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Temperature-induced spin-coherence dissipation in quantum dots

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The temperature dependence of electron-spin coherence in singly negatively charged (In,Ga)As/GaAs quantum dots is studied by time-resolved Faraday rotation. The decoherence time T_2 is constant on the microsecond scale for temperatures below 15 K; for higher temperatures it shows a surprisingly sharp drop into the nanosecond range. The decrease cannot be explained through inelastic scattering with phonons, and it may be related to elastic scattering due to phonon-mediated fluctuations of the hyperfine interaction.

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Solid-state systems are interesting for implementation of quantum information processing because they may provide controllable qubits sufficiently protected from environment-induced classicality.[1,2] Specifically, in semiconductor quantum dots (QDs), a qubit can be defined by the two-level system of a confined electron spin,[3] which currently attracts great attention because of its long relaxation times. The spin relaxation can be characterized by two time scales, the longitudinal relaxation time T_1, limited by inelastic scattering, and the transverse relaxation time T_2 (also called decoherence time), for which limitations may arise also from elastic scattering. The relation between these times is nontrivial and is often summarized by the simple relation $(T_2)^{-1}=(2T_1)^{-1}+(T_2')^{-1}$, where T_2' is the pure or elastic decoherence time.

For the T_1 time of a QD electron spin, a number of investigations exist, from both experiment and theory. Compared to higher-dimensional systems, the T_1 times are very much enhanced because the QD confinement protects the spin from the main inelastic-scattering mechanism: the electron-spin coupling with its orbital motion. In high magnetic fields, T_1 has been shown to persist over tens of milliseconds or even longer at cryogenic temperatures,[4,5] in accord with theoretical calculations.[6] Further, its dependence on external parameters such as temperature and magnetic field for neutral and charged quantum dots has been studied.[7–10]

On the other hand, the information about the T_2 time is still limited. Considering that inelastic scattering would be the only channel for decoherence, T_2 may be as large as $2T_1$. However, studies at cryogenic temperatures show T_2 times in the microsecond range, showing that the elastic relaxation channel due to hyperfine interaction plays the dominant role under these conditions.[11,12] Recently, several calculations for T_2 times have been reported.[13–21]

An important figure of merit of electron-spin qubits is stability under temperature changes. A temperature increase enhances the lattice phonon occupation, so that decoherence mechanisms involving phonons gain importance. Here we study the QD electron-spin coherence as a function of temperature. We show that coherence can be initiated by short laser pulses for temperatures up to ~100 K. The coherence time, however, is temperature independent only up to 15 K; above it shows a sharp drop. From model calculations we conclude that this sharp drop is not related to spin-orbit coupling but arises from hyperfine interaction fluctuations involving phonons.

Time-resolved Faraday rotation (FR) studies using a pump-probe technique have been performed on an ensemble of singly negatively charged (In,Ga)As/GaAs QDs (see Ref. 22 for details). The sample was immersed in the variable temperature insert of a superconductor magnet for fields B aligned perpendicular to the optical axis. For optical excitation a mode-locked Ti:sapphire laser was used, emitting pulses with 1.5 ps duration at a rate of 75.6 MHz (corresponding to $T_R=13.2$ ns pulse separation) with a photon energy tuned to the QD ground-state optical transition. Using a laser-pulse picker, we were able to increment the laser repetition period T_R.

Decoherence time measurements on QD ensembles are constrained by dephasing due to inhomogeneities in the ensemble. The electron-spin dephasing time $T_2^\star$ has been found to be on the order of 10 ns only.[22–24] This fast dephasing can be overcome by exciting with a train of laser pulses which synchronizes precessional phase modes of electron-spin subsets in the ensemble.[12,25] This mode locking produces constructive interference patterns in the FR spectrum due to focusing of ensemble inhomogeneities. Consequently it allows one to recover the dynamics of a single QD by filtering out T_2 from a $T_2^\star$ measurement.[12]

Figure 1(a) shows FR traces at $B=2$ T for different temperatures as a function of delay between pump and probe. After initialization of a spin pure state at time zero, coherent oscillations due to spin precession about the magnetic field are observed. Within the first nanosecond of delay, the ensemble signal arises either from resident electrons in singly charged QDs or from exciton electrons in neutral QDs. The exciton lifetime is about 300 ps, as measured by differential transmission spectroscopy. Therefore the FR signal after ~1 ns can be related to resident electrons only.[22]

At temperatures $T<30$ K, the resident electron signal at positive delays is accompanied by coherent signal on the negative delay side. This signal arises from mode locking of electron spins whose precession frequencies are synchronized with the exciting laser. When the temperature is increased above 40 K, the negative delay signal disappears rather abruptly, while the positive delay signal is still pronounced up to 100 K.[26] Thus the data show that resident electron-spin coherence can be efficiently created at elevated temperatures. For an electron with arbitrary spin, the excitation creates a superposition of an electron state that blocks

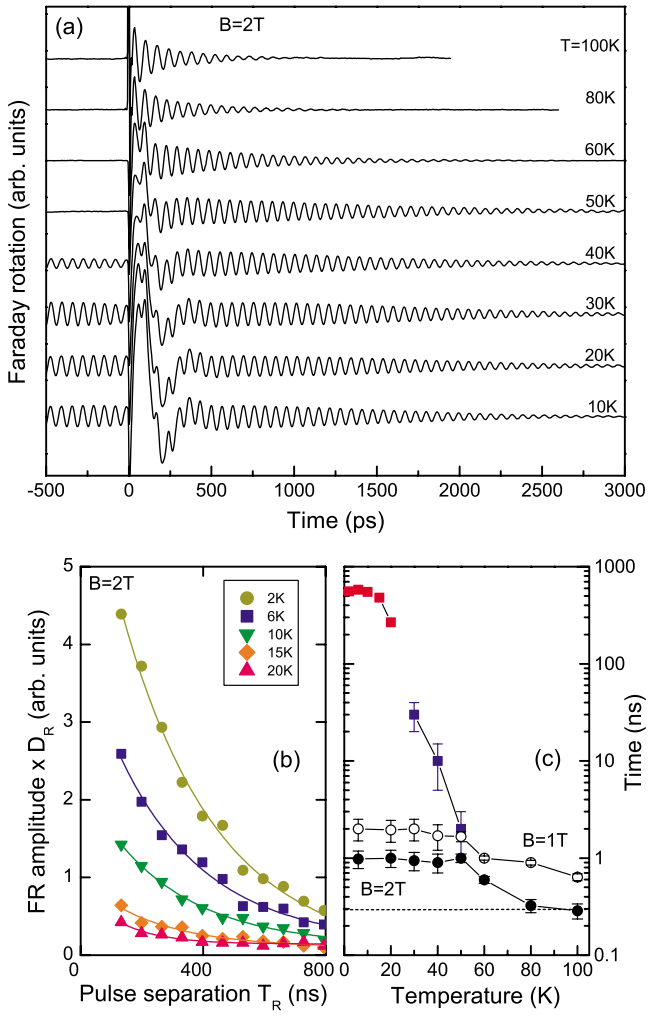

FIG. 1. (Color online) (a) Normalized FR spectra vs pump-probe delay at $B=2$ T for different temperatures; $P_{\text{pump}}=180$ W/cm^2, $P_{\text{probe}}=25$ W/cm^2. (b) FR amplitude at negative delay vs laser repetition period T_R for different temperatures. Solid lines are fits using exponential decay forms with time T_2. (c) Decoherence time T_2 (squares) and dephasing time $T_2^\star$ (full circles) vs temperature at $B=2$ T. Open circles give $T_2^\star$ at $B=1$ T. Dotted line marks the exciton/trion lifetime.

excitation due to the Pauli principle and a charged exciton consisting of a spin singlet electron doublet and a hole. After decay of the trion, an electron is left whose polarization along the optical axis has been increased by the excitation. This mechanism works, however, only if the hole spin is not scattered during the pump pulse.[22]

For discussing spin coherence, we focus on the negative delay signal. As reported earlier,[12] the spin decoherence time T_2 may be inferred by measuring the FR amplitude at negative delay for increasing separation T_R between the laser pump pulses. A change in T_R can be expressed in terms of the division rate $D_R=T_R/(13.2\ \text{ns})$. In our studies T_R was changed from 132.0 ns ($D_R=10$) up to 794.4 ns ($D_R=60$). The upper D_R limit is set by the negative delay signal becoming too weak due to the low cycling rate.

In Fig. 1(b) the negative delay FR amplitude at $B=2$ T, multiplied by D_R to correct the spectroscopic response for decreasing average power at fixed signal recording time, is plotted versus the laser repetition period for different temperatures. The experimental data are fitted by exponential decays with times T_2 (solid lines).[12] T_2 as a function of temperature is plotted in Fig. 1(c) as squares. At low temperature the measured spin-coherence time T_2 is about 600 ns, in good accord with previous reports.[11,12] T_2 remains constant with temperature increment up to 15 K. However, we find a surprisingly sharp drop in T_2 down to 250 ns at 20 K.

Heating up further, the negative delay FR signal can be seen only for small pump laser separations; but a systematic increase in D_R, as required for measuring T_2, is not possible. For example, strong mode-locking signal is seen at $T=30$ K for $T_R=13.2$ ns, but for $D_R=10$ the signal becomes already immeasurably small. This clearly suggests that the spin coherence is destroyed on time scales far below 132 ns. In this temperature range ($T>30$ K), we have therefore used the mode-locking amplitudes at negative delay for $T_R=13.2$ ns to obtain estimates for T_2 [blue squares in Fig. 1(c)]. Calculations show that the coherence time cannot exceed 30 ± 10 ns in order to lose the mode-locking signal completely when increasing D_R from 1 to 10 at $T=30$ K. The mode-locking amplitude for $D_R=1$ decreases strongly going from 30 to 40 K, which can be explained by a further reduction in T_2 to 10 ± 5 ns. At $T=50$ K the mode-locking signal has vanished completely for all D_R, which can be explained by a drop in T_2 into the 2 ns range.

The decay times of the FR signal for positive delays are also shown in Fig. 1(c) as circles for $B=1$ and 2 T. These times have been determined by fitting the FR traces by exponentially damped harmonics with damping time $T_2^\star$. At low temperatures the decay is determined by dephasing due to ensemble inhomogeneities such as electron g-factor variations or nuclear-spin fluctuations. The relation between $T_2^\star$ and T_2 is given by $(T_2^\star)^{-1}=(T_2)^{-1}+(T_{\text{inh}})^{-1}$, where the second term is the inhomogeneity-related scattering rate.

For $T<30$ K the dephasing time is basically constant and exceeds 1 ns for the chosen experimental conditions. As T_2 is more than 2 orders of magnitude longer in this range, $T_2^\star$ is basically identical to T_{inh}. While for $B<0.5$ T the nuclear field fluctuations become important, for higher fields the g-factor variations dominate. These variations are translated into a precession frequency variation scaling linearly with B, so that the dephasing occurs faster at 2 T than at 1 T [see Fig. 1(c)]. For completeness we note that under mode-locking conditions the dephasing depends on optical pump power. We use large bars to indicate this variation and not the experimental error (see Ref. 12 for details). Above 30 K we find a drop in $T_2^\star$, which we attribute to the increased importance of the homogeneous relaxation channel $1/T_2$. From extrapolating the T_2 data, one expects that T_2 becomes shorter than $T_2^\star$ for $T>50$ K.

As mentioned, the main sources of electron-spin decoherence are the spin-orbit coupling and the hyperfine interaction. As for the first case, because of the coupling of the orbital electronic motion to acoustic phonons, the spin-orbit interaction leads to an indirect dissipative channel. The spin-orbit coupling comprises two interaction mechanisms due to bulk inversion asymmetry of the crystal lattice (Dresselhaus) and asymmetry of the QD confining potential (Rashba).[2,27,28]

Both decoherence contributions can be investigated by mapping the interaction Hamiltonians onto bath-of-oscillator models in which the spin is directly coupled to the bath.[29,30] The corresponding spin-boson Hamiltonian is given by

$$\hat{H}_{\text{eff}} = -\frac{\hbar}{2}\Delta\hat{\sigma}_x + \sum_i \hbar\omega_i \hat{b}_i^\dagger \hat{b}_i + \hat{\sigma}_z \sum_i c_i(\hat{b}_i^\dagger + \hat{b}_i), \qquad (1)$$

where $\Delta = g\mu_B B/\hbar$ is the Zeeman frequency with electron g factor g, ω_i is the phonon frequency, and $\hat{\sigma}$ is the Pauli matrix. The second and third terms describe the oscillator bath. Here $\{\hat{b}_i, \hat{b}_i^\dagger\}$ are bosonic annihilation and creation operators. The third term accounts for the spin-bath coupling.

The details of the Dresselhaus and Rashba interactions are comprised in the effective spectral function $J_{\text{eff}}(\omega)$ of the bath "seen" by the electron spin. If the applied magnetic field B is such that the Zeeman frequency Δ is much less than the bath resonance peak Ω $(\Delta/\Omega \ll 1)$, the spin dissipative dynamics occurs in the low-frequency regime of the effective spectral function, given by[30]

$$J_{\text{eff}}(\omega) \approx m^* \gamma^2 \delta_s \left(\frac{\omega_D}{\omega_0}\right)\left(\frac{\omega}{\omega_D}\right)^{s+2}, \qquad (2)$$

where m^* is the electron effective mass, ω_0 is the splitting between the confined electron states, δ_s is the dimensionless electron-phonon coupling, and ω_D is the Debye frequency. The parameter γ corresponds to the spin-orbit coupling β and α for the Dresselhaus and Rashba contributions, respectively. The exponent s distinguishes piezoelectric $(s=3)$ and deformation-potential $(s=5)$ interactions.

In the spin-bath weak-coupling limit, the Bloch-Redfield equations[31] can be used to determine the spin expectation values, $\sigma_i = \text{Tr}\,\rho\hat{\sigma}_i$. Solving these equations,[32] we find T_1 and T_2 times related by $T_2 = 2T_1$ (in agreement with Ref. 33):

$$\frac{1}{T_{2,\text{SO}}} = \frac{1}{4} J_{\text{eff}}(\Delta) \coth\left(\frac{\hbar\Delta}{2k_B T}\right). \qquad (3)$$

The left panel of Fig. 2 shows the calculated decoherence times T_2 as a function of temperature up to 60 K, assuming piezoelectric interaction for the Dresselhaus and Rashba interactions.[34] We assumed an $In_{0.5}Ga_{0.5}As$ QD composition with an electron level splitting $\hbar\omega_0 = 20$ meV and a Zeeman energy $\hbar\Delta = 69.5$ μeV (for example, corresponding to $B = 2$ T, $g = 0.6$), as for the experimentally studied QDs. In addition, the following parameter values were used: $m^* = 0.041 m_e$, $\delta_3 = 298.5$, $\omega_D = 27.5$ meV, and the spin-orbit couplings $\beta = 3 \times 10^3$ m/s (Dresselhaus) and $\alpha = 9.6 \times 10^4$ m/s (Rashba).[27,35] We note that the calculated relaxation rates vary with In composition only by factors of order unity. For both interactions we see in panel (a) a strong drop in the transverse spin relaxation time. From the comparison we see that the Rashba interaction is 3 orders of magnitude more efficient than the Dresselhaus interaction. Still over the whole range the calculated times are orders of magnitude longer than the experimentally observed T_2. Therefore we exclude spin-orbit coupling as a source of the observed spin decoherence.

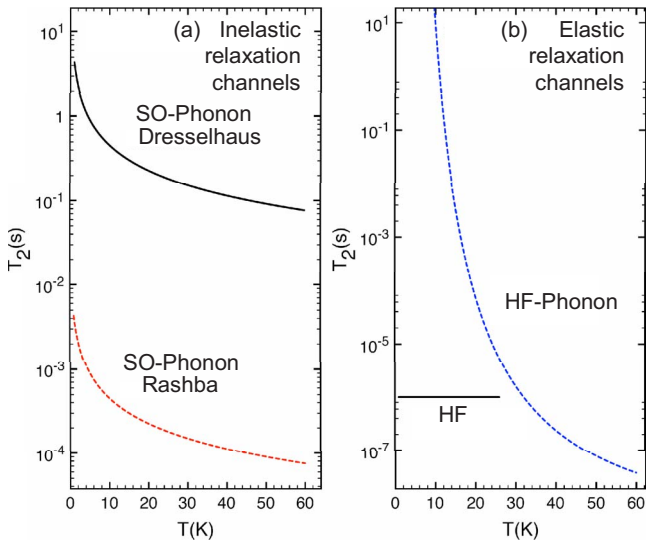

FIG. 2. (Color online) Calculated temperature dependence of T_2 for $In_{0.5}Ga_{0.5}As$ QDs; T_2 due to (a) inelastic spin-orbit-phonon scattering and (b) elastic-scattering time for hyperfine interaction [solid horizontal line (Ref. 19)]. In the right panel, the dashed line includes hyperfine interaction fluctuations due to phonon involvement (Ref. 17).

This leaves us with the hyperfine interaction described by a Hamiltonian which couples the electron spin **S** and the ith nuclear spin $\mathbf{I}_i$ in the QD:

$$\hat{H}_{\text{HF}} = \sum_i A_i |\psi(\mathbf{R_i})|^2 (\hat{S}_z \hat{I}_{i,z} + \hat{S}_+ \hat{I}_{i,-} + \hat{S}_- \hat{I}_{i,+}), \qquad (4)$$

where the sum goes over all nuclei in the QD electron localization volume. The interaction strength is determined by the hyperfine constant A_i and the electron density $|\psi(\mathbf{R_i})|^2$ at the nuclear site $\mathbf{R_i}$. $\hat{H}_{\text{HF}}$ mediates processes in which the spins of electron and nucleus are mutually flipped, as described by the products of raising and lowering operators $\hat{S}_\pm$ and $\hat{I}_{i,\pm}$, which increase and decrease the spin projections S_z and $I_{i,z}$ along the quantization axis z, respectively.

Indications of an inelastic-scattering channel have been found in studies of the dynamic nuclear polarization (DNP) by interaction with an optically oriented electron.[10] The DNP was found to be moderately increased for temperatures <50 K. This was attributed to a temperature-induced increase in the spin flip-flop efficiency by phonon-induced broadening of the electron level. This efficiency is restricted at cryogenic temperatures because of the mismatch in energy splittings between the electron and the nuclear Zeeman levels. The phonons required for compensating the energy mismatch are "frozen" under these conditions. By a temperature-induced level broadening, this energy mismatch may be softened. The data in Ref. 10, however, suggest that the change in the inelastic scattering by $\sim 10\%$ is too weak to explain the strong drop observed experimentally.

Independent of inelastic scattering, calculations of elastic mechanisms involving the hyperfine interaction have found decoherence times in the microsecond range,[15,16] as indicated in Fig. 2(b) by the horizontal line adopted from Ref. 19. Recently, theoretical calculations proposed an efficient deco-

herence mechanism due to modulations of the hyperfine field by phonons that may be dominant at low magnetic fields and high temperatures.[17] The corresponding decoherence time can be estimated by

$$\frac{1}{T_{2,\mathrm{HF}}} = \Gamma(I_i, n_{Ii}, V_{\mathrm{QD}}, A_i) F\left(\frac{\hbar\omega_0}{2k_B T}\right), \tag{5}$$

where Γ is a function of the nuclear-spin concentration n_{Ii} in the QD volume V_{QD}; and $F(x)=(1-\tanh^2 x)\tanh x$ contains the temperature dependence.

Equation (5) in combination with our QD parameters ($\Gamma^{-1}\sim 2.89$ ns) is plotted in Fig. 2(b) as a dashed curve. The results agree with the T_2 drop observed experimentally at about the same temperatures. The calculation deviation from the data can be related to the difficulty to determine the precise QD composition and the resulting nuclear environment of the electron spin.

In conclusion, we observed that the temperature-induced decoherence time dependence in (In,Ga)As self-assembled QDs shows two regimes: (i) $T\leq 15$ K: T_2 is temperature independent and limited by the hyperfine interaction, and (ii) $T>15$ K: T_2 is strongly temperature dependent and the main driven decoherence mechanism may be related to phonon-mediated hyperfine interaction fluctuations. One can see from Eq. (5), describing the resulting decoherence time, T_2 may be stabilized toward higher T by increasing the level splitting ω_0 of the QDs.

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