Ultralong Spin Memory of Optically Excited Single Magnetic Quantum Dots

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We study the magnetization dynamics in CdMnTe quantum dots using sub-wavelength optical microscopy imaging at B=0T. For continuous laser illumination each dot exhibits strong and unique circular polarization despite completely unpolarized ensemble emission. This implies that after an exciton recombines, the spontaneous ferromagnetic alignment of magnetic impurities persists for over 100 microseconds, which is million times longer than in CdMnTe quantum wells. The spin memory effect points towards qualitatively different picture of magnetization dynamics in zero-dimensional limit.

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Significant progress in the synthesis of nanostructures together with the ability to precisely control their morphology and chemical composition has opened previously unexplored research directions in basic and applied science [1-3]. As one of many remarkable achievements, it has been recently possible to incorporate magnetic impurities into semiconductor quantum dots (QDs) [4,5] and nanocrystals [6]. Such structures are ideal for studying magnetic interactions in the zero-dimensional limit [7,8], and at the same time are being actively considered as promising candidates for spintronics devices [9]. A central focus of these efforts is to understand the magnetization dynamics in these systems [10,11] and simultaneously develop feasible ways to control the spin properties of magnetic QDs [12].

Semiconductor II-VI QDs containing magnetic impurities can be divided into two classes, which, on a single dot level, exhibit distinct optical properties. The emission of a single CdTe QD with only one Mn ion (with angular momentum of 5/2) consists of a series of six narrow lines [8] attributed to doubly degenerated exciton levels coupled to six spin orientations of the Mn ion. In contrast, a single CdTe QD, where several percent of Cd cations are replaced with Mn, features a broad luminescence associated with exciton magnetic polarons (EMPs) [5,8,13]. The formation of EMPs, where magnetic ions are aligned within an exciton Bohr radius, is energetically favorable due to strong exchange interaction of the Mn spins and the electron-hole pair. In the case of CdMnTe quantum wells, the exciton must first be localized to an impurity or potential fluctuation before the EMP is formed. On the other hand, in magnetic QDs excitons are already strongly confined in all three dimensions and, in addition, the EMPs form always within the same dots with their position precisely defined through the spatial dot distribution. The strong and immediate electronic confinement should dramatically influence both the EMP formation and magnetization dynamics in the QDs.

Here we focus on the spin properties of EMPs localized in CdTe QDs doped with Mn. The results of simultaneous imaging of many QDs demonstrate that each dot within the ensemble features strong circular polarization at zero magnetic field. Remarkably, this spontaneous ferromagnetic alignment persists for over 100 microseconds, which is million times longer than in Mn-containing quantum wells. We conclude that the dominant demagnetization mechanism must be spin-lattice relaxation, and not spin-spin interactions. Such ultralong spin-memory effect renders these QDs a possibility to become an attractive system for manipulating local magnetic field on a nanometer scale as well as for designing circularly-polarizing single photon emitters.

The sample containing CdMnTe QDs was grown by molecular beam epitaxy [13]. A 4-μm-thick ZnTe buffer layer was grown first on (100)-oriented GaAs substrate at a temperature of 350°C. The ZnTe buffer was then exposed to Mn flux for approximately 4.5 s in order to incorporate Mn ions into the self-assembled dots formed by depositing 4 monolayers of CdTe by atomic layer epitaxy. Finally, a 50-nm thick ZnTe capping layer was grown. The average diameter and Mn concentration in the dots are 3-5 nm and about 2 percent, respectively [13] which translates into 5 to 20 Mn ions in each dot.

The results of low-temperature (T=6K) photoluminescence (PL) for the QD ensemble excited above the ZnTe barrier (2.39 eV) have not revealed any unique or unexpected behavior [13]: The overall shapes and intensities of PL spectra measured at zero magnetic field for two circular polarizations (σ^+ and σ^-) were identical within the limits of our measurement. Thus, there is no polarization for emission integrated from the large QD ensemble.

To understand this result, frequently observed for other magnetic QDs [4-6] and attributed to fast demagnetization of EMP via spin-spin interactions, it is important to draw a

distinction between global magnetization of the ensemble and local magnetization associated with a single magnetic dot. If the formation time of EMP is longer than the exciton lifetime, no EMPs form in a QD and both the local and global magnetization would be zero. On the other hand, even if EMP forms in a dot but the ferromagnetic alignment is not affected by the previous occupation, i.e. the magnetization decays before the dot is occupied again, then the time-averaged local (and global) magnetization of the dot also would be zero. This is the case for EMPs in CdMnTe quantum wells [14]. In contrast, the local magnetization of the dot averaged over time might be nonzero provided that the magnetization of the magnetic impurities decay slowly enough that some remnant magnetization is present when the dot is occupied by the next exciton. Such a spin memory effect would yield a significant polarization of the PL emission. Remarkably, also in this case, no global polarization of the ensemble would be observed, because the magnetization of each of the dot in the ensemble is oriented along random directions.

Simultaneous imaging of many dots allows us to directly measure the polarization of each individual dot within the distribution and sharply distinguish between the two above scenarios. We performed this experiment using slit-confocal imaging combined with a truncated solid immersion lens (SIL) [15,16] placed directly on the sample surface. In this way, with the 514 nm line of an Argon ion laser defocused to a 20 μm spot, a 5 μm x 250 nm area of the sample was imaged directly onto the spectrometer slit with 250 nm resolution. The PL was dispersed spectrally using a DILOR triple monochromator and the image was detected by a two-dimensional CCD matrix detector. Such an image contains the emission energies of QDs as well as their spatial positions along the slit [16]. The polarization of the PL emission was examined using a combination of a Babinet-Soleil compensator and a Glan-Thomson linear polarizer.

In Fig. 1 we display σ^+ and σ^- – polarized PL images taken subsequently for the same set of CdMnTe QDs at zero magnetic field. The vertical axis represents spatial position, while the horizontal axis represents the QD emission energy. Well-resolved spots in these images correspond to the emission from single CdMnTe QDs. The nearly perfect correspondence of the QD patterns between the two images confirms that we image the same subset of QDs for both circular polarizations. Despite unpolarized ensemble emission [13], individual QDs exhibit substantial circular polarization. In Fig. 1c we plot the PL spectra of four single QDs marked in Figs. 1a,b with blue and red curves representing σ^+ and σ^- polarization, respectively. While the emission of QD1 and QD2 is σ^+ – polarized and QD4 features σ^- – polarized emission, QD3 shows almost no polarization. We emphasize that high-resolution simultaneous imaging appears to be the only approach to unravel the complex behavior of the PL polarization, and thus spin alignment, of non-resonantly excited magnetic QDs.

The observation of circularly polarized PL emission of non-resonantly excited single CdMnTe QDs demonstrates that local ferromagnetic order of the EMPs at zero magnetic fields extends over many thousands of occupations. This strongly suggests that the formation of the EMP in QDs depends upon the previous occupations. Since, due to above-the-barrier excitation, the excitons are randomized before they are captured by the QDs, the EMP should form along directions determined by any small alignment of the magnetic impurities (presumably from spin fluctuations). We conclude that the strong polarization of single CdMnTe QDs must involve a spin-memory effect where the magnetization of the magnetic impurities persists after recombination of the exciton for a time long enough to provide a seed for formation of an EMP with the next occupation. In other words, the decay time of magnetization has to be much longer that an average time between consecutive occupations.

To make a simple estimate of this time, we note that for the detection of a typical QD emission line about 3000 photons are accumulated over 30 sec. Taking into account the 1% collection efficiency of the optics, the 10% throughput of the triple spectrometer and the 80% quantum efficiency of the detector, we estimate that the time between consecutive occupations for continuously illuminated sample is no less than 10 μsec. Therefore, in order to account for the measured single dot polarizations the relaxation time of the magnetization must be about 100 μsec, over six orders of magnitude longer than measured for CdMnTe quantum wells [14], where the remnant local magnetization rapidly decays through efficient spin-spin interactions. Such a long-lived magnetization implies that spin-spin relaxation in the QDs is inhibited and the demagnetization takes place predominantly via interaction with lattice vibrations [17].

The validity of this interpretation can be checked by turning off the excitation laser for a time sufficient for the magnetic impurities in the QDs to relax to a paramagnetic state, and then turning it on again. In this case, the polarization of individual dots should reflect no correlation with the previous image. In Fig. 2 we compare the PL polarization maps obtained for continuous laser excitation for over 40 minutes (left column) and chopped excitation with the laser blocked for 5 seconds and the maps taken immediately afterwards (right column). In the first configuration the polarization patterns of the dots feature essentially no change. In contrast, in the second case we observe complete randomization of the polarization of single CdMnTe QDs: The consecutive maps are dramatically different. This indicates that 5 second intervals are sufficient to randomize magnetization within a single QD.

This difference is displayed in Fig. 3, where we plot the value and sign of polarization measured for representative single QDs. Indeed, for continuous illumination the polarization varies within only 10-15% of its average value, while the polarization measured after 5 second

dark intervals exhibits huge changes, both in value and sign. Remarkably, for both dots there are iterations, where no net polarization is observed, similarly as in the case of QD3 in Fig. 1. Taken together, these measurements strongly support our interpretation that the EMP formation in CdMnTe QDs depends on the previous occupation. After the exciton recombines, the relaxation time of the ferromagnetically aligned magnetic ions is slow enough to present a non-zero magnetization to the next exciton occupying the dot.

We summarize by emphasizing that the experimental approach based on simultaneous imaging of many single quantum dots reveals spontaneous ferromagnetic alignment of magnetic impurities confined to CdMnTe QDs. The direction and magnitude of the magnetization vary from one dot to the other and are determined by local fluctuation of the spin system. This finding implies that the magnetic impurities within the dot remain polarized long after the exciton recombines. We estimate that the spin memory persists over tens of microseconds due to the inhibition of the normally dominant spin-spin relaxation processes. We believe that observation of such extremely stable magnetization makes this system particularly attractive for manipulating spin properties of nanostructures.

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Figure captions

Figure 1. (Color online) PL images of single magnetic QDs collected for (a) σ^+ and (b) σ^- polarizations at B=0. (c) PL spectra extracted for the four marked single QDs with blue (red) color representing σ^+ (σ^-) polarization.

Figure 2. (Color online) Left column: polarization images of the same set of single magnetic QDs obtained for continuous laser illumination. Right column: polarization images of the same set of single magnetic QDs obtained for chopped laser illumination with the laser being turned off for 5 seconds between two consecutive maps. Blue and red colors correspond to positive and negative polarization, respectively.

Figure 3. (Color online) (a) Representative time and iteration dependencies of single QD polarization for continuous (red) and chopped (blue) laser illumination. The distributions of measured polarization values in these two regimes are shown in (b) and (c), respectively.

Figure 1.

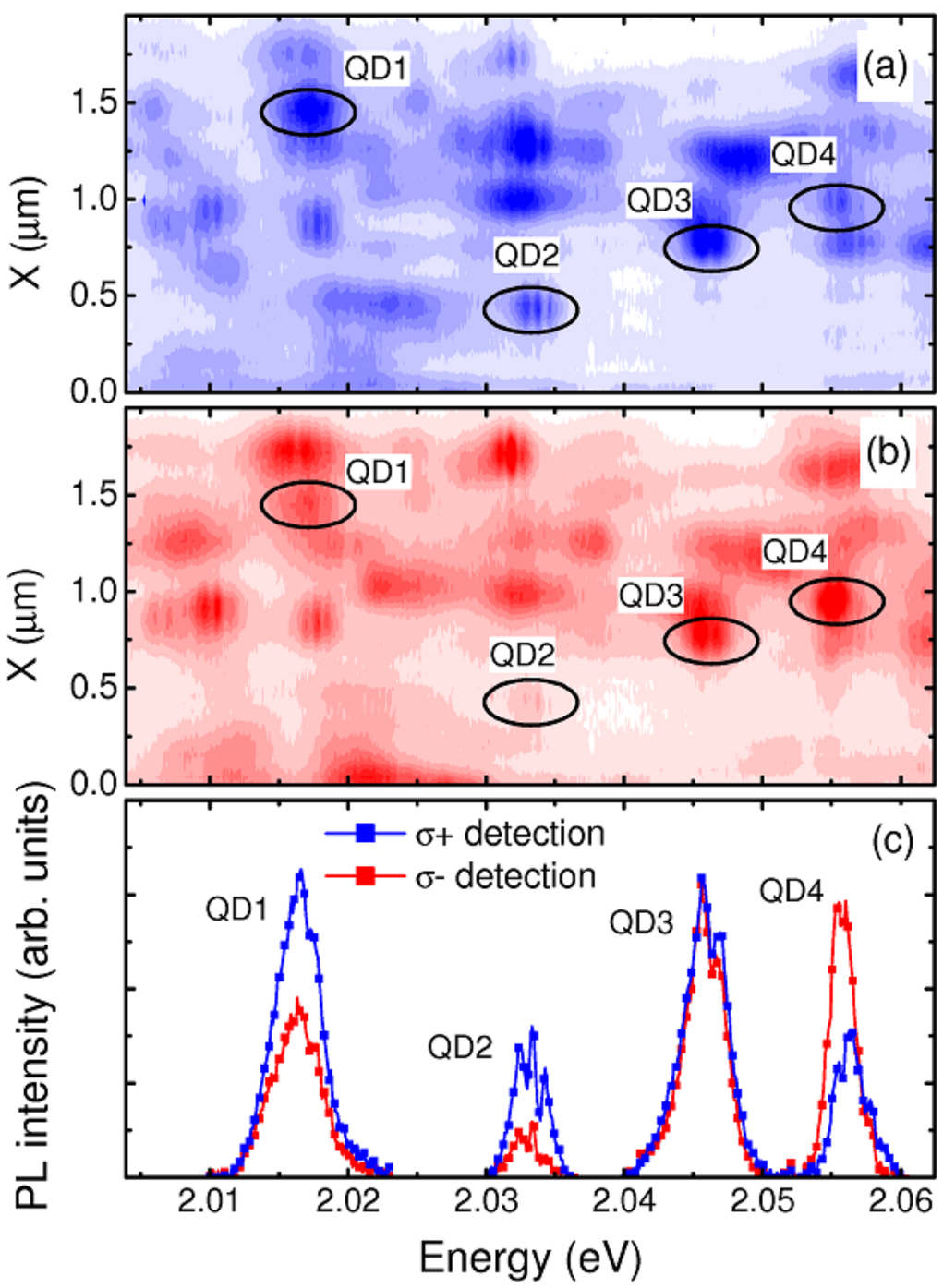

Figure 2.

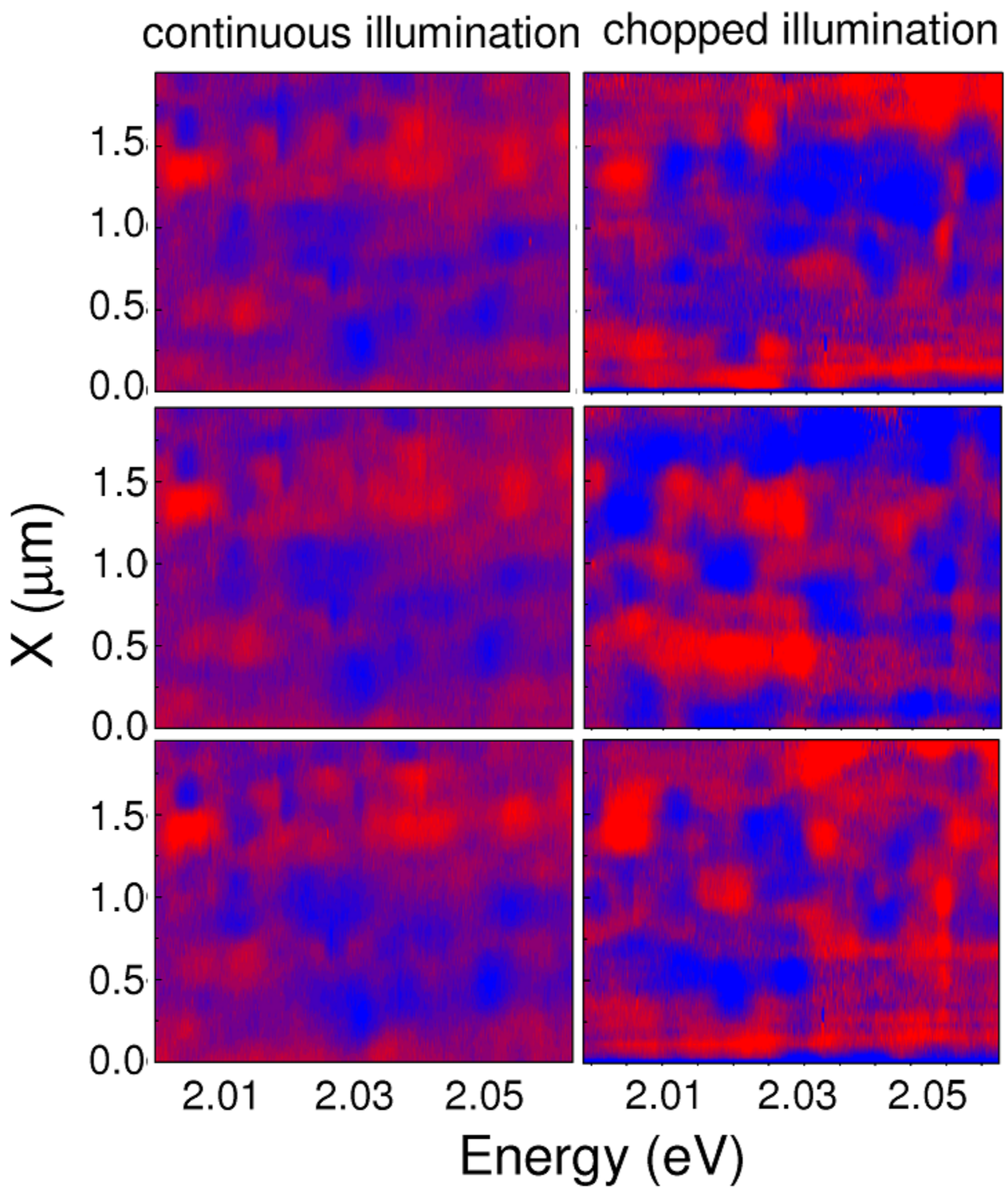

Figure 3.