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Size dependence of electron spin dephasing in InGaAs quantum dots

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We investigate ensemble electron spin dephasing in self-assembled InGaAs/GaAs quantum dots (QDs) of different lateral sizes by employing optical Hanle measurements. Using low excitation power, we are able to obtain a spin dephasing time T_2^* (in the order of ns) of the resident electron after recombination of negative trions in the QDs. We show that T_2^* is determined by the hyperfine field arising from the frozen fluctuation of nuclear spins, which scales with the size of QDs following the Merkulov-Efros-Rosen model. This scaling no longer holds in large QDs, most likely due to a breakdown in the lateral electron confinement. © 2015 AIP Publishing LLC.

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The endeavor of bringing spin functionalities to conventional solid state devices has been viewed as an attractive route to combine information processing relying on control of charge with information storage built on orienting magnetic domains and information communications utilizing photons, as well as preliminary steps towards realization of spin-based quantum computing.^{1–4} In this context, semiconductor quantum dots (QDs) are promising since major spin relaxation processes via spin-orbit (s-o) interactions are suppressed in QDs due to the three-dimensional carrier confinement leading to a long electron spin relaxation time exceeding ms in the QD ground state.^{5,6} Moreover, the fact that most of these zero-dimensional materials systems (III–V QDs for instance) are highly optically active has made them even more appealing as they provide optical accessibility of spins that not only facilitates studies of spin dynamics but also oversees innovations in photonic and optoelectronic applications. During the last two decades, there has been great advance in our understanding of fundamental spin properties of QDs that are also highly relevant to spintronics and quantum information technology. Innovative devices like spin-LEDs,⁷ spin filters,⁸ and spin memory devices⁹ have been fabricated based on QDs. Coherent manipulation of spins has been demonstrated by various means in both electrostatic¹⁰ and optically active QDs.¹¹ Large scale production of III–V self-assembled QDs has been demonstrated, with the capability of being integrated with the main-stream silicon technology,¹² suggesting that a wide range of applications of QD devices may soon be possible.

Despite an expected long electron spin lifetime, however, it has become clear that an average electron spin dephasing time T_2^* in QD ensembles is rather short due to the hyperfine interaction between electron and nuclear spins. A short spin dephasing/decoherence time is undesirable for QD-based spin information processing as it limits the maximum number of quantum error-correction protocol¹³ such that unavoidable errors can be detected and compensated. This has motivated in-depth studies that aim at a better understanding of the dominant physical mechanism

responsible for spin dephasing such that an effective strategy to prolong T_2^* can be devised. The role of a fluctuating nuclear field (FNF) on electron spins in QDs was discussed theoretically within the frame work of the Merkulov-Efros-Rosen (MER) model,¹⁴ which suggested that the dominant factor governing T_2^* is the spin precession induced by the frozen fluctuation of the hyperfine nuclear field. Experimental evidence for the validity of the MER model was provided in self-assembled GaAs/GaAlAs and InGaAs/GaAs QDs, where optically generated electron-spin polarization decreased to 1/3 of its initial value within a characteristic time of ~ 1 ns.¹⁵ This was attributed to the fact that 2/3 of the randomly distributed frozen FNF was oriented in the plane perpendicular to the electron spin direction that caused spin dephasing, whereas the remaining 1/3 of the frozen FNF was aligned parallel to the spin direction and thereby did not lead to electron spin dephasing.¹⁴ Another prominent feature of the spin dephasing induced by the FNF is that T_2^* is closely related to the number of interacting nuclei and thus to the size of QDs. A further verification of the MER model and a better understanding of spin dephasing processes therefore demand systematic experimental investigations of the dependence of T_2^* on QD sizes, which is still lacking. In this work, we intend to carry out such a study by examining T_2^* in a set of self-assembled InGaAs/GaAs QDs with a fixed height but different lateral sizes. Through this study, we also hope to shed light on the limit of QD sizes within which the scaling between T_2^* and QD size still holds.

The studied Stanski-Krastanov (SK) grown $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}$ /GaAs QDs was prepared by molecular beam epitaxy (MBE) on a semi-insulating (001) GaAs substrate, with a 350–400 nm thick GaAs buffer layer. Two or three layers of QDs were prepared under the same conditions, of which the topmost layer is uncapped for topographic measurements of the QDs. By carefully controlling and varying the growth conditions, QDs with a small size deviation and different lateral diameters were fabricated. They are of a typical height of 3.5 nm and an average diameter ranging from 23.3 nm to 100.9 nm. They are nominally undoped, though common contamination by C_{As}

FIG. 1. (a)–(c) PL and (d)–(f) P_{PL} spectra measured from the QDs of different average lateral diameters d , under optical excitation above E_g^{GaAs} (at 800 nm). The P_{PL} spectra were obtained under the σ^+ (the thick lines) and σ^- (the thin lines) excitation. The maximum counter-polarization point is marked by a dash line for each sample as the detection photon energy in the Hanle measurements.

acceptors in GaAs is evident from the observation of the related free-to-bound optical transition. Optical orientation spectroscopy was performed at 4 K under a non-resonant excitation condition with a photon energy above the GaAs bandgap energy (E_g^{GaAs}). The optical excitation was done by using a Ti-sapphire laser beam of circular polarization that alternated between σ^+ and σ^- at a frequency of 50 kHz by means of a photo-elastic modulator (PEM), to avoid dynamic nuclear spin polarization of the QDs.¹⁵ The excitation light was directed along the direction normal to the sample surface. The resulting photoluminescence (PL) was spectrally resolved by a single grating monochromator and collected by a liquid-nitrogen-cooled Ge detector, in a backscattering geometry. The degree of circular polarization of the PL emission under either σ^+ or σ^- excitation, $P_{PL} = (I^{\sigma^+} - I^{\sigma^-}) / (I^{\sigma^+} + I^{\sigma^-})$, from the exciton ground state of the QDs was analyzed by a rotatable broadband quarter-wave plate in conjunction with a linear polarizer. Here, $I^{\sigma^\pm}$ refers to the intensity of the circularly polarized PL component (σ^+ or σ^- as given in the superscript). Hanle measurements were carried out by measuring P_{PL} at a selected emission energy as a function of an applied transverse field (B_T), i.e., a field perpendicular to the light path as well as the spin direction of the optically oriented electrons.

Fig. 1 shows typical PL and P_{PL} spectra from the studied QD samples. The observed PL bands are typical for QD ensembles and are due to an inhomogeneous broadening effect introduced by a statistical distribution in the QD height as well as variations in the chemical composition and strain. A distribution in the lateral diameter of the QDs plays a negligible role in the PL linewidth of our QDs, due to a large aspect ratio between the lateral diameter and the height.¹⁶ Over the entire PL band of each QD sample, P_{PL} exhibits a strong spectral dependence, which we recently showed to arise from a strong spectral overlap between the positive X^+ and negative X^- trion emissions of opposite optical polarizations.¹⁷ The X^+ , leading to the co-polarized PL on the high-energy side, is favored when holes from the residual C_{As} acceptors in GaAs are transferred to the QDs. This occurs when optical excitation energy is below E_g^{GaAs} or at a low excitation power under excitation above E_g^{GaAs} . The X^- giving rise to the counter-polarized contribution on the low-energy side, on the other hand, is promoted under strong excitation above E_g^{GaAs} , due to a combined effect of neutralization of the ionized C_{As} acceptors in GaAs and more efficient injection of more mobile electrons than holes to the QDs.¹⁷

To determine electron spin dephasing times in the studied QDs, we resorted to Hanle studies of electron spin polarization under optical orientation conditions. In Hanle measurement, see Fig. 2(a), B_T drives electron precession around the applied field with a field-dependent Larmor frequency determined by $\Omega = g_e \mu_B B_T / \hbar$. Here, g_e is the electron g factor, μ_B is the Bohr magneton, and $\hbar$ is the reduced Planck constant. Within the limit of a spin lifetime T_s , accelerating spin precession with increasing magnetic field reduces electron spin polarization as well as $|P_{PL}|$. The resulting Hanle curve follows a Lorentzian lineshape¹⁸

$$|P_{PL}(B_T)| = \frac{|P_{PL}(0)|}{1 + (B_T/B_{1/2})^2}. \quad (1)$$

FIG. 2. (a) Typical Hanle measurement geometry to deduce T_s^* for the QD samples. (b)–(d) Experimental Hanle curves (the symbols) obtained under optical excitation at two different power levels. The solid lines are the fitting curves for the co- and counter-polarized Hanle components, whereas each dashed line is the sum of the co- and counter-polarized Hanle components. (e) Effective electron spin lifetime $g_e^* T_s$ as a function of PL intensity. The saturation values marked by the dashed correspond to $g_e^* T_s^*$ for the specified QDs.

T_s can be determined from the half-width-at-half-maximum (HWHM) $B_{1/2}$ through the well-known relation $g_e^* T_s = \hbar / (\mu_B B_{1/2})$. Note that T_s is the ensemble spin lifetime, and g_e^* is the effective g-factor representing the given QD ensemble. Several events can interrupt the spin precession and contribute to T_s , including a decay process (characterized with a time constant τ) and spin relaxation (τ_s) due to spin flips (T_1) and spin dephasing (T_2^*) with $\tau_s^{-1} = T_1^{-1} + T_2^{*-1}$. They are linked to T_s with a simple relation $T_s^{-1} = \tau^{-1} + \tau_s^{-1}$.

Typical Hanle curves obtained from the QD samples are presented in Figs. 2(b)–2(d), under optical excitation above E_g^{GaAs} to enhance the contribution from X^- as mentioned above. Here, we choose to detect at the emission energy where the maximum degree of the counter-polarization from X^- was observed. Hanle curves at different excitation power levels were obtained at a fixed detection energy for a given sample, as indicated by the dashed lines in Fig. 1, as the maximum counter-polarization point is rather insensitive to excitation power.¹⁹ The reason why X^- is particularly suitable here is that while depolarization of the electron spin in X^+ is limited by a short recombination time τ of the trion, depolarization of the electron in X^- takes place in the resident electron at the QD ground state after the trion recombination. The lifetime τ of this resident electron is only limited by re-capturing of an electron-hole pair to form X^- , which can be conveniently tuned by excitation power and thus the number of available e-h pairs. With a low excitation power, the capture process can be significantly slowed down resulting in τ that can be orders of magnitude larger than the spin relaxation time τ_s , such that T_s is solely governed by the latter. This is indeed the case for the counter-polarized component of the Hanle curves, as shown in Figs. 2(b)–2(d). With a high excitation power, a single component of the counter-polarization is resolved that can be assigned to optical depolarization of X^- . For examples, as shown in Fig. 2(b), the HWHM (~ 307 G) of the Hanle curve obtained at 12 mW from the QDs with a diameter of 23.3 nm corresponds to an effective spin lifetime ($g_e^* T_s$) of 370 ps, which is predominantly determined by the lifetime τ of the resident electron. As excitation power is reduced, the HWHM of this counter-polarized component significantly reduces to 210 Gauss signifying an increasing value of $g_e^* T_s$ (A co-polarized component also appears at the low excitation power, with a much broader HWHM of ~ 970 Gauss that can be ascribed to X^+ due to the favorable condition as discussed above.¹⁷ By fitting the experimental Hanle curve by a sum of two Lorentzian lines using Eq. (1) with opposite signs, $B_{1/2}$ can be obtained for both co- and counter-polarized components as shown by the dashed lines in Fig. 2.). From a plot of the value of $g_e^* T_s$ determined from the Hanle curves as a function of PL intensity, which scales with excitation density and directly reflects the density of e-h pairs captured by the QDs, a clear trend can be observed for all of the studied QD samples as shown in Fig. 2(e): $g_e^* T_s$ continuously rises with the decrease in PL intensity as a result of a prolonged τ of the resident electron in the ground state of the negatively charged QDs. $g_e^* T_s$ eventually reaches a saturation value, marked by the dashed line in Fig. 2(e), when a further reduction of the excitation power no longer has an effect. This corresponds to the situation when $g_e^* T_s$ is no longer governed by τ

but instead by τ_s , such that $g_e^* T_s = g_e^* \tau_s$. Since T_1 is often found to be in the order of $\sim$ ms,⁹ which is much larger than T_2^* , τ_s is mainly determined by T_2^* .

To experimentally determine the dependence of $g_e^* T_2^*$ on QD sizes, detailed Hanle studies following the aforementioned procedure were conducted in the samples of different QD sizes. To obtain values of the QD volumes, we averaged atomic-force-microscopy (AFM) profiles of uncapped single dots in each sample and took into account the fact that the optically active QDs were the buried ones with a truncated top and an actual height of around 3.5 nm as measured by the high-angle annular dark field scanning transmission electron microscopy. As shown in Fig. 3, $g_e^* T_2^*$ increases with the increase in QD size, experiencing a change in its value by close to 3 times from the smallest dots to the largest ones. Earlier studies have shown that, though the electron g-factor in InGaAs/GaAs self-assembled QDs is dependent on emission energies²⁰ and therefore on QD sizes, this size dependence is largely negligible.²¹ As the estimated dispersion of the electron g-factor in our QD structures is less than 8%,²⁰ it is reasonable to assume that the variation in g_e^* plays a minor role in the observed large change in $g_e^* T_2^*$. A g_e^* value of -0.8 is adopted here, which is commonly accepted in the literature for InGaAs/GaAs QDs with a similar emission energy as that in our samples.^{20,21} Following the MER model, the ensemble spin dephasing time governed by the FNF can be calculated¹⁴ by

$$T_2^* = \hbar \sqrt{\frac{3V_L}{2v_0 \sum_j I^j (I^j + 1) (A^j)^2}}. \quad (2)$$

Here, V_L is the volume of electron localization, which characterizes the extension of the envelope part of the ground state wavefunction. v_0 is the volume of the two-atom unit cell. The summation runs over all atoms in the unit cell with nuclear spin I^j and hyperfine interaction constant A^j . By referring to the reported hyperfine constants $A^{Ga} = 38 \mu\text{eV}$ ($I^{Ga} = 3/2$), $A^{As} = 46 \mu\text{eV}$ ($I^{As} = 3/2$), and $A^{In} = 56 \mu\text{eV}$ ($I^{In} = 9/2$)²² and taking V_L to be the average QD

FIG. 3. Measured effective spin dephasing times $g_e^* T_2^*$ (the filled squares) as a function of the QD sizes, together with the predicted dependence based on the MER model (the dashed line). a_B is the Bohr radius in $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}$. The AFM images of two selected QD samples are also shown.

volume, the dependence of $g_e^*T_2^*$ on QD sizes can be calculated based on Eq. (2) without any adjustable parameters and is shown by the dashed line in Fig. 3. The calculated relation is in good agreement with the experimental data for small QDs, confirming the validity of the MER model in these QDs. From Fig. 3, it can also be clearly seen that the calculations start to significantly deviate from the experimental values for large QDs when the lateral diameter exceeds 3–4 times of the Bohr radius. This discrepancy is unlikely to be caused by a distribution of In composition commonly occurring along the growth direction in InGaAs QDs.^{23,24} This is because electron localization along the growth direction is not significantly altered in our QDs since the QDs' height is considerably smaller than the electron Bohr radius, such that the overall In composition and the total number of atoms the electron interacts are largely unaffected. The discrepancy cannot be accounted for by fluctuations in In composition or strain, even assuming their upper-bound values.¹⁹ Instead, it most likely stems from a breakdown in the lateral electron confinement in the large QDs such that: (i) the effective volume of the electron localization is no longer restricted by the QD size and (ii) electron dephasing mechanisms via s-o interactions begins to gain importance due to an increasingly quasi-2D character, yielding a shorter spin lifetime than that predicted by the MER model.

In summary, we have studied electron spin dephasing as a function of QD sizes in a series of nominally undoped InGaAs/GaAs QDs. By performing Hanle measurements on the X^- PL emission, electron spin dephasing time T_2^* has been determined. We found T_2^* to be strongly dependent on the QD sizes, which confirmed the description of electron spin dephasing by the frozen fluctuations of the hyperfine field according to the MER model for the QDs with a lateral diameter smaller than ~ 3 –4 times of the Bohr radius. For the larger QDs, the scaling between T_2^* and QD size no longer holds. Our results shed light on a critical QD size beyond which the lateral electron confinement breaks down and more efficient s-o mediated spin dephasing/relaxation processes can be activated, which can severely undermine the potential of the QDs for future applications in spintronics and spin-based quantum information technology. This work thus provides a useful guideline on the design of QDs for optimal spin functionality.

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