Monitoring Statistical Magnetic Fluctuations on the Nanometer Scale

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Statistical fluctuations of the magnetization are measured on the nanometer scale. As the experimental monitor we use the characteristic photoluminescence signal of a single electron-hole pair confined in one magnetic semiconductor quantum dot, which sensitively depends on the alignment of the magnetic ion spins. Quantitative access to statistical magnetic fluctuations is obtained by analyzing the linewidth broadening of the single dot emission. Our all-optical technique allows us to address a magnetic moment of only $\approx 100\mu_B$ and to resolve statistical changes on the order of a few μ_B.

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Magnetic semiconductors hold promise for closing the gap between information storage and information processing in microelectronics. Indeed, a completely new research area—referred to as "spintronics"—has evolved around this idea [1,2]. The unique possibility to tailor electronic and magnetic properties independently has triggered a variety of visionary device concepts, based on controlling the interaction of carrier spins and the spins of magnetic ions [3,4]. However, as one proceeds to ultrasmall magnetic systems, there arises a *fundamental* limit for operating such devices: statistical magnetic fluctuations are expected to severely restrict the device functionality. In this Letter, statistical magnetic fluctuations have been analyzed in magnetic semiconductor single quantum dots (SQDs) by means of spatially resolved optical spectroscopy.

The first successful approaches to fabricate self-organized magnetic semiconductor QDs have quite recently been reported, incorporating Mn^{2+} ions into the crystal matrix of II-VI semiconductors [5,6]. This offers an interesting opportunity: due to the exchange interaction between the carriers and the magnetic ions, variations of the magnetization within the environment of a given dot is expected to be reflected by the PL signal of the quasi-zero-dimensional electron-hole pair bound by that dot. Here the advantage of studying magnetic semiconductor QDs instead of metallic nanomagnets becomes evident when it is realized that the sensitivity of spatially resolved magneto-PL spectroscopy is sufficient to address one SQD with typical dimensions in the 10 nm range [7]. Since the optical response of a SQD at low temperatures is spectrally quite narrow, even minute variations in the *magnetic* environment within the exciton wave function can be monitored, quite similar to what is known from *charge* fluctuations, which results in spectral diffusion and linewidth broadening [8].

We investigated self-assembled CdSe/ZnMnSe QDs dots prepared by molecular beam epitaxy. A nominal CdSe thickness of 2.5 monolayers and a Mn content of $x = 0.25$ have been chosen. SQD selection is obtained by defining lithographically a metal mask with apertures down to $\phi = 100$ nm in diameter on top of the samples [5]. Time-resolved PL spectroscopy reveals a recombination lifetime of about 580 ps, roughly 1 order of magnitude longer than that usually found in CdZnSe/ZnSe quantum wells [9]. The lifetime is found to be *temperature independent* in the temperature range under investigation. This gives clear proof of three-dimensional exciton confinement; i.e., the exciton is sharply localized within a size comparable to or even smaller than the 3D Bohr radius (about 4–5 nm). This is in good agreement with typical QD sizes found in the CdMnSe/ZnSe [6,10,11] systems. Second, the formation of an excitonic magnetic polaron (EMP), i.e., a ferromagnetically aligned spin complex consisting of one exciton and several hundred Mn^{2+} ions with strongly correlated spins, is found to occur on a time scale much faster than the recombination lifetime [12]. Thus, the QD PL spectrum reflects the equilibrium energy of the EMP, redshifted relative to the absorption peak by the EMP binding energy ($E_{\mathrm{MP}} \approx 17$ meV at a bath temperature of 2 K).

At first glance, one would expect that in the equilibrium case the EMP in a given SQD is characterized by a well-defined energy—i.e., the PL signal should be spectrally quite narrow. This is definitely not true, as is evident from Fig. 1, where for a bath temperature of 2 K spatially resolved PL spectra of magnetic CdSe/ZnMnSe QDs are compared to a nonmagnetic CdSe/ZnSe reference sample. Low excitation density was used in order to ensure that only one single electron-hole pair was generated per QD. In both samples, individual PL peaks can be resolved, each attributed to the recombination of an

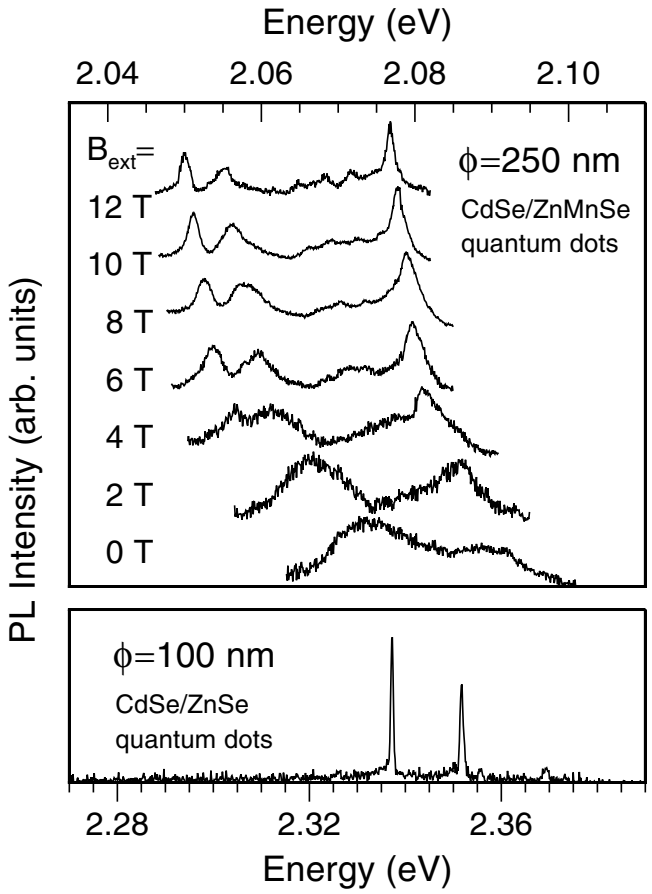

FIG. 1. Spatially resolved PL spectra for nonmagnetic (bottom) and magnetic (top) QDs. The upper part of the figure shows σ^+ polarized PL spectra of the CdSe/ZnMnSe SQDs at different magnetic fields.

electron-hole pair in a SQD. However, while for nonmagnetic SQDs the PL linewidth is limited by our spectral resolution of ≈ 0.4 meV, the individual emission peaks of the magnetic SQDs are characterized by a rather large linewidth of several meV. Moreover, the emission linewidth of the magnetic QDs drastically narrows when an external magnetic field is applied in Faraday geometry. This behavior, which is characteristic for all the SQDs studied, is obviously related to the influence of the magnetic ions located within the spatial extent of the exciton wave function. It should be noted that, for ensemble measurements on the magnetic QDs, the inhomogeneous broadening of the PL signal [full width at half maximum (FWHM) ≈ 44 meV] hides this effect and no magnetic-field dependence of the PL linewidth can be resolved.

We relate this finding—i.e., the large magnetic-field-dependent PL linewidth in magnetic SQDs—to statistical magnetic fluctuations [13]: as the SQD is being probed repeatedly, statistical variations of the magnetization within the exciton wave function result in a broadening of the SQD emission peak in time-integrated experiments. In diluted magnetic semiconductors, the magnetization M can be described by a modified Brillouin function [14]

$$M(B,T) = xN_0 g\mu_B S_{\text{eff}} B_{5/2}\left(\frac{5\mu_B gB}{2k_B T_{\text{eff}}}\right) \tag{1}$$

where $g = 2$ is the g factor of Mn^{2+} ions, and N_0 is the number of cations per unit volume. The effective spin $S_{\text{eff}} < 5/2$ and the effective temperature $T_{\text{eff}} = T + T_0$ take into account the antiferromagnetic interaction between neighboring Mn^{2+} spins [15]. The field $B = B_{\text{ext}} + B_{\text{MP}}$ includes both the external field B_{ext} and the exchange field B_{MP} between the charge carriers and the spins of the magnetic ions [5,16]. Because of the sp-d exchange interaction between the three dimensionally confined carriers and the spins of the Mn^{2+} ions, we are in the unique situation that the energy $E(B,T)$ of the PL signal from a SQD depends directly on the magnetization $M(B,T)$ within the exciton wave function. We can then write

$$\begin{aligned} E(B,T) - E_0(T) &= -\frac{\gamma(\alpha-\beta)}{2\mu_B g} M(B,T) \\ &= -B_{\text{MP}} V_{\text{eff}} M(B,T), \end{aligned} \tag{2}$$

where α and β are the well-known exchange constants of the electrons and holes, respectively, and $E_0(T)$ includes the temperature dependence of the band gap. The effective volume is given by $V_{\text{eff}} = \gamma V$, where V is the volume occupied by the exciton wave function, and γ (which is less than unity) takes into account the fact that only a part of the exciton wave function actually overlaps with the Mn^{2+} spins [17].

The distribution $\Phi(\mathbf{M})$ of the magnetization $\mathbf{M}$ within the single exciton wave function can be written in Gaussian form

$$\Phi(\mathbf{M}) = \frac{1}{(2\pi\langle M^2\rangle)^{3/2}} \exp\left(\frac{|\mathbf{M} - \langle\mathbf{M}\rangle|^2}{2\langle M^2\rangle}\right). \tag{3}$$

The characteristic width (FWHM) of this distribution $\Delta M = \sqrt{\langle M^2\rangle 8\ln 2}$ is directly related to the SQD emission linewidth (FWHM) ΔE via the sp-d exchange interaction. As our dots are most likely disk-shaped (in accordance to what is known from TEM measurements in the similar system CdSe/ZnSe), the in-plane g factor of the heavy hole is expected to be small. Being aware that the exchange interaction between the carrier spins and the magnetic ions is dominated by the heavy hole-Mn^{2+} spin interaction, we monitor the longitudinal magnetic fluctuations, as the magnetic field is applied in Faraday geometry. Using the well-known fluctuation-dissipation theorem $\langle M^2\rangle = (dM/dB)k_B T/V_{\text{eff}}$, one obtains [18]

$$\begin{aligned} \Delta E(B,T) &= B_{\text{MP}} V_{\text{eff}} \sqrt{\langle M^2\rangle 8\ln 2} \\ &= \sqrt{B_{\text{MP}}\, 8\ln 2\, k_B T \left(-\frac{dE}{dB}\bigg|_{B=B_{\text{ext}}+B_{\text{MP}}}\right)}. \end{aligned} \tag{4}$$

This leads us to a very fundamental result: ΔE is proportional to $\sqrt{\langle M^2\rangle}$; i.e., the optical response of the SQD is directly correlated to the statistical magnetic fluctuations on a scale defined by the spatial extent of

the exciton wave function. As B_{MP} is inversely proportional to the localization volume of the exciton [19], ΔE does depend on the volume of the localized exciton and decreases as $V^{-1/2}$, if one approaches large volumes. Note that applying the magnetic field in Voigt geometry would trace the transverse magnetic fluctuations, which cannot be efficiently suppressed in an external field. In fact, preliminary experiments in Voigt geometry confirm this picture.

If the emission linewidth is primarily controlled by longitudinal magnetic fluctuations, ΔE should be proportional to $(-dE/dB)^{1/2}$ [see Eq. (4)]. As the emission peaks narrow with increasing field, the optical response of individual QDs can be easily resolved, and the Zeeman shift (and therefore dE/dB) can be determined very accurately. Furthermore, at a fixed temperature, the proportionality factor between ΔE and $(-dE/dB)^{1/2}$ is given by the internal exchange field B_{MP}, thus providing an experimental handle for determining B_{MP}. Using this procedure, we obtain values between $B_{MP} \approx 2.3$ T and $B_{MP} \approx 3.2$ T varying from dot to dot due to variations in the dot size and/or composition [20]. In Fig. 2 we plotted ΔE (we took the FWHM of a Gaussian line shape fit) as a function of $(-B_{MP} \times dE/dB)^{1/2}$ for several QDs. The proportionality between these two quantities for all the SQDs indicates that the emission linewidth of the magnetic SQDs is primarily controlled by statistical magnetic fluctuations. It is worth noting that the relation between B_{MP} and V allows a rough estimate of the spatial extent of the exciton wave function. For example, $B_{MP} = 3$ T corresponds to $V \approx 80$ nm^3. Taking the exciton volume as a first approximation for the dot size, this would result in a dot diameter of about 7 nm if one assumes a dot height of 2 nm. This is in good agreement with both the values usually found by TEM for the CdSe/ZnSe system [10,11] and with the conclusions drawn above from our time-resolved experiments.

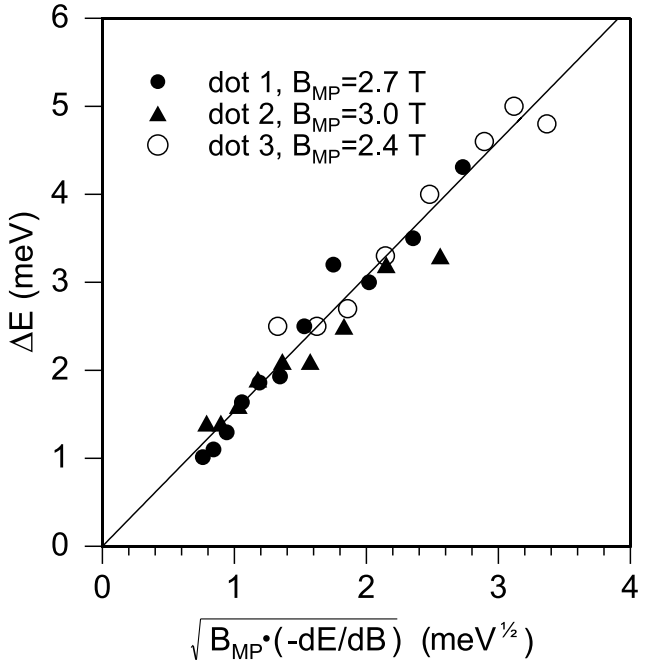

FIG. 2. Emission linewidth ΔE plotted for several SQDs (different symbols) as a function of $(-B_{MP} \times dE/dB)^{1/2}$. The experimental data are obtained at a bath temperature of 2 K. The solid line represents a fit according to Eq. (4).

In the insets of Fig. 3, the Zeeman shift for two SQDs with strongly different PL energies is depicted. The solid lines represent fits using Eq. (2) with B_{MP} extracted from the data of Fig. 2, showing rather excellent agreement with the experiment. We are thus in a position to describe quantitatively the linewidth broadening due to magnetic fluctuations. This can be seen in Fig. 3, where ΔE is plotted for the same two SQDs as a function of B_{ext}. It is evident that the value of the PL linewidth as well as the magnetic-field-induced linewidth narrowing, which is ascribed to the suppression of magnetic fluctuations, is quantitatively reproduced by our model without any additional parameters. A quite similar behavior is found for all the SQDs investigated. Note that even for high magnetic fields the linewidth exceeds the values obtained for nonmagnetic SQDs, most likely due to an incomplete saturation of the magnetization. Our model predicts an increase of the PL linewidth with temperature—and this is exactly what we find in experiment, as can be seen in the inset of Fig. 4. The linewidth broadening cannot be

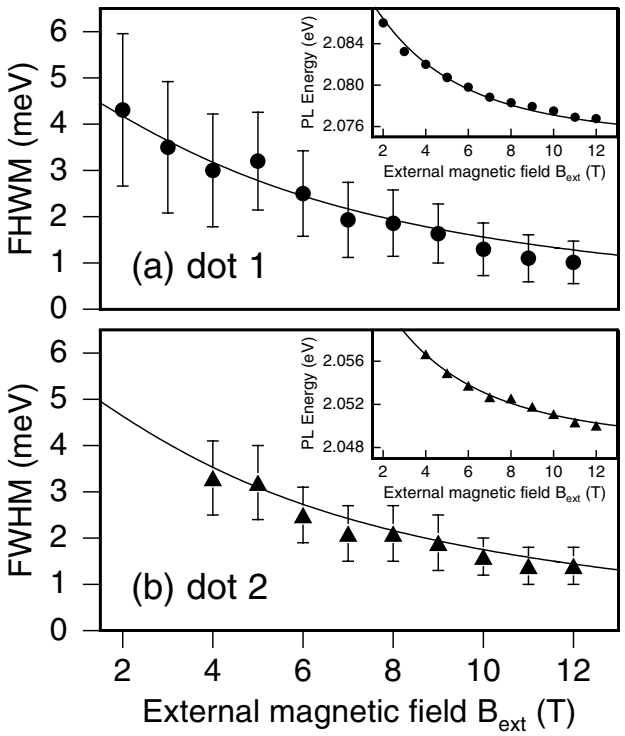

FIG. 3. PL linewidth ΔE as a function of external magnetic field B_{ext} for two SQDs with strongly different PL energies. The insets show the magnetic-field-induced energy shifts of the PL lines for the same SQDs. Symbols correspond to experimental data, solid lines to calculations according to Eqs. (2) and (4).

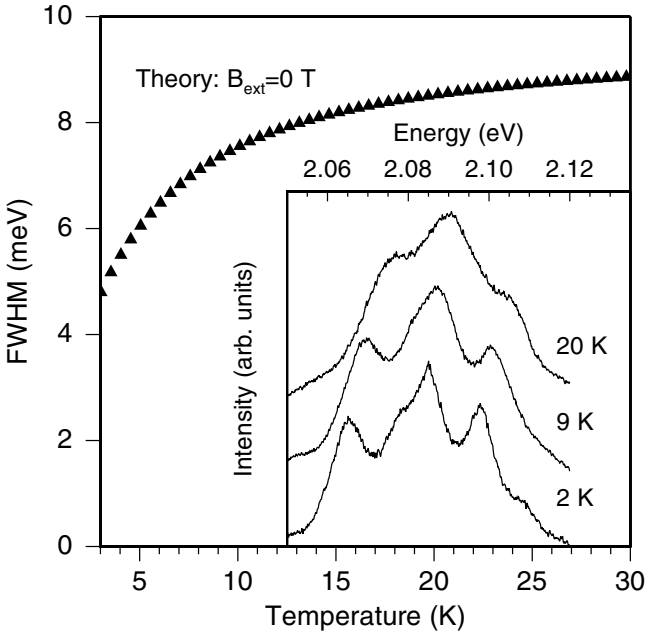

FIG. 4. Temperature dependence of the PL linewidth as expected from theory. The inset shows SQD PL spectra for different temperatures obtained with an aperture of $\phi = 175$ nm.

attributed to phonon scattering, as shown by comparison with data obtained on nonmagnetic SQDs. For example, from our calculations using the same parameters as above, we obtain an increase of the linewidth of about 50% when the temperature increases from 5 to 20 K (see Fig. 4), in rather good agreement with experimental data. However, a detailed quantitative comparison between experiment and theory is quite difficult, as the strong broadening at $B_{\text{ext}} = 0$ leads to a significant spectral overlap of the lines emitted by different SQDs within the aperture area (see inset).

In conclusion, we demonstrate that the PL linewidth from a single magnetic semiconductor QD can be used to analyze statistical magnetic fluctuations on a scale of a few nanometers. Our results are based on the fact that the EMP formation time is much smaller than the recombination lifetime. Otherwise, the transient energy shift during EMP formation as well as memory effects due to an incomplete magnetic relaxation between two recombination events have to be taken into account when discussing the linewidth of optical transitions in magnetic SQDs [17]. Moreover, the electronic localization of the exciton must not change with magnetic field and temperature, which is fulfilled for the magnetic field and temperature range under investigation [12]. We wish to note that investigating a magnetic SQD corresponds to detecting a magnetic moment of only $\mu \approx 100\mu_B$, as estimated using the relation $\mu \approx E_{\text{MP}}/B_{\text{MP}}$. The linewidth broadening at low temperatures corresponds to fluctuations of $\pm 20\mu_B$ at $B_{\text{ext}} = 0$, which are reduced to about $\pm 3\mu_B$ at $B_{\text{ext}} = 12$ T—i.e., we are able to resolve magnetic moment changes in the magnetic ion ensemble of only a few μ_B. We expect that our results are of importance not only for devices based on magnetic semiconductor nanostructures, but also for applications of metallic nanomagnets [21,22], or other concepts involving magnetic nanoscale structures.

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