circular polarization reflects directly the probability distribution of the total spin of the Mn^{2+} ion system. In such a case, the calculation of the first moment of the PL intensity distribution at a certain value of the magnetic field yields a quantity reflecting, in the first approximation, the magnetization of the Mn^{2+} ions in a QD. The circular polarization resolution in detection allows us to account for the excitonic relaxation processes, which for some QDs are clearly manifested in zero-field spectra as the nonsymmetric shape of the excitonic lines. Therefore, we use a difference between the magnetization of Mn^{2+} ions calculated from the PL spectra detected in opposite circular polarizations to obtain a value unperturbed by the excitonic relaxation. Such a value is presented in Fig. 2(c) as a function of the magnetic field. A clear transition is seen, starting at ~ 5 T and progressing

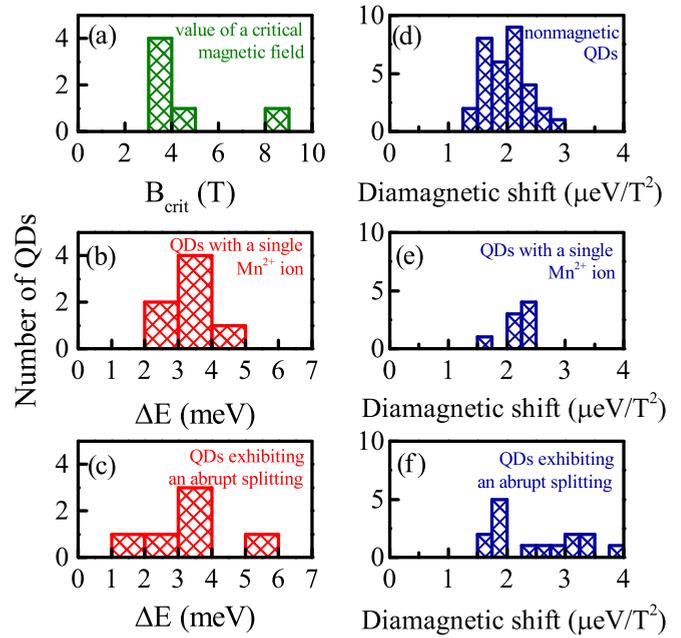


FIG. 3. (a) Histogram presenting the value of a critical magnetic field, which induces an abrupt transition of the PL lines from QDs with a few Mn^{2+} ions. (b),(c) Zero-field effective exchange splittings of the neutral exciton measured for the QDs containing single Mn^{2+} ions (b) and for the dots with a few Mn^{2+} ions (c). (d)–(f) Histograms of diamagnetic shift coefficients determined for (d) nonmagnetic QDs, (e) QDs with single Mn^{2+} ions, and (f) for the QDs with a few Mn^{2+} ions exhibiting a magnetic-field-induced transition. The data clearly show that in the latter case the diamagnetic shift is higher, which strongly indicates that the QDs exhibiting the transformation are of larger size.

until ~ 7 T, when a saturation value is reached. For the reference, the evolution of the magnetization is compared with the two Brillouin function curves for a 5/2 spin at an effective temperature of 2 K (close to the helium bath temperature equal to ~ 1.7 K) and 20 K. The values of the temperature are chosen so that the Brillouin curves match the magnetization data in the higher and lower field regimes (after and before transition, respectively). Even though the Brillouin function, originating from a mean field approximation applied to a paramagnetic system, may not be expected to accurately describe the spin state of a few interacting Mn^{2+} ions, the comparison suggests the existence of a mechanism suppressing the spin polarization of the Mn^{2+} ions in the low field regime, which is abruptly switched off at the field when the transition is seen.

An analysis of the magnetic field evolution of the PL spectra for several QDs exhibiting a transition allows us to establish basic properties of the cluster of Mn^{2+} ions in such specific QDs. First, an important parameter describing the phenomenon is the value of a critical magnetic field at which the transition is seen. Its values for several QDs are presented in Fig. 3(a). The critical value of the magnetic field is a particular property of a specific QD and typically ranges from 3 T up to 5 T, however for some QDs can reach as high as 10 T. Second, we can determine an effective zero-field exchange splitting between σ^+/σ^- exciton interacting with highly spin-polarized Mn^{2+} ions by extrapolating the evolution

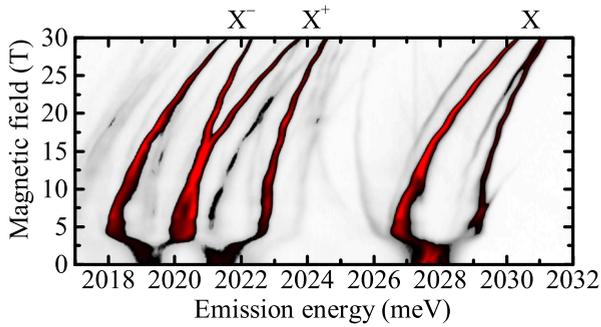


FIG. 4. The magnetic field evolution of a PL spectrum of a single CdTe/ZnTe QD4 with a few Mn^{2+} ions presented in a wider spectral range. The data show that the PL line transformation appears for all excitonic complexes visible in the QD PL spectrum. In particular, both neutral and charged complexes reveal the same kind of transformation at exactly the same value of the critical magnetic field.

of the two narrow lines above the critical field down to the zero magnetic field. Such a splitting depends on the number of Mn^{2+} ions in the QD, their position in the lattice structure, and the shape of the excitonic wave function governed by the geometry of the QD, all of which finally determine the wave function overlap between the Mn^{2+} ions and the exciton. It is interesting to compare the average value of this splitting with a typical value of the zero-field exchange splitting for the QDs doped with single Mn^{2+} ions from the same sample, which is directly available as a splitting between the two outermost lines in a neutral exciton sextuplet. Relevant statistical data are presented in Figs. 3(b) and 3(c) in the form of histograms for QDs with a single and with a few Mn^{2+} ions, respectively. On average the value of the splitting in both cases is roughly the same, which suggests that for the QDs exhibiting the transition the Mn^{2+} ions are located rather at the perimeter of the QD than in its center (one has to note, however, than in the case of the statistics for the QDs with a single Mn^{2+} ion, the average value is shifted towards higher energy by our choice of QDs with clearly resolvable six lines). An additional factor that further reduces the Mn-exciton wave function overlap is related to a larger size of the QDs with a few Mn^{2+} exhibiting a transition, which is revealed by the higher value of the diamagnetic shift for these QDs as compared to the QDs without or with a single Mn^{2+} ion [Figs. 3(d)–3(f)].

At this point a question arises about the origin of the observed effect: whether the transition is caused by some process mediated by photocreated carriers, which could be controlled by means of optical techniques, or is it an intrinsic property of the cluster of the magnetic ions due to a particular spin configuration defined by their position in the crystal lattice, mutual exchange interaction, and interaction with the residual carriers. In order to address these two possibilities, we first analyze the magneto-PL spectra in a wider energy range, covering the emission of different excitonic complexes. Figure 4 illustrates the magnetic field evolution of a spectrum for a selected QD exhibiting a transformation. The complete spectrum of a CdTe QD consists of a set of lines, which form a characteristic pattern enabling the identification of particular excitonic complexes. Hence, it is possible to recognize the emission of both neutral and charged states. The lines related to

the neutral exciton and negatively/positively charged excitons are indicated in the figure. Importantly, the same kind of transformation is seen for all excitonic lines. Particularly interesting in this context are the trion states, which are in fact composed of three carriers, however due to the singlet state of a pair of carriers of the same type the trions effectively interact with the Mn^{2+} spins in the same way as a single minority carrier (the electron in the case of a positive trion and the hole in the case of a negative trion). Apparent equal robustness of the observed transformation regardless of the presence of an electron, a hole as well as an electron-hole pair suggests that there is no link between observed excitonic complex and the actual origin of the effect.

The type of transformation of the PL lines in the magnetic field that we describe here is not known to appear in other magnetic systems. Seemingly similar deviations from the typical giant Zeeman effect were observed for CdTe/ZnTe quantum wells [33] or QDs [34] with high concentration of magnetic ions. These deviations were interpreted as related to the influence of carriers trapped in a wetting layer, which can effectively interact with the Mn^{2+} ions only in the low magnetic field regime. More specifically, at low fields the spin-flip processes between the angular momentum of such carriers and the spin of the Mn^{2+} ions enable the transfer of energy and may lead to an increased temperature of the Mn^{2+} spins. However, a similar scenario is highly unlikely in our case for the reasons discussed below.

First, we verify this hypothesis experimentally by changing the excitation conditions (energy and power). A typical macro-PL spectrum of an ensemble of QDs from our samples is presented in Fig. 5(a). It covers a spectral range roughly from 1850 meV to 2250 meV. A potential wetting layer in our sample (however not appearing in the PL signal) would reside at higher energies but below the ZnTe (barrier) band gap. In Figs. 5(b) and 5(c) we present the magnetic field evolution of a PL line for a QD exhibiting a rapid transformation measured under two excitation regimes: just below the band gap (with a 532 nm diode laser) and quasisonant (with a tunable rhodamine dye laser set for 580 nm). While the creation of carriers in a potential wetting layer is feasible for the 532 nm excitation, the quasisonant excitation excludes such possibility. The presence of an equally robust PL line transformation for both regimes of excitation directly confirms that the carriers in the potential wetting layer cannot be responsible for the observed effect. Moreover, any heating mechanism should be strongly dependent on the excitation power. For example, in the case of QDs with single Mn^{2+} ions, the heating of the ion spin by the laser excitation is known to be very efficient with the corresponding effective temperatures of the Mn^{2+} ion reaching tens of K for sufficiently high excitation powers [1,7,8,35]. Here, we do not observe any significant change in the nature of the transformation by varying the laser excitation power by two orders of magnitude, as shown in Figs. 5(d)–5(f).

Second, the magnetic field needed to suppress the potential heating mediated by the carriers in the wetting layer is proportional to the concentration of the magnetic ions. Such concentration in our samples is almost an order of magnitude lower than in the samples used in the previous reports. This is revealed, for instance, by the energy splitting between

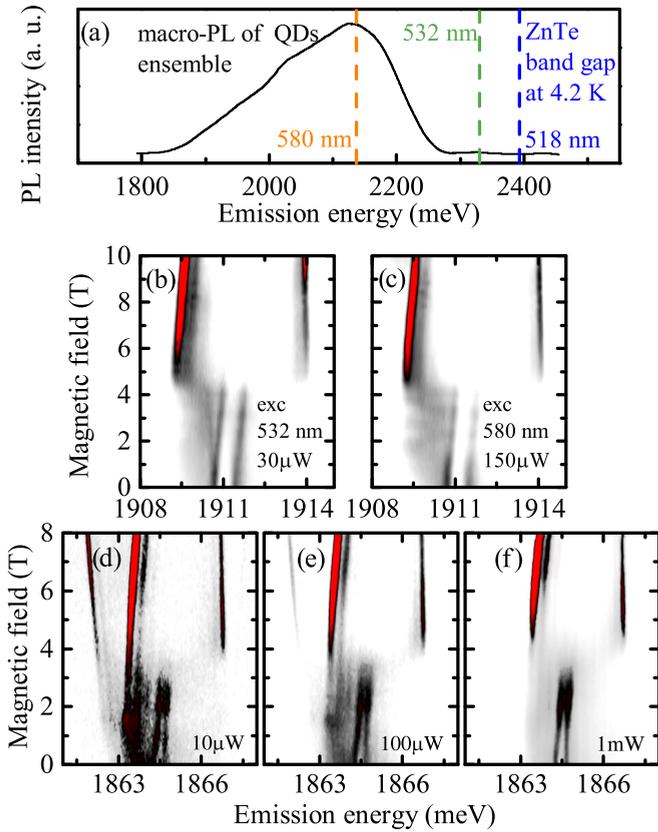


FIG. 5. The presence of magnetic-field-induced abrupt transformation of the PL line for CdTe/ZnTe QDs with a few Mn^{2+} ions is found to be independent of the laser excitation conditions. This includes two different regimes of the excitation energy schematically depicted in panel (a): just below the barrier band gap with 532 nm diode laser and quasiresonant with 580 nm tunable dye laser. The color-scale maps presenting the magneto-PL spectra for QD5 obtained under both excitation regimes (b) and (c) confirm that the character of the abrupt transformation remains unaffected by the choice of the laser energy. Additionally, no particular influence of the laser excitation power on the transformation is observed, as seen in the data for QD6 presented for (d) 10 μW , (e) 100 μW , and (f) 1 mW laser excitation power of 514 nm Ar laser line.

the two σ^+/σ^- polarized lines in the magnetic field that highly polarizes the magnetic ions spins. The splitting equals 2–4 meV in our samples as compared to the splitting of about 20 meV observed for the samples used before. Consequently, in our case the heating could be effective only for the magnetic fields remaining in the range of tens of mT, which are much smaller than the values of critical magnetic fields 3 T–10 T at which the rapid transformation of the PL lines appears. This observation finally shows that the transformation cannot be due to the invoked heating mechanism.

IV. EVIDENCE FOR A STEADY SPIN STATE OF Mn^{2+} IONS IN TIME-RESOLVED EXPERIMENTS

Valuable information relevant for establishing the origin of the abrupt field-induced transformation of the PL lines in the studied QDs may come from the time-resolved photo-

luminescence measurements. Such experiments can provide deeper insight into the character of the transformation and help to distinguish between the intrinsic static spin properties of the Mn^{2+} ions and dynamical processes related to the capture/recombination of the photocreated carriers in the QD. In fact, the data obtained from the time-integrated PL experiments do not exclude the possibility that the spin of the Mn^{2+} ions becomes highly polarized at the critical value of the magnetic field due to the interaction with excitonic complexes. One approach to verify this scenario is to investigate the PL transients. Such measurements would reveal the possible variations of the Mn^{2+} ions magnetization in the timescale shorter or comparable to the excitonic lifetime (~ 300 ps for CdTe/ZnTe QDs). In order to trace the temporal evolution of the PL spectra with high resolution (~ 10 ps) we use a streak camera. As the line transformation in the magnetic field is observed for all excitonic complexes, we focus on a neutral exciton PL line. The temporal evolution of the neutral exciton PL line measured for a selected QD in the two values of the magnetic field (4 T and 5.4 T) are presented in Figs. 6(a) and 6(b). The corresponding time-integrated PL spectra obtained in the same fields are shown in Figs. 6(c) and 6(d). The values of the magnetic field were chosen to be just below and just above the transformation [the evolution of the PL spectrum with the magnetic field is presented in Fig. 6(e)]. The temporal profiles in both regimes of the magnetic field do not indicate any transformation of the spectral lines during the lifetime of a single excitonic complex.

The measurement of the PL decay does not entirely exclude that the observed transformation of the PL lines is related to the photocreated carriers. Another possibility is that the transformation originates from a stationary state established by a series of multiple capture-recombination events. The second time-resolved experiment was designed to verify this hypothesis. In these measurements we used a continuous-wave (CW) 405 nm laser with a module of fast, electrically controlled turn on/turn off system, which enabled us to introduce a dark period (50 μs) in the excitation. The dark period of such duration is significantly longer than the characteristic spin relaxation times of clusters of Mn^{2+} ions embedded in CdTe crystal [17]. Consequently, at the moment of excitation relaunch the system of Mn^{2+} ions may be considered as fully thermalized. During the excitation period (1 μs) the PL spectrum was recorded with a gated CCD camera in a series of temporal windows of 30 ns, allowing us to probe the stationary state of the Mn^{2+} ions in darkness (i.e., absence of excitation). Again, a QD exhibiting a rapid transformation was selected [the magneto-PL map shown in Fig. 6(f)] and the experiment was performed at two values of the magnetic field (4.8 T and 7 T), chosen in the middle and just above the transformation. In Figs. 6(g) and 6(h) the two PL spectra are compared: the one under CW excitation and the second one recorded during 30 ns time window just after the dark period. No particular difference observed between these two spectra allows us finally to conclude that the field-induced transformation of the excitonic lines is not mediated by the photocreated carriers. This in turn strongly suggests that the origin of the effect is related to the intrinsic properties of the magnetic cluster of Mn^{2+} ions.

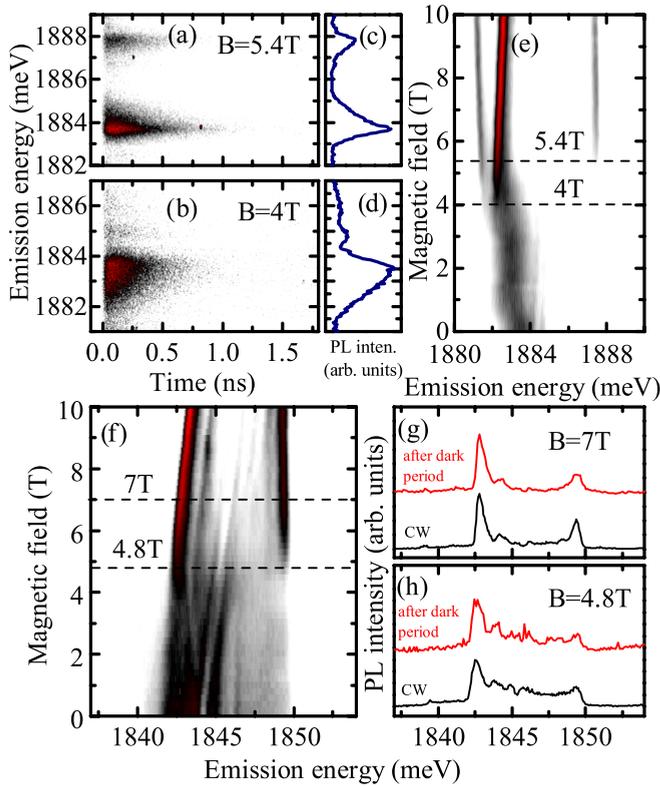


FIG. 6. The temporal traces of the neutral exciton PL line measured for the QD7 containing a few Mn^{2+} ions with a streak camera in the two values of the magnetic field: (a) 4 T and (b) 5.4 T. The respective time-integrated spectra are shown in (c) and (d) for comparison. The critical value of the magnetic field for this QD is equal to about 5 T, as seen in the magnetic field evolution of the PL spectrum (e). For the second time-resolved experiment the neutral exciton PL spectrum after a dark period was compared with the one obtained under CW excitation. QD8 with a transition at 5 T was selected, as seen in (f). The comparison of the two spectra measured in the magnetic field (g) below and (h) above the transformation indicate no influence of the introduced dark period on the character of the transformation.

V. CONCLUDING REMARKS

At present we consider mainly two scenarios. One possibility is that the Mn^{2+} ions were incorporated into the crystal lattice so close to each other that their mutual exchange interaction is significant. In such a case they could form a highly frustrated system exhibiting a transition in the magnetic field, in a similar manner as, i.e., magnetic ions in single molecules, which can be found in particular configurations such as, e.g., triangular clusters [36]. On the other hand, even distant Mn^{2+} ions can interact with each other via exchange interaction with a carrier [12], which can stochastically occupy the QD in the absence of optical excitation. Such an interaction

can have nontrivial character, especially in the case of the hole, with its intrinsic anisotropy additionally complicated by possible effects of spin textures [37,38]. It is also worth noting that potential abrupt changes in the magnetization of coupled Mn^{2+} ions could originate from large anisotropy of their mutual interaction. In an extreme case, if the Mn-Mn exchange interaction would be purely of an Ising type (i.e., when the spins couple exclusively along a single direction), then at sufficiently low temperature the multiple magnetization steps [39] would merge into a single transition, directly from nonmagnetic to a fully polarized state of the Mn^{2+} ions. Moreover, there exist mechanisms that could induce certain anisotropy in the Mn-Mn interaction. One of them is the coupling of the Mn^{2+} ion with the excited states of the hole, which leads to a quadratic in the spin of the Mn^{2+} ions perturbation along the heavy hole anisotropy axis. A similar effect can also be induced by strain, which can be particularly strong in a QD. However, the estimation of the magnitude of these effects shows that in real systems they cannot dominate over the Mn-Mn exchange coupling. For that reason, such reasoning still remains at conceptual level.

VI. SUMMARY

The magneto-optical studies of CdTe/ZnTe QDs doped with a few Mn^{2+} ions revealed an abrupt transition from a nonpolarized to a highly polarized state of the magnetic ions system inside the dot, which occurs upon application of the magnetic field in the Faraday geometry. The uncovered effect has been thoroughly characterized with various spectroscopic techniques. The influence of the excitation energy and the laser power has been investigated as well as temporal transients of the emission lines in two different time-resolved experiments. Our findings lead to a conclusion that the origin of the transition is most likely unrelated to the photocreated carriers. Therefore further exclusively optical studies may not be helpful in providing a reliable test against various hypotheses that have been put forward. Currently, more efforts are needed to study the origin of the transition, possibly involving new research directions, as the PL measurements most likely cannot provide a definitive answer about the origin of the effect. Therefore, in the future studies one should include other techniques, such as optically detected magnetic resonance or nonoptical magnetization measurements.

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