Millisecond-range electron spin memory in singly-charged InP quantum dots

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We report millisecond-range spin memory of resident electrons in an ensemble of InP quantum dots (QDs) under a small magnetic field of 0.1 T applied along the optical excitation axis at temperatures up to about 5 K. A pump-probe photoluminescence (PL) technique is used for optical orientation of electron spins by the pump pulses and for study of spin relaxation over the long time scale by measuring the degree of circular polarization of the probe PL as a function of pump-probe delay. Dependence of spin decay rate on magnetic field and temperature suggests two-phonon processes as the dominant spin relaxation mechanism in this QDs at low temperatures.

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Electron spin in semiconductors may be suitable for use as a quantum memory in quantum repeaters as semiconductors are capable of converting photons to electrons (and holes) while transferring quantum information from conductors. While transferring information from conductors to photons is also possible, the process is more complex and the fidelity of the information is not as high as in the opposite direction. The transfer of information from conductors to photons is also more energy-efficient.

In this letter we report observation of optically created electron spin orientation dynamics by measuring the circular polarization [defined as P = (I↑↑ + I↑↓ + I↓↑ + I↓↓)/(I↑↑ + I↑↓ + I↓↑ + I↓↓), where I↑↓ (I↓↑) is the PL intensity for excitation with σ+ probe and detection of σ− probe] of the probe pulse PL in presence of a preexcitation by a pump pulse. Our experimental setup is schematically shown in Fig. 1(a).

A CW Ti:sapphire laser beam is split into pump and probe beams. Two acousto-optic modulators (AOM) driven by programmable function generator (PFG) generates pump and probe pulses with controllable pulse width and delay (τ) between them. Glan-Thompson polarizers (GTP) and wave plates are used to control the circular polarization of the pump and probe beams. The probe PL signal is sent through a combination of a photelastic modulator (PEM) and a GTP before dispersing in a monochromator and detecting in a GaAs photomultiplier tube (PMT). The PMT output is connected to a two-channel gated photon counter (GPC). The PEM acts as an oscillating λ/4-plate and when combined with GTP, allows detection of PL intensity in the σ+ and σ− channels. The PEM frequency, fP = 42 kHz, is reduced to fP = fP/(n + 0.5) (typically n = 40 in our measurements) to trigger the PFG and GPC. Thus, one probe pulse (and A-gate of the GPC) is centered at the +λ/4 and the next probe pulse (and B-gate of the GPC) is centered at the −λ/4 retardation peaks of the PEM [Fig. 1(b)]. We usually use a GPC gate width of 5 µs, while the pump (probe) pulse width is 60 (3) µs, giving a pump (probe) power density [Wpump (probe)] of about 0.5 (0.05) W cm−2. The low probe power density ensures that the pump-induced spin polarization is not fully erased by the probe pulse. The excitation energy is tuned to about 1.771 eV (below-barrier, QD excited state excitation) and the QD ground state PL is detected at about 1.729 eV. An external electric bias of Ubias = −0.1 V is applied to the sample. We find that under this condition the PL polarization is negative and reaches maximum. A study of trionic quantum beats in this sample showed that at Ubias ≈ −0.1 V each QD contains one resident electron on an average. This suggests that the negative PL polarization arises from trionic state, as is discussed e.g., in Refs. 6, 14, 15.

In our experiments, a σ+ (or σ−) polarized pump induces ↓ (or ↑) spin orientation of the resident electrons. A probe pulse, variably delayed with respect to the pump pulse, tests this pump-induced spin-orientation. The probe beam (always σ+ polarized) creates a hot trion with parallel ↓↓-QDs or anti-parallel ↑↑-QDs electron spins. After a flip-flop process in ↓↓-QDs shown schematically in Fig. 2(a), the probe PL polarization becomes negative while it is positive for the ↑↑-QDs [Fig. 2(b)]. At any given τ the net probe PL polarization is determined by the ratio of ↓↓- and ↑↑-QDs.

We measure the probe PL polarization for (i) co-
circularly polarized pump-probe ($P_{CO}$) [pump creates more $\downarrow\downarrow$-QDs] and (ii) cross-circularly polarized pump-probe ($P_{CR}$) [pump creates more $\uparrow\downarrow$-QDs] [pump and probe polarizations for the two cases are indicated in Fig. (b)]. A small static magnetic field of $B = 0.1$ T is applied parallel to the optical excitation (and sample growth) axis to suppress the effect of fluctuating nuclear magnetic field.\textsuperscript{18,19} Polarisations $P_{CR}$ and $P_{CO}$ as a function of $\tau$ are shown in Fig. (c). The difference $P_{CR} - P_{CO}$ is a good measure of the pump induced spin orientation of the resident electrons.\textsuperscript{20} A semilogarithmic plot of $P_{CR} - P_{CO}$ obtained from Fig. (c) shows that the spin memory decay is nonexponential [Fig. 2(d)]. Thus, a spin relaxation time cannot be defined in a simple way. However, it is clear from this data that the spin memory decays on a millisecond time-scale.

![FIG. 1: Schematic of the experimental setup (a) and time synchronization (b) of the PEM retardation, the probe pulses, and the GPC gates.](image)

The observed long-lived spin polarization could result from a dynamic nuclear polarization which may appear under the experimental condition used.\textsuperscript{17,18,21} However, in a recent study of this aspect we have shown that very small effective magnetic field ($< 0.02$ T) in InP QDs\textsuperscript{22,23} arising from dynamic nuclear polarization, is not consistent with the large amplitude of PL polarization observed in this sample. Thus, the long spin memory observed here should be related to the lack of efficient spin decay path in QDs. To investigate the spin relaxation mechanisms effective in this case we study temperature and magnetic field dependence of the spin decay process.

![FIG. 2: Schematics of $\downarrow\downarrow$-QDs (a) and $\uparrow\downarrow$-QDs (b). Probe PL polarization for co- ($P_{CO}$) and cross- ($P_{CR}$) circularly polarized pump-probe (c), and the difference $P_{CR} - P_{CO}$ (d) as a function of $\tau$.](image)

Figure 2 shows decay of $P_{CR} - P_{CO}$ at a few temperatures for $B = 0.1$ T. A faster decay is seen with increasing $T$. As noted earlier, the decay is nonexponential and suggests a distribution of decay rates, which may arise due to inhomogeneous environment and size-distribution of the QDs.\textsuperscript{11} Theoretical analysis shows (see e.g., Ref. 24) that a spread of the relaxation rate results in a nonexponential decay of the form $\sim \exp[-(\gamma_s \tau)\gamma]$ (the so-called stretched exponential function), where the parameter $c$ depends on the physical processes causing the spread. We find that the function fits our data very well (dashed lines in Fig. 3) if we use $c$ as a fitting parameter. Effective spin decay rate $\gamma_s$ obtained from such fits is plotted in the inset of Fig. 3 as a function of $T$. A rapid increase in $\gamma_s$ is seen for $T > 8$ K. Such an increase is expected for thermally activated spin relaxation due to the phonon-mediated coupling of the ground and excited electron states (two-phonon Orbach process).\textsuperscript{25} We find that the function $\gamma_s \sim (\exp[\Delta E/k_B T] - 1)^{-1} + \gamma_0$ ($\Delta E = $ activation energy, $k_B = $ Boltzmann constant, and $\gamma_0$ stands for spin decay rate arising from temperature independent relaxation mechanisms) describing this process fits the data very well (solid line in the inset of Fig. 3). From the fit we obtain $\Delta E \approx 5$ meV. This value is smaller than that obtained experimentally for electron level spacing of 15 meV in Ref. 13. This discrepancy is probably due to the difference in QD sub-ensemble probed in the two cases.
Various mechanisms of optical orientation of spins are discussed e.g., in Refs. 18, 19. Possible mechanisms of optical orientation of resident electron spins in InP QDs under study will be discussed elsewhere.

We now present the magnetic field dependence of the spin decay process. Decay of $P_{\text{CR}} - P_{\text{CO}}$ at a few values of $B$ at $T = 2$ K is shown in Fig. 4. We find that the decay becomes increasingly faster with increase in $B$. An effective spin decay rate obtained from stretched exponential fit to the data is plotted as a function of $B$ in the inset of Fig. 4. Decay rate $\gamma_s$ is found to increase superlinearly with $B$. Several possible mechanisms for such an increase are discussed in the literature. Magnetic field couples the higher energy states with nonzero orbital momentum to the electron spin states split by the magnetic field (Zeeman splitting) that allows a small admixture of the states of opposite spin to each Zeeman sublevel. At low temperature this enables spin-flip transition between Zeeman sublevels via participation of acoustic phonons to dissipate energy (one-phonon resonant process). With increasing $B$ the Zeeman splitting increases. Due to higher density of resonant phonons at increased energy and more efficient mixture of the states by the magnetic field, the spin relaxation rate increases. Theoretical calculations have predicted $\gamma_s \sim B^3$ at very low temperature and large magnetic field if the spin-orbit interaction and the one-phonon scattering dominate. However, for $T$ of about a few kelvin, the two-phonon nonresonant (Raman) scattering may become important. In that case, the magnetic field dependence is only determined by the admixture of the excited states and becomes quadratic.

Our data in Fig. 4 inset can be fitted very well with $\gamma_s = \alpha + \beta B^2$. This argues for the two-phonon scattering as the main mechanism of acceleration of the spin relaxation in magnetic field.

The acceleration of spin relaxation could also result from hyperfine interaction. However, this is unlikely in our case due to very small nuclear spin polarization in the InP QDs we studied.

In conclusion, we have observed long spin memory, persisting over 1 ms, in an ensemble of singly negatively charged InP QDs at small magnetic field (0.1 T) and at moderate temperature ($\sim$ 5 K). Our data on the magnetic field and temperature dependence of spin decay rate suggests two-phonon scattering may be the dominant spin relaxation mechanism. Long spin memory observed here in III-V semiconductor QDs is relevant for quantum information communication and storage. Though our study is made at about 0.7 $\mu$m wavelength ($\lambda$), III-V semiconductor system can be easily adapted to $\lambda = 1.3$ and 1.5 $\mu$m, suitable for fiber optic communication.

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