Electron spin relaxation in a single InAs quantum dot measured by tunable nuclear spins

X. M. Dou, B. Q. Sun,* D. S. Jiang, H. Q. Ni, and Z. C. Niu
SKLSM, Institute of Semiconductors, CAS, P.O. Box 912, Beijing 100083, China
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The electron-spin dynamics of positively charged excitons in a single InAs quantum dot (QD) were measured by time-resolved photoluminescence spectroscopy. Using alternating σ^+/σ^- pulse sequences to excite the QD, the lattice nuclear spins remained randomly oriented. This method enables us to check the theory of electron-spin relaxation in the randomly distributed frozen fluctuation of the nuclear field. The experimental results are in qualitative agreement with theoretical prediction, showing that the electron-spin polarization decreases to a minimum value at first, and then increases again up to a steady value of about 1/3 of its initial value.

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Self-assembled single quantum dots (QDs) are of great interest in current research because of their promising applications in spin-based quantum information processing.[1,2] At low temperature, the dominant mechanism of spin relaxation for localized electrons in semiconductor QDs is due to the hyperfine interaction with nuclear spins.[3–5] Single QDs present an excellent system for studying optically induced dynamic nuclear spin polarization (DNSP)[6,7] and electron-spin relaxation on an isolated ensemble of $\sim 10^5$ nuclear spins.[8,9] Studying the dynamics of DNSP and electron-spin relaxation in a single QD has the advantages of avoiding effects of the inhomogeneous broadening of spectral lines due to ensemble QDs and differently charged excitons over doped QD ensembles, which may become a major obstacle in accessing fully detailed intrinsic information on QD nuclear spins and the electron-spin relaxation properties in QDs. Controlling and understanding of QD electron-nuclear spin system can lead to many interesting experiments, such as the coherent exchange of information between electron and nuclear spins.[10,11]

Hyperfine interaction between electron and nuclear spins in single and ensemble QDs has been widely studied to elucidate electron-spin relaxation.[3,8,9,12–15] The electron-spin relaxation predicted by theory[3,8] is due to the hyperfine interaction between the ground state electron and randomly oriented nuclear spins. Note that optically polarized nuclear spins will be induced by a repeated pumping of QDs using circularly polarized pulses during the photoluminescence (PL) experiment.[16,17] It is therefore crucial to investigate the electron-spin relaxation in QDs under the condition of random orientation of nuclear spins, which requires a delicate technique and setup for the experiment.

In this work, we investigated the electron-spin relaxation in a single InAs QD with positively charged exciton (X^+) by time-resolved photoluminescence (TRPL) spectroscopy. The randomly oriented nuclear spin bath is kept in place by an alternating σ^+/σ^- sequence of excitation pulses under the condition that the switching-over period of the pumping pulse in a few nanoseconds is shorter than the nuclear spin buildup time, which is approximately on the order of milliseconds.[6] The observed time dependence of electron-spin polarization is in qualitative agreement with the model of interaction with randomly orientated nuclei predicted in Ref. 8, i.e., the electron-spin polarization decreases to a minimum value at first, and finally increases to a steady value of about 1/3 of its initial value.

The investigated QD samples were grown by molecular beam epitaxy on a semi-insulating GaAs substrate. They consist of, in sequence, an *n*-doped GaAs buffer layer, a 20-period *n*-doped $GaAs/Al_{0.9}Ga_{0.1}As$ distributed Bragg reflector (DBR), a 2λ GaAs cavity with an InAs QD layer at the cavity antinode, and a top *p*-doped GaAs layer. An ultra-low-density InAs QD layer was formed by depositing nominally 2.35 monolayers (ML) of InAs at a growth rate of 0.001 ML/s. In experiments, the QD sample was mounted in a continuous-flow liquid helium cryostat at 5 K. A mode-locked Ti:sapphire laser with 2 ps pulses and 80 MHz repetition frequency was tuned to a wavelength of 902 nm to excite the QD sample. The excitation intensity was about 5 μW. Figure 1(a) shows the photoluminescence of excitation (PLE) spectrum detected at 927 nm of the X^+ emission, where the peak at 902 nm is induced by the GaAs LO-phonon-assisted resonance excitation.[18] The emission line of X^+ was identified previously and is reported in Ref. 19. The emitted luminescence was collected by an objective (NA: 0.5), spectrally filtered by a 0.5 m monochromator, and detected by a silicon charge coupled device (CCD). For measuring high-resolution PL spectra (HRPL), a scanning Fabry-Perot interferometer (FPI) with free spectral range of 15 GHz (62 μeV), a multi-channel scaler (MCS), and an avalanche photodiode (APD) were used. TRPL measurements were carried out by a time-correlated single-photon counting (TCSPC) setup with a time resolution of 400 ps. For the polarization PL measurements, the excitation pulses were circularly polarized (σ^+) using a $\lambda/4$ wave plate. The luminescence emission was analyzed by a $\lambda/4$ wave plate and a linear polarizer to distinguish different circular polarization components.

As mentioned above, in order to control the nuclear spins in QD, we used two different kinds of pulse sequences. In the first configuration, the pulses are alternating with σ^+ and σ^- (T-arm and R-arm, respectively, as shown in Fig. 1(b)) polarizations, and these pulses are separated by 6.25 ns to excite the QD sample. Similar modulation techniques, such as those using photo-elastic and electro-optical modulators to study the nuclear spin dynamics in QDs, have been reported.[20,21] The hyperfine interaction-induced spin relaxation has also been discussed in continuous wave (cw) PL measurement under constant helicity or modulated polarized excitation.[22,23] In our case, TRPL is measured by TCSPC. In this configuration, the nuclear spins are not polarized as two opposite pulses are working alternately. However, in another configuration, the nuclear spins will be polarized when only one pulse

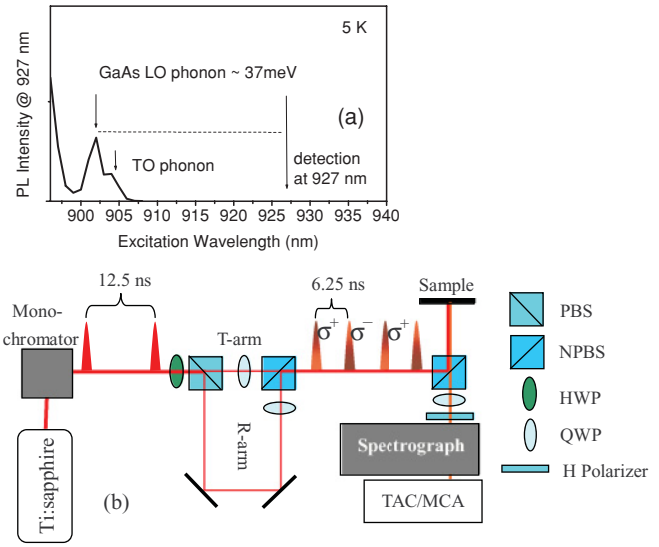

FIG. 1. (Color online) (a) PLE spectrum detected at 927 nm at 5 K. The cw excitation wavelength scans from 890 to 908 nm. (b) Schematic diagram of experimental setup of TRPL in which the T-arm and R-arm represent the optical paths of alternating σ^+ and σ^- excitation pulse sequences, separated by 6.25 ns. The 2 ps pulses are generated by 100 fs laser pulses going through a monochromator. TAC: time-amplitude converter. MCA: multichannel analyzer.

sequence with σ^+ polarization (T-arm in Fig. 1(b)) excites the QD sample. Using this setup, for the first time in TRPL measurement, we can truly test the mechanism of electron-spin relaxation under the influence of either random or polarized orientation of nuclear spin baths.

Figure 2(a) displays the TRPL intensity of X^+ emission under excitation with alternating σ^+/σ^- pulse sequences. The top and bottom curves represent the detected time-dependent emission intensities of σ^- and σ^+ polarizations, respectively. The time interval of two neighboring TRPL peaks pumped by alternating σ^+ and σ^- pulses is about 6.25 ns, corresponding to the σ^+/σ^- pulse sequences. From these curves, we can obtain the time-dependent circular polarization degree (P_c) according to the expression $P_c = (I_{\sigma+} - I_{\sigma-})/(I_{\sigma+} + I_{\sigma-})$, where $I_{\sigma+}$ and $I_{\sigma-}$ are the emission intensities of TRPL with σ^+ and σ^- components. Then, σ^+ (black line) and σ^- (red

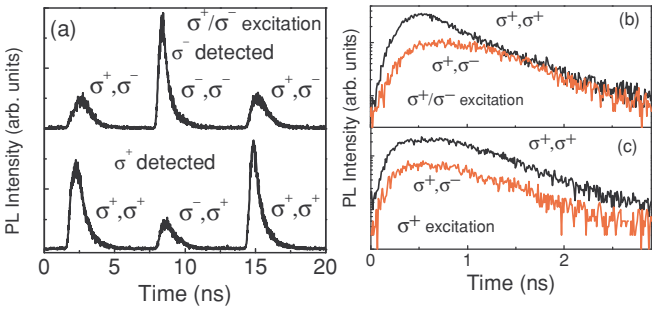

FIG. 2. (Color online) (a) TRPL intensity of X^+ emission under the excitation of alternating σ^+/σ^- pulse sequences, where the top and bottom decay curves represent σ^- and σ^+ components of TRPL, respectively. (b–c) Decay curves of σ^+ ([σ^+, σ^+], black) and σ^- ([σ^+, σ^-], red) components in a semilogarithmic scale, corresponding to either σ^+/σ^- (b) or σ^+ (c) pulse sequence excitation.

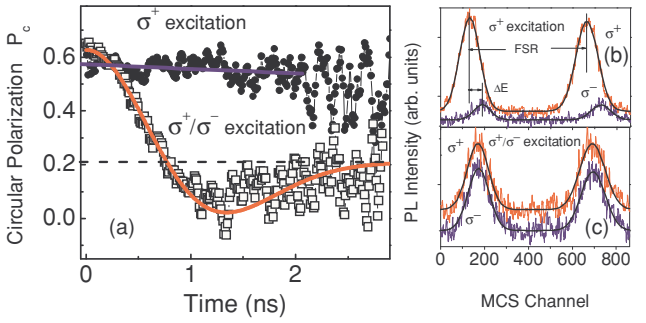

FIG. 3. (Color online) (a) Time-dependent circular polarization P_c under σ^+/σ^- (open squares) and σ^+ (solid circles) pulse sequence excitation. The red line is a fit to Eq. (1), and the blue line is the fit to an exponential function. Dashed lines are a marker value of 1/3 of initial P_c. The curves are σ^+ (red) and σ^- (blue) components of HRPL under σ^+ (b) and alternating σ^+/σ^- (c) pulse sequence excitation. The HRPL is measured by an F-P interferometer and recorded by a multichannel scaler (MCS). The corresponding Zeeman splittings (ΔE) are 7 ± 0.1 μeV (b) and ~ 0 μeV (c), calculated by fitting the experimental data with two Gaussian functions.

line) components obtained during the period of σ^+ pulse excitation were measured and are shown as (σ^+, σ^+) and (σ^+, σ^-) respectively in Fig. 2(b). Both of them correspond to the TRPL peaks at the time t of ~15 ns in Fig. 2(a). The measured time of each component is 20 min in order to get a higher ratio of signal to noise. The derived P_c data are shown in Fig. 3(a) by open squares, demonstrating that a rapid decrease of P_c exists at first, and afterwards P_c turns to slightly increase again. In contrast to the case of σ^+/σ^- excitation, the QD is also excited by only the σ^+ pulse sequence, and the measured σ^+ (black line) and σ^- (red line) emission components are shown in Fig. 2(c). The corresponding P_c values are indicated in Fig. 3(a) by solid circles. A quite long decay time of $\sim 34 \pm 7$ ns compared to the exciton lifetime of ~1 ns is derived by a single exponential fitting of the experimental data within 2 ns due to the large scattering of the data beyond 2 ns. In the case of QD excited by only the σ^+ pulse, the nuclear spins will be partly aligned by polarized electrons due to hyperfine interaction with nuclei. The orientated nuclear spins will give rise to an effective field B_{OS} (so-called Overhauser field), and this field will in turn suppress electron-spin relaxation,[16,24] leading to a slow exponential-like decay as shown in Fig. 3(a). Figure 3(b) presents a 7 ± 0.1 μeV of Zeeman splitting (ΔE) between the σ^+ and σ^- components of PL detected under the influence of B_{OS} field, which corresponds to a magnetic field of 149 mT if an electron g-factor g_e of ~0.8 is taken.[12,25] We find that the Zeeman splitting ΔE is close to zero if alternating σ^+/σ^- pulse sequences are used to excite the QD, which results in a randomly distributed nuclear spin orientation, as indicated in Fig. 3(c).

Electron-spin relaxation in the randomly distributed frozen fluctuation of the nuclear field has been theoretically investigated,[3,8] and the time dependence of the ensemble averaged electron-spin polarization can be written as[8,12]

$$\langle S(t)\rangle = \frac{S_0}{3}\left\{1 + 2\left[1 - 2\left(\frac{t}{2T_\Delta}\right)^2\right]\exp\left[-\left(\frac{t}{2T_\Delta}\right)^2\right]\right\} \quad (1)$$

where S_0 is the initial electron-spin polarization, $T_\Delta = \hbar/\mu_B g_e \Delta_B$ is the dephasing time, $\hbar$ is the reduced Planck constant, μ_B is the Bohr magneton, g_e is the electron g factor, and Δ_B is the dispersion of the nuclear hyperfine field distribution. For TRPL measurements of a single QD, Eq. (1) is valid for describing the electron-spin polarization averaged over a large number of successive measurements.[8] The dephasing time due to variable electron precession frequencies during successive measurements is termed "spin relaxation," like the term defined in ensemble QDs.[12] It is found that the experimental result for randomly oriented nuclear spins (open squares in Fig. 3(a)) can be described by Eq. (1), where the red curve is a fitting result with T_Δ of 0.55 ns. A value of 0.5 ns in the same order has been reported for *p*-doped ensemble QDs.[12] The short dephasing time indicates that the electron-spin relaxation induced by randomly orientated nuclei is very efficient. Merkulov *et al*. show that this dephasing time T_Δ can be written as,[8,12]

$$T_\Delta = \hbar \left[n^2 \sum_j I^j (I^j + 1)(A^j)^2/(3N) \right]^{-1/2} \qquad (2)$$

where N is the number of nuclei interacting with the electron in the QD, A^j is the hyperfine constant, I^j is the spin of the jth nucleus, and n is the number of nuclei per unit cell. The sum goes over all the atoms in the primitive unit cell. Based on Eq. (2), the dephasing time can be estimated by using the parameters of the hyperfine constants of As ($I^{\mathrm{As}} = 3/2$) and In ($I^{\mathrm{In}} = 9/2$) nuclei, $\mathrm{A_{As}} = 47\ \mu$eV and $\mathrm{A_{In}} = 56\ \mu$eV, and $n = 2$. For $N \sim 10^5$, Eq. (2) yields $T_\Delta \sim 0.58$ ns for an InAs QD, which is in good agreement with the experimental value of 0.55 ns. From the obtained dephasing time T_Δ, we can estimate the dispersion Δ_B of the nuclear hyperfine field. It is found that $\Delta_B = \hbar/\mu_B g_e T_\Delta \sim 26$ mT, for electron g factor of $g_e \sim 0.8$.[12,25] This is close to the value of 28 mT reported by Braun *et al*.[12] On the other hand, when only the σ^+ pulse sequence is used to excite QD, the Overhauser field, $B_{OS} \sim 149$ mT, will suppress the electron-spin relaxation, prolonging the dephasing time T_Δ to 34 ± 7 ns, as obtained by single exponential fitting of the experimental data (blue line in Fig. 3(a)). Using the expression of $\Delta_B = \hbar/\mu_B g_e T_\Delta$, the estimated dispersion Δ_B of nuclear hyperfine field corresponding to a dephasing time of 34 ± 7 ns is 0.4 ± 0.08 mT. This value is much smaller than the value of 26 mT obtained in the condition of randomly oriented nuclei, implying a much narrower distribution of the nuclear field when only the σ^+ pulse sequence is used to excite QD. We have also found that the electron-spin polarization increases with increasing cw or pulsed σ^+ excitation power, leading to an increase in the dephasing time of electron spin.[9] Therefore, the measured dephasing time depends on the excitation power of constant helicity. In our experiment, the obtained time of $\sim$34 ns corresponds to an excitation power of $\sim$5 μW when the PL intensity is close to the saturated value. Note that in order to suppress the electron-spin dephasing induced by hyperfine interaction, the required magnetic field applied along the Oz growth axis must be larger than Δ_B.[8,12] The TRPL measurements here show that the optically orientated nuclear spins can effectively suppress the electron-spin dephasing, even in the absence of applied magnetic field.

The circular polarization P_c (t) ($P_c[t] \propto$ electron-spin polarization $S[t]$) of the excitonic radiative recombination X^+ in Fig. 3(a) probes the electron-spin relaxation.[12] The value of circular polarization $P_c(t)$ in Fig. 3(a) is found to decrease down to about 60% of its initial value $P_c(0)$ at time $t = T_\Delta \sim 0.55$ ns, then it further decreases to a minimum value of $\sim$8% of $P_c(0)$ at time $t \sim 1.34$ ns, and then increases again to a steady value of (0.32 ± 0.17) of $P_c(0)$ at time $t \sim 2.8$ ns, which is close to the expected value of $P_c(\infty) = P_c(0)/3$. A relatively large error and a deviation from the expected $P_c(0)/3$ value are due to the weak PL signal and a large scattering of the data when time is longer than radiative lifetime. In fact, a steady value of $P_c(0)/3$ was clearly manifested by Braun *et al*. using *p*-doped ensemble InAs QDs[12] and transport measurements in single GaAs QDs.[2,26] Note that in Fig. 3(a), the minimum value of fitted red curve at 1.34 ns is 4%, which is smaller than the averaged value of the experimental data of $\sim$8%. This difference occurs probably due to an imbalance in laser intensity of σ^+/σ^- pulse sequences during the TRPL measurement, which may lead to a nuclear spin bath that is not perfectly random. In addition, the APD time response could affect the depth of the detected minimum value.[27] It is also noted that the absence of the turning point (minimum value) in the optical measurement of ensemble QDs has been reported and discussed, ascribing it to the dot size fluctuations of ensemble QDs.[12,28]

In summary, by using alternating σ^+/σ^- or σ^+ pulse sequences to optically excite the QD, either randomly oriented or polarized nuclear spin baths are generated. This enables us to test electron-spin relaxation under different configurations of nuclear spin baths. The obtained result of electron-spin relaxation is in qualitative agreement with the theoretical prediction for randomly orientated nuclear spins, showing that the electron-spin polarization decreases to a minimum value at first, and finally increases to a steady value of about 1/3 of its initial value.

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*bqsun@semi.ac.cn

[1] D. Loss and D. P. DiVincenzo, Phys. Rev. A **57**, 120 (1998).
[2] R. Hanson, L. P. Kouwenhoven, J. R. Petta, S. Tarucha, and L. M. K. Vandersypen, Rev. Mod. Phys. **79**, 1217 (2007).
[3] A. V. Khaetskii, D. Loss, and L. Glazman, Phys. Rev. Lett. **88**, 186802 (2002).
[4] M. H. Mikkelsen, J. Berezovsky, N. G. Stoltz, L. A. Coldren, and D. D. Awschalom, Nature Phys. **3**, 770 (2007).
[5] X. Xu, B. Sun, P. R. Berman, D. G. Steel, A. S. Bracker, D. Gammon, and L. J. Sham, Nature Phys. **4**, 692 (2008).
[6] P. Maletinsky, A. Badolato, and A. Imamoglu, Phys. Rev. Lett. **99**, 056804 (2007).

[7]J. R. Petta, J. M. Taylor, A. C. Johnson, A. Yacoby, M. D. Lukin, C. M. Marcus, M. P. Hanson, and A. C. Gossard, Phys. Rev. Lett. **100**, 067601 (2008).
[8]I. A. Merkulov, Al. L. Efros, and M. Rosen, Phys. Rev. B **65**, 205309 (2002).
[9]X. M. Dou, X. Y. Chang, B. Q. Sun,Y. H. Xiong, Z. C. Niu, H. Q. Ni, and D. S. Jiang, Appl. Phys. Lett. **95**, 221903 (2009).
[10]T. D. Ladd, D. Press, K. D. Greve, P. L. McMahon, B. Friess, C. Schneider, M. Kamp, S. Höfling, A. Forchel, and Y. Yamamoto, Phys. Rev. Lett. **105**, 107401 (2010).
[11]H. Christ, J. I. Cirac, and G. Giedke, Phys. Rev. B **75**, 155324 (2007).
[12]P. F. Braun, X. Marie, L. Lombez, B. Urbaszek, T. Amand, P. Renucci, V. K. Kalevich, K.V. Kavokin, O. Krebs, P. Voisin, and Y. Masumoto, Phys. Rev. Lett. **94**, 116601 (2005).
[13]R. Oulton, A. Greilich, S. Yu. Verbin, R. V. Cherbunin, T. Auer, D. R. Yakovlev, M. Bayer, I. A. Merkulov, V. Stavarache, D. Reuter, and A. D. Wieck, Phys. Rev. Lett. **98**, 107401 (2007).
[14]B. Pal, S. Yu. Verbin, I. V. Ignatiev, M. Ikezawa, and Y. Masumoto, Phys. Rev. B **75**, 125322 (2007).
[15]M. Yu. Petrov, I. V. Ignatiev, S. V. Poltavtsev, A. Greilich, A. Bauschulte, D. R. Yakovlev, and M. Bayer, Phys. Rev. B **78**, 045315 (2008).
[16]M. I. Dyakonov, in *Spin Physics in Semiconductor*, edited by M. I. Dyakonov (Springer Series in Solid-State Sciences, 2008), p. 1–28.
[17]I. A. Merkulov, G. Alvarez, D. R. Yakovlev, and T. C. Schulthess, Phys. Rev. B **81**, 115107 (2010).
[18]K. Kowalik, O. Krebs, A. Lemaître, J. A. Gaj, and P. Voisin, Phys. Rev. B **77**, 161305(R) (2008).
[19]X. Y. Chang, X. M. Dou, B. Q. Sun, Y. H. Xiong, Z. C. Niu, H. Q. Ni, and D. S. Jiang, J. Appl. Phys. **106**, 103716 (2009).
[20]R. I. Dzhioev, B. P. Zakharchenya, V. L. Korenev, and M. V. Lazarev, Phys. Solid State **41**, 2014 (1999).
[21]R. V. Cherbunin, S. Yu. Verbin, T. Auer, D. R. Yakovlev, D. Reuter, A. D. Wieck, I. Ya. Gerlovin, I. V. Ignatiev, D. V. Vishnevsky, and M. Bayer, Phys. Rev. B **80**, 035326 (2009).
[22]O. Krebs, B. Eble, A. Lemaître, B. Urbaszek, K. Kowalik, A. Kudelski, X. Marie, T. Amand, and P. Voisin, Phys. Stat. Sol. (a) **204**, 202 (2007).
[23]M. Yu. Petrov, G. G. Kozlov, I. V. Ignatiev, R. V. Cherbunin, D. R. Yakovlev, and M. Bayer, Phys. Rev. B **80**, 125318 (2009).
[24]R. Oulton, A. Greilich, S.Yu. Verbin, R.V. Cherbunin, T. Auer, D. R. Yakovlev, M. Bayer, I. A. Merkulov, V. Stavarache, D. Reuter, and A. D. Wieck, Phys. Rev. Lett. **98**, 107401 (2007).
[25]M. Bayer, A. Kuther, A. Forchel, A. Gorbunov, V. B. Timofeev, F. Schäfer, and J. P. Reithmaier, Phys. Rev. Lett. **82**, 1748 (1999).
[26]J. R. Petta, A. C. Johnson, J. M. Taylor, E. A. Laird, A. Yacoby, M. D. Lukin, C. M. Marcus, M. P. Hanson, and A. C. Gossard, Science **309**, 2180 (2005).
[27]C. Becher, A. Kiraz, P. Michler, A. Imamoğlu, W. V. Schoenfeld, P. M. Petroff, Lidong Zhang, and E. Hu, Phys. Rev. B **63**, 121312(R) (2001).
[28]P. F. Braun, L. Lombez, X. Marie, B. UrbaszeK, T. Amand, P. Renucci, J. L. Gauffier, V. K. Kalevich, K. V. Kavokin, O. Krebs, and P. Voisin, Braz. J. Phys. **36**, 482 (2006).