Spin-lattice relaxation of an individual Mn^{2+} ion in a CdTe/ZnTe quantum dot

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We present the study of the spin-lattice relaxation of an isolated Mn^{2+} ion in a single CdTe/ZnTe quantum dot. The measurements are done in a wide range of magnetic field. The spin-lattice relaxation rate is determined in a time resolved experiment. The ion spin state is driven out of equilibrium using optical orientation of the Mn^{2+} spin in a system of two coupled dots. Then the light is switched off and the Mn^{2+} ion spin relaxes. The Mn^{2+} spin state is measured after switching the light on again. We discuss the magnetic field dependence of the spin-relaxation rate in light of two theoretical models: one based on scattering of transverse acoustic phonons in the presence of a finite uniaxial Mn^{2+} spin anisotropy, and the second relying on presence of quantum dot charge state fluctuations.

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I. INTRODUCTION

The spin dynamics caused by relaxation processes has been studied for years in bulk and two-dimensional (2D) diluted magnetic semiconductor (DMS) materials [1–12]. The main interactions which keep the system of magnetic ions in thermodynamical equilibrium with the crystal lattice were identified as a spin-lattice interaction [13], which can lead to a global change of magnetization, and spin-spin interaction [1,2,6], which can act locally and lead to the spin diffusion to non-spin-conserving centers (e.g., clusters of more than two coupled magnetic ions, which statistically exist within the random distribution of the ions [5,7]). It has also been shown that carriers can mediate the energy transfer between magnetic ions [11,14–16], as well as between the ions and the crystal lattice [8].

Although many features of the spin-lattice relaxation (SLR) of a system of magnetic ions in a semiconductor crystal are well understood, there are still several aspects that remain little investigated. First of all, due to the limitations of the sample growth and experimental techniques, the spin dynamics of a big ensemble of magnetic ions was studied in previous experiments. This was done either with diluted magnetic bulk materials [1–5] or quantum wells [6–12]. However, even for very diluted materials the distance l between adjacent magnetic ions was not negligible, the largest studied l being of order of a few nanometers (e.g., about 3 nm in Ref. [4]). Therefore the spin-spin interactions between these ions always had to be considered, which hindered the analysis and understanding of the single-spin SLR mechanism.

Among many advantages of a single quantum dot (QD) with a single magnetic ion, the large distance to any other magnetic ion is particularly useful in investigations of the SLR. This distance, orders of magnitude larger than in a DMS bulk material or a quantum well system, allows us to consider the magnetic ion in the QD as a spin truly isolated from any other magnetic ions. Thus the only mechanism which can lead to the relaxation of the ion spin is its coupling with the lattice (SLR).

In this work we present a detailed study of the SLR of an individual magnetic ion embedded in a semiconductor QD in a wide range of magnetic field and discuss the experimental results in light of two theoretical models.

II. SAMPLE AND EXPERIMENTAL SETUP

The studied sample is grown by molecular beam epitaxy. It contains a single layer of self-assembled CdTe QDs embedded in a ZnTe matrix. Only the material of the dots is weakly doped with Mn^{2+} ions. Their concentration is adjusted to maximize the probability of formation of QDs with single Mn^{2+} ions [17]. The density of magnetic ions in the QD formation layer yields about 3×10^9 cm^{-2}, and is estimated based on the information on the Mn flux and the opening time of the Mn effusion cell during the growth of the sample. Such density corresponds to the mean distance between nearest-neighboring ions equal to about 140 nm.

The excitation of the sample is realized with a continuous-wave rhodamine 6G laser tunable in the range 560–600 nm. It allows us to perform the photoluminescence excitation (PLE) measurements in the energetic region corresponding to the maximum of the photoluminescence band of the ensemble of QDs. An acousto-optic modulator is used to repeatedly deflect the laser beam with an arbitrary frequency and deflection time. This introduces a dark period in excitation, with a transition time below 10 ns.

A microscope objective and piezoelectric x-y-z stages enables the focalization of the laser beam to a spot with a diameter of about 1 μm and scanning of the sample surface with nanometer precision. The system is kept at the temperature equal to 12 K in a cryostat situated in a magnet. The direction of the applied magnetic field is parallel to the growth axis of the sample and the detection of the emitted photons is realized in Faraday configuration. The spectrally resolved detection is provided by a spectrometer with a charge-coupled device (CCD) camera attached to one of the two outputs. An avalanche photodiode, attached to the second output, together with the time-resolved photon counting system, is used to record temporal profiles of the photoluminescence at selected wavelength.

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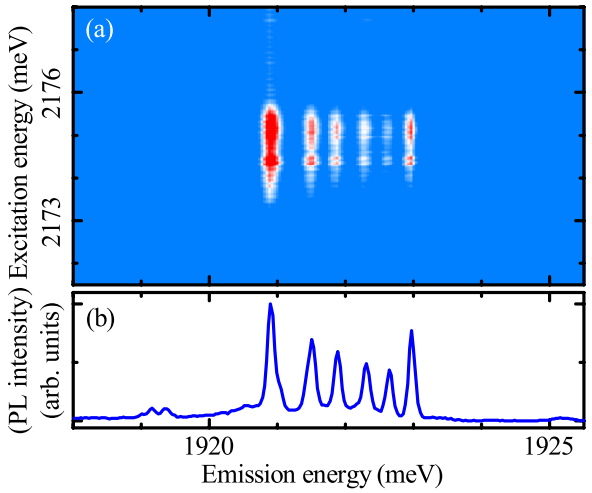

FIG. 1. (Color online) (a) The photoluminescence excitation map of a system of two coupled QDs with a single Mn^{2+} ion in the emitting one. The visible resonance corresponds to the neutral exciton states in both emitting and absorbing QDs. (b) The emission spectrum of the Mn-doped QD, taken for the resonant excitation energy at 2175.2 meV, showing the characteristic sixfold splitting of the neutral exciton line.

III. EXPERIMENTAL METHOD

In order to study the spin-lattice relaxation of the Mn^{2+} ion one has to drive its spin out of equilibrium. In our experiment we exploit the mechanism of optical orientation of a single Mn^{2+} spin in a system of two coupled QDs [18]. Due to the tendency towards formation of QD clusters in our samples, it is very likely to find a pair of QDs close enough to enable a transfer of an exciton between them [19,20]. The orientation of the Mn^{2+} spin is achieved by taking advantage of the spin conservation property of the transfer. As it was shown in Ref. [19] its efficiency can be as high as 70%. This value is also confirmed in the present experiment. The spin polarized excitons intentionally injected into a QD with a single Mn^{2+} ion lead to the orientation of the ion spin towards the spin-up or spin-down state, depending on the excitonic angular momentum [18,21–23]. Therefore, we can induce a nonequilibrium state of the single Mn^{2+} ion using the laser excitation and by turning the excitation off we let the spin relax back to the equilibrium.

An identification of a pair of coupled QDs is possible in the PLE measurement. By tuning the laser energy in resonance with excitonic energy level in one QD, a robust enhancement of the photoluminescence signal from the other dot is observed, usually 100–250 meV below the excitation energy [see Fig. 1(a)]. In the case of the presence of a single Mn^{2+} ion in the emitting QD a characteristic sixfold splitting of the neutral exciton (X) emission line is observed [24] [see Fig. 1(b)]. The splitting originates from the exchange interaction between the Mn^{2+} ion and the exciton. The energy of the X-Mn system depends on the Mn^{2+} spin projection on the exciton quantization axis (parallel to the sample growth axis). As a result, each of the observed six lines, detected in circular polarization, corresponds to one of the six possible projections of the Mn^{2+} 5/2 spin [25,26], and the intensities of those lines reflect the probability of occupation of corresponding states. This provides a convenient method to read out the spin state of the Mn^{2+} ion.

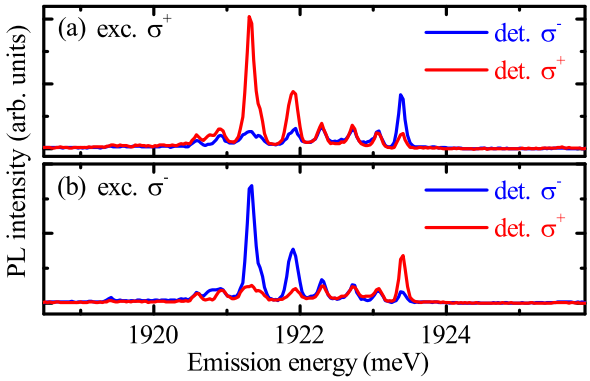

FIG. 2. (Color online) Photoluminescence spectra of a QD containing a single Mn^{2+} ion excited at the resonance with (a) σ^+ and (b) σ^- polarized light, detected in indicated polarizations.

The feasibility of the Mn^{2+} spin orientation for one of the studied QDs without an external magnetic field is illustrated in Fig. 2. The four spectra are obtained under resonant excitation of the absorbing QD with circularly polarized light [σ^+ in Fig. 2(a) and σ^- in Fig. 2(b)] and circular polarization resolution in detection. In the case of cross-polarized excitation and detection a robust enhancement of the low-energy lines occurs while for co-polarized configuration the high-energy lines are enhanced. Such observation proves the possibility of efficient manipulation of the Mn^{2+} spin state with light.

In the time-resolved experiment, both the excitation and the detection are σ^- polarized. During the experiment, we introduce a dark period in the laser excitation and monitor the temporal evolution of intensity of the lowest energy line of the QD PL spectrum after switching the excitation on again [see Fig. 3(a)]. Due to the Mn^{2+} relaxation, during the dark period the occupation of the monitored state ($S_z = +5/2$) decreases. Thus, after the relaunch of the laser excitation the line intensity is lower than before the dark period. Subsequently, a slow increase of the line intensity is observed, which is related to the Mn^{2+} optical orientation toward the

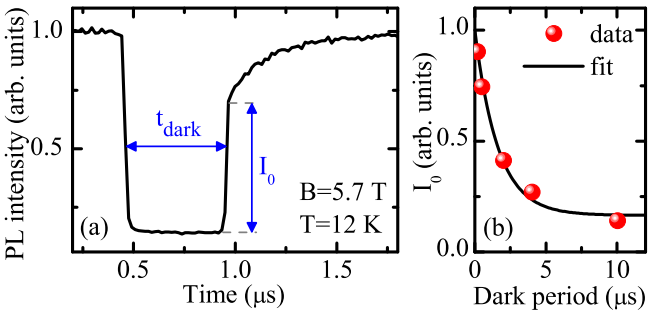

FIG. 3. (Color online) (a) An example of the intensity temporal profile of the lowest energy line from the neutral exciton sextuplet for an excitation scheme with a dark period introduced. (b) The dependence of the line intensity at the moment of the excitation relaunch (I_0) on the dark period duration (t_{dark}) fitted with an exponential decay.

state polarized opposite to the thermalized state. With the increase of the duration of the dark period the initial intensity after excitation relaunch (I_0) diminishes until saturation. It occurs when the dark period is long enough to enable full Mn^{2+} relaxation [see Fig. 3(b)]. The spin-relaxation rate is determined by fitting an exponential decay to the dependence of the I_0 on the dark period duration.

The described method is valid for a wide range of magnetic field. The only limiting factor of the measurement feasibility is the temporal resolution of the acousto-optical modulator, preventing the measurements of relaxation times shorter than 10 ns.

IV. RESULTS AND DISCUSSION

Figure 4 shows the dependence of the SLR rate on the magnetic field observed for a representative QD together with data measured in the former experiments on bulk diluted $Cd_{1-x}Mn_xTe$ material [4]. The relaxation rate of the single Mn^{2+} ion in the QD for low magnetic field (below 2 T) and temperature of 12 K is of the order of 10^4 s^{-1}, significantly lower than in bulk material at 4.7 K, and weakly depends on the value of the field. However, for higher fields it increases rapidly. For magnetic field in the range 2–10 T the SLR rate follows roughly the $1/\tau \propto B^3$ dependence. For higher fields this dependence is even stronger and the SLR rate reaches almost 10^8 s^{-1} above 11 T. This is over two orders of magnitude higher than the value measured for bulk $Cd_{1-x}Mn_xTe$ (for $x = 0.002$ and 0.01) [4].

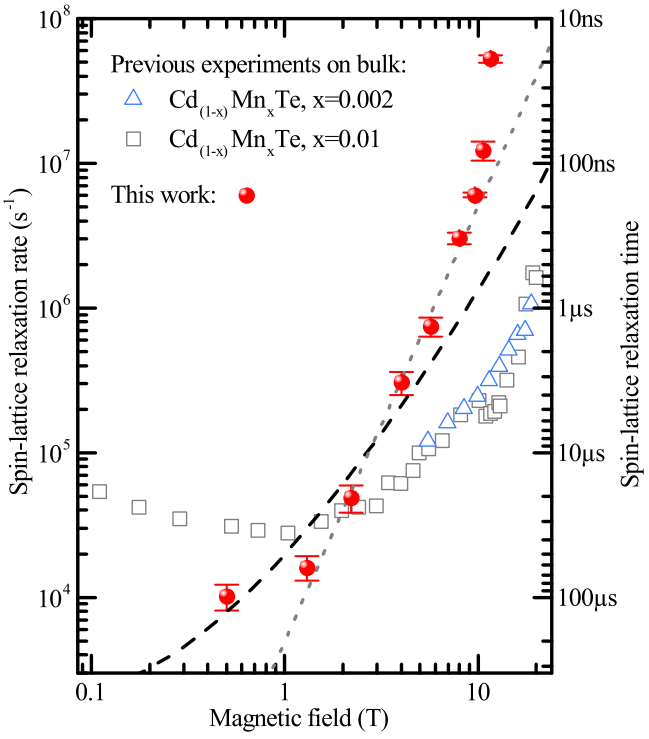

FIG. 4. (Color online) Spin-lattice relaxation rates for a single Mn^{2+} ion in a CdTe QD as a function of magnetic field measured at 12 K (dots) compared with results of previous experiments performed on bulk materials at 4.7 K (empty symbols), model of the intrinsic spin-relaxation process described in text and multiplied by a factor of 500 (black dashed line), and the $1/\tau \propto B^3$ dependence (gray dotted line).

Strong dependence of the measured SLR rate on the magnetic field clearly suggests that the relaxation is mediated by a single acoustic-phonon emission. In such a case, the rapid changes of the SLR rate are mostly related to the energy dependence of the phonon density of states (and of the matrix element for phonon scattering), since only the phonons with energies matching the Zeeman splitting $\Delta(B)$ of the involved Mn^{2+} spin states can participate in the relaxation process. On the other hand, any particular multiphonon relaxation mechanisms [e.g., a process involving simultaneous absorption of a phonon and emission of another phonon, with the difference of their energies equal to $\Delta(B)$] is unlikely to exhibit significant rate dependence on the magnetic field [27,28]. This is due to the absence of any strict requirement imposed on the energy of each individual phonon in such processes, as only the total energy transferred to the lattice (i.e., the difference of the energies of the two phonons) must be equal to $\Delta(B)$. On this basis we focus on the single phonon assisted relaxation.

First, we analyze the intrinsic spin-relaxation processes, involving only the isolated Mn^{2+} spin and the lattice. We start with the Hamiltonian of a single Mn^{2+} ion confined in a CdTe crystal [29]:

$$\mathcal{H} = g\mu_B \vec{B}\cdot\vec{S} + A\vec{I}\cdot\vec{S} + D_0\left(S_z^2 - \frac{S(S+1)}{3}\right) + \frac{a}{6}\left(S_x^4 + S_y^4 + S_z^4 - \frac{S(S+1)(3S^2+3S-1)}{5}\right) \quad (1)$$

where the first term represents the Zeeman splitting with Mn^{2+} Landé factor $g = 2$, $A = 680$ neV represents the hyperfine coupling to the $I = 5/2$ nuclear spin, $a = 320$ neV is related to the cubic crystal field, and D_0 describes the effect of a uniaxial strain component. The value of the latter parameter cannot be directly inferred from typical PL measurements. However, based on the results of EPR experiments performed on strained CdTe layers [29] and different experiments performed on CdTe/ZnTe QDs [21,30] we assume $D_0 \simeq 10$ μeV. Such value clearly indicates that the uniaxial strain dominates over the cubic crystal field and enables us to neglect the latter in our considerations of the SLR rate. One should also remember that hyperfine coupling with the ion nuclear spin introduces a mixing between Mn^{2+} spin states and thus opens another possible Mn^{2+} spin-relaxation channel. However, since A is much smaller than D_0, the invoked mixing is expected to be a secondary effect. More importantly, the efficiency of the hyperfine-related relaxation channel rapidly decreases with the Zeeman splitting induced by the magnetic field, in contrast to the experimentally determined ion spin-relaxation rate. This observation leaves us with a Zeeman term accompanied by a $D_0S_z^2$ term coupling the ion spin to the lattice. In such a case, one of the possible mechanisms of the SLR is related to rotation of the axes of the local crystal field by a single transverse acoustic phonon. It was quantitatively studied theoretically by Chudnovski *et al.* in Ref. [31], and the possible relevance of this relaxation channel for Mn^{2+} ion in a QD was recently pointed out [32]. Applying the general formula [31] to the case of the Mn^{2+} spin relaxation between 5/2 and 3/2 spin states

studied in our experiment, one obtains the following SLR rate:

$$\Gamma(B) = \frac{10}{3\pi\hbar^4 v^5 \rho} D_0^2 \Delta(B)^3 [2n_B(\Delta(B)) + 1], \qquad (2)$$

where $\Delta(B) = 4D_0 + g\mu_B B$ is the energy splitting of 5/2 and 3/2 spin states of the Mn^{2+} ion, $v = 1.79$ km s^{-1} is the speed of TA phonons in CdTe, $\rho = 5870$ kg m^{-3} is the mass density of the CdTe unit cell, and $n_B(\Delta) = (e^{\Delta/k_BT} - 1)^{-1}$ is the phonon occupation number at temperature T.

In general, the SLR rate field dependence is mostly related to the $\Delta(B)^3$ term in Eq. (2) and is further modified by the presence of the Bose-Einstein occupation term. At very low field ($B < \frac{4D_0}{g\mu_B} \simeq 0.3$ T) the relaxation rate rises slowly with B, as $\Delta(B)$ splitting is dominated by the uniaxial strain component. At higher fields $\Gamma(B)$ increases more rapidly, since $\Delta(B)$ is almost proportional to B. However, at $B < \frac{k_BT}{g\mu_B} \simeq 8.5$ T (for $T = 12$ K) the occupation term $n_B \approx k_BT/\Delta(B) > 1$ weakens the field dependence of the relaxation rate. Finally, at very high $B \gtrsim 20$ T the SLR rate $\Gamma(B) \propto B^3$.

Such theoretically predicted SLR rate dependence on the magnetic field qualitatively well describes the experimental results at sufficiently low field (dashed line in Fig. 4). At $B \gtrsim 5$ T the measured SLR rate increases more rapidly, which can be related to the presence of an additional spin-relaxation process not included in the above-mentioned theoretical model. However, there is a significant quantitative discrepancy between the model and the experimental results. The actual $\Gamma(B)$ calculated from Eq. (2) is more than two orders of magnitude lower compared to the experimentally determined SLR rate in the entire range of the magnetic field. This discrepancy may be a result of a larger phonon density of states or a stronger spin-phonon coupling in a QD when compared to the bulk case assumed in Ref. [31] in derivation of Eq. (2). It may be also due to the fact that the dominant mechanism of coupling of the Mn^{2+} spin to the lattice is not a result of phonon-induced rotations of the axes of the local crystal field, but a result of local crystal deformations. Modeling of such coupling is, however, far more complicated and to the best of our knowledge was not considered in the literature so far.

The discrepancy between the calculation of the intrinsic single-Mn SLR and the experiment clearly shows that we cannot exclude the possibility that SLR is also mediated by another relaxation mechanism. We will discuss another possible mechanism, which is based on assumption that during the dark period the charge state of the QD actually exhibits fluctuations; i.e., for a certain fraction of time the QD is actually occupied by an electron captured from the surrounding bulk material. Since this mechanism involves an additional entity (a carrier appearing in the QD), we refer to it as the extrinsic one.

Due to the presence of s-d exchange interaction, $\hat{H}_{sd} = -A_e \overrightarrow{s} \cdot \overrightarrow{S}$ (where $\overrightarrow{s}$ is the electron spin operator), the eigenstates of a coupled electron-Mn complex are not the eigenstates of S_z, i.e., there is some mixing of states with various S_z projections caused by the electron. Considering only the $S_z = 5/2, 3/2$ states of the Mn^{2+} ion in an empty QD, when an electron with a random spin is captured, one of the four states $|\Psi_{S_z,s_z}\rangle$ become occupied. These states are labeled by their dominant spin component, i.e., we have $|\Psi_{5/2,1/2}\rangle \propto |5/2,1/2\rangle$, $|\Psi_{5/2,-1/2}\rangle \approx |a\rangle|5/2,-1/2\rangle + |b\rangle|3/2,1/2\rangle$ (with $|\langle a|a\rangle|^2 \gg |\langle b|b\rangle|^2$), etc., where $|S_z,s_z\rangle$ are eigenstates of S_z and s_z, and $|a\rangle$, $|b\rangle$ are the electron orbital states. The fact that the latter are not simply identified with the lowest-energy electron orbital in a QD is crucial for correct description of spin dynamics: the transitions considered below are possible only because either the spin-orbit interaction [33] or the flip-flop part of the s-d exchange interaction [34–36] are mixing the lowest-energy orbitals with the excited ones.

When a spin-up electron is captured in the QD with a Mn^{2+} ion in the $S_z = 5/2$ state, the only possible transition involving Mn^{2+} spin relaxation is from the resulting $|\Psi_{5/2,1/2}\rangle$ state to the $|\Psi_{3/2,1/2}\rangle \approx |a\rangle|3/2,1/2\rangle + |b\rangle|5/2,-1/2\rangle$ state, and this transition involves electron spin relaxation. The latter process is made possible by the spin-orbit interaction admixing spin-flipped states from higher orbitals to the nominal electron spin "up" and "down" states in the lowest energy orbital. The probability for such a transition scales with the spin splitting of the electron states Δ_e as Δ_e^2 due to the so-called Van Vleck cancellation [33]. The total transition rate acquires also a dependence on the energy transferred to the lattice, $\Delta(B) \approx g\mu_B B - A_e/2$, due to the density of states of bulklike phonons [which contributes $\Delta^2(B)$], the square of the relevant phonon-carrier coupling [which contributes $\Delta(B)$ or $1/\Delta(B)$ for deformation potential and piezoelectric couplings, respectively], and from the interorbital matrix element of the phonon interaction [which contributes another $\Delta^2(B)$]. Using the typical values of electron g factor $|g_e| \approx 0.4$ [37–39] and the s-d coupling $A_e \approx 0.1$ meV [25,37,40] we see that for all the B fields used in the experiment the electron-spin splitting in the presence of the Mn^{2+} spin in the 5/2 state is dominated by interaction with this spin, $\Delta_e \approx \frac{5}{2}A_e$. This means that the rate of $S_z = 5/2 \rightarrow S_z = 3/2$ transition caused by electron-spin relaxation scales as $\Delta^5(B)$ for deformation potential coupling, and as $\Delta^3(B)$ for piezoelectric coupling (a very transparent discussion of all the factors determining such scaling laws for spin-relaxation rates is given in Ref. [41]).

On the other hand, when a spin-down electron is captured and a $|\Psi_{5/2,-1/2}\rangle$ state is created, transitions allowed by the orbital mixing, caused by s-d interaction, may occur. For example, the $|\Psi_{5/2,-1/2}\rangle$ state is built mostly from the $|\emptyset\rangle|5/2,-1/2\rangle$ state (where $|\emptyset\rangle$ is the ground-state electron orbital), while the $|\Psi_{3/2,1/2}\rangle$ state contains admixtures of excited states of the form $|\varphi_{\text{exc}}\rangle|5/2,-1/2\rangle$, where $|\varphi_{\text{exc}}\rangle$ denotes the excited-state electron orbital. The amplitude of such an admixture is proportional to $A_e|\langle\emptyset|\varphi_{\text{exc}}\rangle|/[E(\varphi_{\text{exc}}) - E(\emptyset)]$ where $E(\varphi_{\text{exc}}) - E(\emptyset)$ is the energy difference between the excited orbital state and the ground state, which is about 30 meV for the first excited state in CdTe QDs [42]. Thus, such admixtures allow for a transition between invoked states via phonon scattering. The reasoning analogous to the previous case gives the same result for scaling of the Mn^{2+} spin-relaxation rate with $\Delta(B) \approx (g - g_e)\mu_B B - 2A_e$.

In both scenarios (i.e., spin-up and spin-down electron capture), with $A_e \approx 0.1$ meV, in the expected case of dominance of piezoelectric coupling the above mechanisms predict $\Gamma_{\text{SLR}} \propto B^3$ for $B > 2$ T, in agreement with our measurements. It should be clear that obtaining quantitative results (predictions for actual values of Γ_{SLR}) would require a rather realistic multiorbital calculation of electronic structure (with spin-orbit

and carrier-Mn interactions included) of the QD with a single Mn^{2+} ion. While attempts at solving this problem were made [36], we note that due to the strong dependence of the carrier-phonon scattering element on the size of the QD [32,33], a lack of precise estimation of the spatial extent of the carrier wave functions in a given QD can result in quite a serious discrepancy between calculated and observed rates.

Finally let us note that the above-discussed QD charge fluctuation based mechanism, which requires the charge to be captured and then ejected (possibly many times) during the dark period, has to become irrelevant when the relaxation time becomes shorter than the typical carrier dwell time. Γ_{SLR} measured at highest magnetic field is therefore caused by yet another mechanism.

V. SUMMARY

We have presented a purely optical study of the SLR of a single Mn^{2+} ion in a CdTe/ZnTe QD in a wide range of magnetic field. By taking advantage of a spatial confinement of the Mn^{2+} ion in the dot and its isolation from other magnetic ions in the sample we have studied the SLR mechanism without the influence of spin-spin interactions. This constitutes a significant progress with respect to the previous studies of bulk materials and quantum wells. The obtained dependence of the SLR rate on the magnetic field evidently suggests that the relaxation of the Mn^{2+} ion is caused by single-phonon processes. In order to theoretically describe our experimental results we have discussed two mechanisms of a single-Mn SLR: an intrinsic one, in which the relaxation of an isolated ion is mediated by the emission of a single acoustic phonon, and an extrinsic one involving the possibility of existence of QD charge state fluctuations during the dark period. In the latter case the single-Mn spin relaxation is actually caused by relaxation of a carrier-Mn complex. Although these models are clearly not complete, the results of theoretical calculations performed for both relaxation mechanisms qualitatively reproduce the main features of the magnetic field dependence of the SLR rate. We find a quantitative discrepancy between the experimental results and the predictions of the first model in which the coupling of the Mn^{2+} spin to the lattice is a result of phonon-induced rotations of the axes of the local crystal field. This discrepancy may be a result of a larger phonon density of states, a stronger spin-phonon coupling in a QD when compared to the bulk case assumed in calculations, or a different coupling mechanism between the Mn^{2+} ion and the lattice. Quantitative comparison of the extrinsic or more elaborate model with the experiment would require detailed knowledge of the properties of a given QD.

It is noteworthy that the generality of our method of the SLR measurement may also enable the study of more complex magnetic systems, for example small clusters of magnetic ions [43], which are known to play an important role in the SLR dynamics in magnetic materials [5].

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