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Photoluminescence Dynamics of CdSe Quantum Dot with Single Mn^{2+} Ion under Modulated Excitation

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A quantum dot with a single magnetic ion is a perfect model system to investigate spin dynamics of a magnetic ion embedded in semiconductor matrix. In this work we present results of spin dynamics studies of Mn^{2+} ion embedded in CdSe quantum dot, performed under modulated excitation of the dot. In particular, the relaxation time of the Mn^{2+} ion in high magnetic field was determined.

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1. Introduction

A quantum dot (QD) with a single magnetic ion is a simple, yet powerful system for investigation of the ion interaction with charge carriers as well as with the semiconductor matrix. So far, CdTe and InAs QDs with single Mn^{2+} ions were the only systems on which extensive studies were performed [1–4]. A problem with other systems of QDs containing magnetic ions was expected quenching of the exciton luminescence caused by the intra-ionic transitions [5]. However, it was recently shown that such a quenching is negligible in the case of single dopants, even if the intra-ionic transition energy is lower than the exciton energy [6]. This opened the possibility of optical studies of a wide range of new systems on which ion–carriers interaction can be examined.

In this work we investigate one of such systems — a CdSe QD with a single Mn^{2+} ion. We use the photoluminescence (PL) measurements under modulated excitation [3, 6, 7] to investigate the Mn^{2+} spin dynamics. In particular, we determine the spin relaxation time without illumination of the dot in high magnetic field, as well as the spin depolarization process caused by the photocreated excitons confined in the dot.

In contrast to previous studies involving only one of the Mn^{2+} spin states [6], we obtain the full information on the evolution of the whole PL spectrum of the QD. This allows not only more detailed analysis of the Mn^{2+} spin evolution, but also provides full information on the behaviour of the QD PL background, improving the stability and reliability of the experiment.

2. Samples and experiment

The sample containing self-assembled CdSe/ZnSe QDs with single Mn^{2+} ion is grown by molecular beam epitaxy. During QDs formation a small amount of manganese is introduced, so that selection of individual dots containing exactly one Mn^{2+} ion is possible with the use of micro-PL setup. Detailed method of growth is given in Ref. [6].

Micro-PL studies are performed in an experimental setup described in detail in Refs. [8–10]. A liquid helium cryostat ($T = 1.78$ K) equipped with a superconducting split-coil magnet is used to produce high magnetic field ($B = 8$ T) applied in the Faraday configuration. Sample is attached to a microscope objective with high numerical aperture [11]. The PL of a single quantum dot is excited quasi-resonantly with a continuous wave argon laser at 488 nm. Modulated excitation is achieved by the use of acousto-optic modulator (AOM), which makes switching the excitation on and off possible. Light emitted from the QD is collected with monochromator equipped with gated CCD camera. The camera enables measurements with a 5 ns temporal resolution. The polarization optics in the detection path (half-wave and quarter-wave plates and a linear polarizer) allows the selection of one circularly polarized component (σ^-) of the QD PL.

3. Results

In the time-resolved experiment we exploit the effect of depolarization of the Mn^{2+} spin by the photocreated excitons, previously observed for the CdTe QDs with single Mn^{2+} ions [3, 12]. When the magnetic field is applied and the laser excitation is turned off, the Mn^{2+} spin relaxes to the thermalized state (oriented along the field) due to spin–lattice relaxation. When the excitation is switched on, randomly polarized excitons interact with the Mn^{2+} spin leading to the reduction of the mean Mn^{2+} spin projection ($\langle S_z \rangle$) onto the quantization axis given by the magnetic field. Therefore, by switching the excitation off and then on again after a controllable delay one can investigate both the Mn^{2+} relaxation process in darkness and depolarization by the photocreated excitons.

The CdSe QDs in our sample are usually negatively charged [6]. Therefore we use the negatively charged exciton (X^-) PL spectrum to monitor the spin state of the Mn^{2+} ion. Polarization of the Mn^{2+} spin in magnetic field results in strongly non-equal intensity distribution between the X^- PL lines. This is visible for excitation

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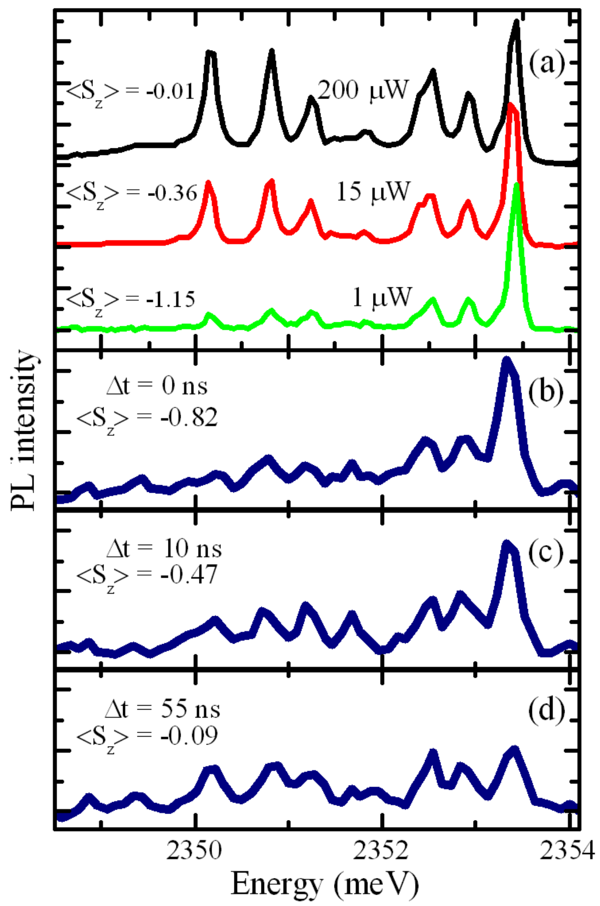

Fig. 1. (a) X^- PL spectrum of the QD with a single Mn^{2+} ion for indicated values of excitation power. (b)–(d) PL of X^- right after, 10 ns after, and 55 ns after switching the excitation on.

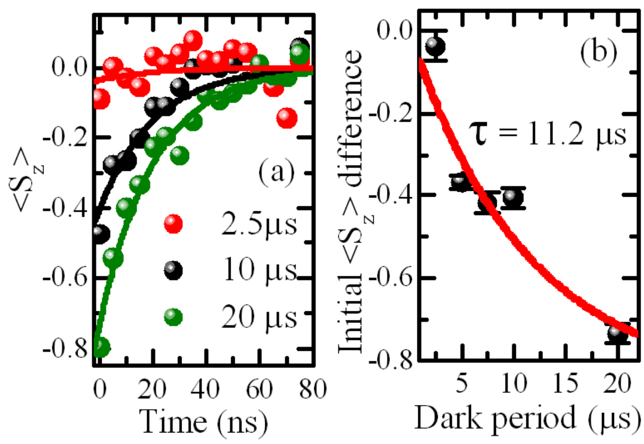

Fig. 2. (a) Evolution of the Mn^{2+} mean spin projection after switching the light on, fitted with exponential curve. (b) Mn^{2+} mean spin projection right after switching the light on versus length of dark period.

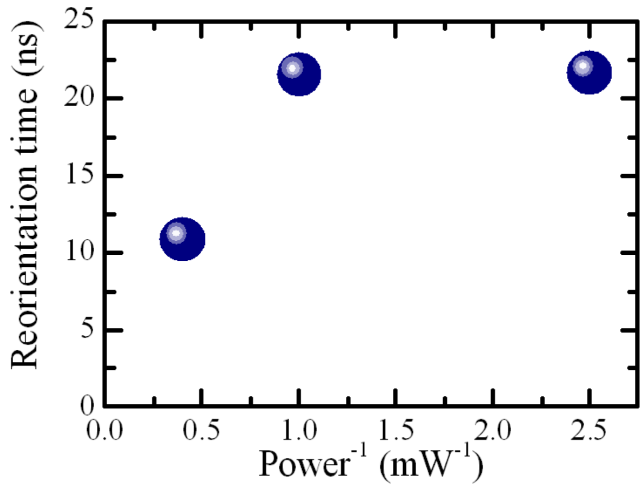

Fig. 3. Dependence of the Mn^{2+} reorientation time on the excitation power.

power low enough not to disturb the thermalized state (see Fig. 1a). With increasing excitation power the intensity distribution between the X^- lines becomes more equal, indicating depolarization of the Mn^{2+} spin. Due to complicated character of the X^-–Mn^{2+} interaction [13] and limited resolution of recorded spectrum the detailed analysis of the X^- fine structure is difficult. For simplicity, to estimate the $\langle S_z \rangle$ we assume that in σ^- polarized detection the lowest and the highest energy lines correspond to $+^5/_2$ and $-^5/_2$ Mn^{2+} spin state, respectively. We also assume that the lines in between correspond to states with linearly interpolated value of S_z.

As shown in Fig. 1a, $\langle S_z \rangle$ is close to 0 for high excitation power. Under such conditions, the X^- PL spectrum evolves after switching the excitation on, which is clearly visible in Fig. 1b–d. The measured evolution of $\langle S_z \rangle$ after different lengths of the dark period is shown in Fig. 2a. The initial $\langle S_z \rangle$ value plotted vs. the dark period length reflects the Mn^{2+} relaxation in darkness. As shown in Fig. 2b it follows the exponential decay, giving the Mn^{2+} spin–lattice relaxation time equal to 11.2 μs.

Simultaneously, the $\langle S_z \rangle$ evolution after switching the excitation on gives information on the depolarization of the Mn^{2+} ion by the photocreated excitons. As shown in Fig. 3, the depolarization time does not follow the linear dependence on the inverse of the excitation power, previously observed for CdTe dots with single Mn^{2+} ions [3]. This suggests that the depolarization mechanism is not the same in the case of the CdSe-based system. Further studies are needed to understand this behaviour.

4. Conclusions

Time-resolved PL measurements of a CdSe/ZnSe QD with a single Mn^{2+} ion under modulated excitation are used to investigate the Mn^{2+} spin dynamics in magnetic field. The spin–lattice relaxation time of the Mn^{2+} ion in darkness is equal to 11.2 μs, which is consistent with previous results obtained with the use of different experimental technique [3, 6, 14]. Surprising power dependence of the Mn^{2+} depolarization time requires further studies.

Acknowledgments

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