Enhancement of Electron Spin Coherence by Optical Preparation of Nuclear Spins

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We study a large ensemble of nuclear spins interacting with a single electron spin in a quantum dot under optical excitation and photon detection. At the two-photon resonance between the two electron-spin states, the detection of light scattering from the intermediate exciton state acts as a weak quantum measurement of the effective magnetic (Overhauser) field due to the nuclear spins. In a coherent population trapping state without light scattering, the nuclear state is projected into an eigenstate of the Overhauser field operator, and electron decoherence due to nuclear spins is suppressed: We show that this limit can be approached by adapting the driving frequencies when a photon is detected. We use a Lindblad equation to describe the driven system under photon emission and detection. Numerically, we find an increase of the electron coherence time from 5 to 500 ns after a preparation time of 10 μs.

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Introduction.—Single electron spins localized in small artificial structures, such as semiconductor quantum dots (QDs), have become available and to a large extent controllable [1–4]. Of particular interest is the phase coherence of electron spins as single quantum objects, both from a fundamental physics point of view and because of their potential use as quantum bits (qubits) for quantum information processing [5,6].

A number of physical mechanisms that lead to the gradual reduction of the quantum phase coherence (decoherence) of the electron spin have been analyzed [7]. It has been established experimentally [2–4] and theoretically [8–13] that, in a GaAs QD, the predominant decoherence mechanism is the hyperfine coupling to the nuclear spins in the host material. For an unpolarized ensemble of N nuclei and an effective hyperfine interaction energy A, the dephasing time in a weak magnetic field is $T_2^* \sim 1/\sigma \sim \sqrt{N}/A$, where σ is the width of the distribution of nuclear field values h_z parallel to the field. In a typical GaAs QD with $A \sim 90$ μeV or $A/g\mu_B = 3.5$ T [14], the number of Ga and As nuclei (spin $I = 3/2$) is $N \sim 5 \times 10^5$ and $T_2^* \sim 5$ ns; this value is supported by the experimental evidence [4,15]. The T_2^* decay originates from nuclear ensemble averaging and can be prolonged by narrowing the nuclear spin distribution [12]. Another strategy is to polarize the nuclear spins [8], but this requires a polarization close to 100% which is currently not available [12]. Two schemes have been proposed to achieve a narrowing of the nuclear spin distribution, based on electron transport [16] and gate-controlled electronic Rabi oscillations [17].

Here we analyze an optical scheme for nuclear spin preparation that makes use of spin-flip two-photon (Raman) resonance in a driven three-level system (TLS), in analogy to electromagnetically induced transparency (EIT) in atoms [18,19]. The lowest electronic states in a QD formed in a III-V semiconductor (e.g., GaAs) that are optically active under σ_+ circularly polarized excitation are the Zeeman-split ground state of a single localized conduction-band (E_C) electron and the negatively charged exciton (trion) $|X\rangle$, i.e., two electrons (spin up and down) plus one valence band heavy hole (hh) with angular momentum $J_{z'} = +3/2$ (Fig. 1). The $J = 3/2$ sector in the valence band is split into light hole and hh states along the axis z' of strong QD confinement. Here we assume excitation from the hh ($J_{z'} = \pm 3/2$) subband only. The axis z in

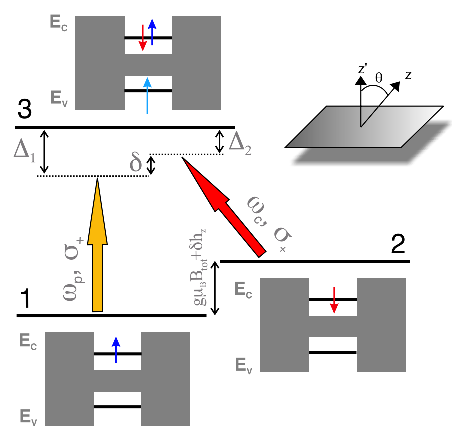

FIG. 1 (color online). Three-level system. State 1 (2) is a spin-up (-down) conduction-band (E_C) electron, with splitting $g\mu_B B_{\rm tot} + \delta h_z$, where δh_z is the z component of the nuclear (Overhauser) field fluctuations. State 3 is a trion with $J_{z'} = 3/2$. Two laser fields with frequencies ω_p and ω_c are applied near the 13 and 23 resonances with detunings $\Delta_{1,2}$. For a σ_+ circularly polarized excitation (along z'), both transitions are allowed for $\theta \neq 0$ and transitions to the $J_{z'} = -3/2$ states are forbidden. Inset: Structural axis z', leading to a splitting in E_V and spin quantization axis $z \parallel \mathbf{B}_{\rm tot}$ in E_C where $\cos\theta = z \cdot z' < 1$.

E_C is parallel to the total magnetic field $\mathbf{B}_{\rm tot}$, and we assume that the axes z and z' enclose an angle $\theta > 0$. The spin-up and -down states in E_C are then $|\Uparrow\rangle \equiv |\Uparrow\rangle_z = \cos(\theta)|\Uparrow\rangle_{z'} + \sin(\theta)|\Downarrow\rangle_{z'}$ and $|\Downarrow\rangle \equiv |\Downarrow\rangle_z = \cos(\theta)|\Downarrow\rangle_{z'} - \sin(\theta)|\Uparrow\rangle_{z'}$. Two circularly polarized (σ_+) continuous-wave lasers at the frequencies $\omega_p = \omega_X - \omega_\Uparrow - \Delta_1$ and $\omega_c = \omega_X - \omega_\Downarrow - \Delta_2$ stimulate the transitions between $|\Uparrow\rangle$ and $|X\rangle$ and between $|\Downarrow\rangle$ and $|X\rangle$, while the trion with $J_{z'} = -3/2$ is not excited.

The narrowing of the nuclear field distribution ν is based on light scattering in a TLS, where two long-lived (spin) states are coupled resonantly to an excited state that decays by spontaneous emission. When the two lasers satisfy exact two-photon resonance $\delta = \Delta_1 - \Delta_2 = 0$, one of the eigenstates of the system is a superposition of the two spin states with a vanishing excited state $|X\rangle$ component. The TLS at $\delta = 0$ is driven to this dark state with a vanishing light scattering rate [19]. The population of $|X\rangle$ and, thus, the photon scattering rate is nonzero for $\delta \neq 0$. In the presence of the nuclear spins, this resonance moves to $\delta = \delta h_z$, where δh_z is the deviation of the nuclear field (along z) from its mean $\langle h_z\rangle$. The absence of photon emission during a waiting time t constitutes a weak measurement of the quantum operator δh_z. In the limit $t \to \infty$, it becomes a strong measurement, projecting the nuclear state onto $|\delta h_z = 0\rangle$ (width $\sigma = 0$), thus eliminating electron decoherence due to the fluctuating field δh_z.

Model.—The Hamiltonian for the TLS coupled to nuclei is $H = H_0 + H_{\rm int} + H_{\rm hf}$, where $H_0 = -(\hbar\omega_z/2)\Sigma_z + \hbar\omega_X P_X$, with $\Sigma_i = \sigma_i \oplus 0$, the block-diagonal 3×3 matrix with the Pauli matrix σ_i in the upper left corner and 0 elsewhere, and $P_X = |X\rangle\langle X| = (001)^T(001)$. The spin splitting is given as $\hbar\omega_z = g\mu_B B_{\rm tot} = |g\mu_B\mathbf{B} + \langle\mathbf{h}\rangle|$, the sum of the external magnetic and the mean nuclear fields. The nuclear (Overhauser) field operator is $\mathbf{h} = \sum_{i=1}^N A_i\mathbf{I}_i$, where $A_i = a_i v_0|\Psi(\mathbf{r}_i)|^2$, and $\Psi(\mathbf{r}_i)$ denotes the electron wave function at the position $\mathbf{r}_i$ of the ith atomic nucleus, v_0 is the volume of the unit cell, and a_i is the hyperfine coupling strength for the nuclear species at site i. The classical laser fields in the rotating wave approximation (RWA) are described by [19] $H_{\rm int} = \Omega_p e^{i\omega_p t}|X\rangle \times \langle\Uparrow| + \Omega_c e^{i\omega_c t}|X\rangle\langle\Downarrow| + {\rm H.c.}$ The coupling of the electron spin to the quantum fluctuations of $\mathbf{h}$ is described by $H_{\rm hf} = -\frac{1}{2}\delta\mathbf{h}\cdot\mathbf{\Sigma}$, where $\delta\mathbf{h} = \mathbf{h} - \langle\mathbf{h}\rangle$. In the rotating frame $\tilde{\Psi}(t) = U(t)\Psi(t)$, with $U(t) = e^{-i\omega_p t}P_\Uparrow + e^{-i\omega_c t}P_\Downarrow + P_X$, where $P_\Uparrow = |\Uparrow\rangle\langle\Uparrow|$ and $P_\Downarrow = |\Downarrow\rangle\langle\Downarrow|$, we find $\tilde{H}(t) = U(t) \times [H(t) + \hbar\omega_p P_\Uparrow + \hbar\omega_c P_\Downarrow]U(t)^\dagger$ and, up to a constant (we drop the tilde and use H for the Hamiltonian henceforth),

$$H(t) = -\frac{\hbar}{2}\begin{pmatrix} \delta & 0 & \Omega_p \\ 0 & -\delta & \Omega_c \\ \Omega_p & \Omega_c & -\Delta \end{pmatrix} - \frac{\hbar}{2}\delta h_z\Sigma_z + H_\perp, \quad (1)$$

where $\Delta = \Delta_1 + \Delta_2$. The hyperfine flip-flop terms $H_\perp = \hbar(\delta h_+\Sigma_- e^{it(\omega_p-\omega_c)} + \delta h_-\Sigma_+ e^{-it(\omega_p-\omega_c)})/4$ are oscillating rapidly at the frequency $\omega_p - \omega_c = g\mu_B B_{\rm tot}/\hbar - \delta$ and can be neglected in the RWA [20], leading to a block-diagonal Hamiltonian $H = {\rm diag}(H_1, H_2, \ldots, H_K)$, with

$$H_k = -\frac{\hbar}{2}\begin{pmatrix} \delta h_z^k + \delta & 0 & \Omega_p \\ 0 & -\delta h_z^k - \delta & \Omega_c \\ \Omega_p & \Omega_c & -\Delta \end{pmatrix}, \quad (2)$$

where δh_z^k for $k = 1, 2, \ldots, K$ are the eigenvalues of the operator δh_z and $K = (2I+1)^N$ is the dimension of the nuclear spin Hilbert space. The state of the TLS combined with the nuclear spins is described by the density matrix ρ, which we divide up into 3-by-3 blocks $\rho_{kk'}$ and which evolves according to the generalized master equation [19]

$$\dot{\rho} = \mathcal{L}\rho \equiv \frac{1}{i\hbar}[H, \rho] + \mathcal{W}\rho, \quad (3)$$

with the Hamiltonian equation (1) and the dissipative term $\mathcal{W}\rho = \sum_{\alpha=\Uparrow,\Downarrow}\Gamma_{X\alpha}(2\sigma_{\alpha X}\rho\sigma_{X\alpha} - \sigma_{XX}\rho - \rho\sigma_{XX})/2 + \sum_{\beta=\Downarrow,X}\gamma_\beta(2\sigma_{\beta\beta}\rho\sigma_{\beta\beta} - \sigma_{\beta\beta}\rho - \rho\sigma_{\beta\beta})/2$, where $\sigma_{ij} = \sigma_{ij} \otimes \mathbb{1} = |i\rangle\langle j|$. The rate $\Gamma_{X\alpha}$ describes the radiative decay of $|X\rangle$ into $\alpha = |\Uparrow\rangle, |\Downarrow\rangle$, while γ_β is the pure dephasing rate of state $\beta = |\Downarrow\rangle, |X\rangle$ with respect to $|\Uparrow\rangle$. Since H is block-diagonal, Eq. (3) leads to the closed form

$$\dot{\rho}_{kk'} = \frac{1}{i\hbar}(H_k\rho_{kk'} - \rho_{kk'}H_{k'}) + \mathcal{W}\rho_{kk'}. \quad (4)$$

The diagonal blocks obey the familiar Lindblad equation,

$$\dot{\rho}_{kk} = \mathcal{L}_k\rho_{kk}, \qquad \mathcal{L}_k = -i[H_k, \rho] + \mathcal{W}\rho. \quad (5)$$

Stationary state.—We start with the factorized state $\rho_0 = \chi_0 \otimes \nu_0$, with arbitrary initial density matrices χ_0 and $\nu_0 = \sum_{kk'}\nu_{kk'}|\delta h_z^k\rangle\langle\delta h_z^{k'}|$ of the TLS and the nuclear ensemble, where $|\delta h_z^k\rangle$ are eigenstates of δh_z. We assume a Gaussian $\nu_{kk} = (2\pi)^{-1/2}\sigma^{-1}\exp[-(\delta h_z^k)^2/2\sigma^2]$, with the width $\sigma = \sigma_0 = A/\sqrt{N}$, plotted as a solid line in Fig. 2(a). For our numerics, we choose $A = 90$ μeV, $N \approx 5 \times 10^5$, corresponding to $\sigma_0 \simeq 0.13$ μeV $\simeq 0.2\hbar\Gamma$, with $\Gamma = 1$ ns, and a sample of $n \ll K$ states ($n \sim 4000$) [21]. Because of the hyperfine coupling, the TLS and the nuclei are entangled in the stationary state $\bar{\rho} = \sum_{kk'}\bar{\rho}_{kk'} \otimes |\delta h_z^k\rangle\langle\delta h_z^{k'}|$ with $\dot{\bar{\rho}} = \mathcal{L}\bar{\rho} = 0$. We derived an analytical expression for the 3-by-3 diagonal blocks $\bar{\rho}_{kk}$ of $\bar{\rho}$ as a function of all parameters, including δh_k.

Evolution of the observed system.—In order to enhance the electron-spin coherence, we aim at *narrowing* the nuclear spin distribution ν_{kk}. For a Gaussian distribution, this amounts to decreasing the width σ, thus increasing the electron coherence time $t_0 \simeq 1/2\sigma$. Ideally, we would perform a projective measurement P on the nuclear spins, $P\bar{\rho}_{kk}P \propto \delta(\delta h_z^k - \delta)$. A successive approximation of P is achieved by monitoring the photon emission from the QD. The longer the period t during which no photon is emitted, the higher is the probability for δh_z to be at the two-photon resonance, $\delta h_z = \delta$.

To describe the state of the system conditional on a measurement record, we use the *conditional density matrix*

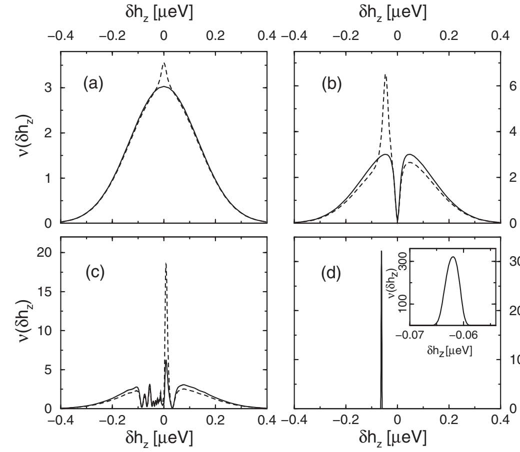

FIG. 2. Conditional evolution of the nuclear spin distribution $\nu(\delta h_z^k) = \nu_{kk}$. (a) During the first period t_1 without photon emission, the initial Gaussian distribution (solid line) develops a peak at the two-photon resonance (dashed line). (b) Change of $\nu(\delta h_z)$ after emission at t_1 (solid line), until before emission time t_2 of the second photon (dashed line). The two-photon resonance δ is shifted to the position of the left maximum (adaptive technique). The depleted region around $\delta h_z^k = 0$ develops at t_1. (c) Analogous situation between t_{11} and t_{12}. (d) $\nu(\delta h_z)$ is obtained after a total time of 10 μs. Inset: Magnification of peak in (d). The width of $\nu(\delta h_z)$ is reduced by a factor of ≈ 100 compared to the initial width in (a). The parameters are $\Omega_c = \Omega_p = 0.2\ \text{ns}^{-1}$, $\Delta = 0$, $\Gamma_{X\uparrow} = \Gamma_{X\downarrow} = 1\ \text{ns}^{-1}$, and $\gamma_\downarrow = \gamma_X = 0.001\ \text{ns}^{-1}$.

ρ^c. In the absence of photon emission, ρ^c obeys Eq. (5) with $\mathcal{L}_k$ replaced by $\mathcal{L}_k - \mathcal{S}$, where the collapse operator $\mathcal{S}$ describes spontaneous emission of the state $|X\rangle$ into $|\uparrow\rangle$ and $|\downarrow\rangle$ with rates $\Gamma_{X\uparrow}$ and $\Gamma_{X\downarrow}$ [22],

$$\dot{\rho}^c_{kk} = (\mathcal{L}_k - \mathcal{S})\rho^c_{kk}, \qquad \mathcal{S}\rho = \sum_{\alpha=\uparrow,\downarrow} \Gamma_{X\alpha}\sigma_{\alpha X}\rho\sigma_{X\alpha}. \quad (6)$$

We have numerically calculated ρ^c in the absence of emitted photons for a duration t. We plot the updated distribution ν_{kk} from $\nu = \text{Tr}_\Lambda \rho^c$ as a dashed line in Fig. 2(a). We find that the *a posteriori* ν_{kk} is concentrated around the two-photon resonance. As the off-diagonal elements (coherences) of ν are constrained by positivity, $|\nu_{kk'}| \leq \sqrt{\nu_{kk}\nu_{k'k'}}$, they are also reduced by the narrowing of ν_{kk}. This process is eventually stopped by a photon emission.

Photon emission.—The stationary emission rate is [22]

$$\Gamma_{\text{em}} = \text{Tr}\mathcal{S}\bar{\rho}(t) = \Gamma\sum_k (\rho_{kk})_{XX}\nu_{kk}, \quad (7)$$

where $\Gamma = \Gamma_{X\uparrow} + \Gamma_{X\downarrow}$. The average photon number during time t is $\langle N_{\text{ph}}\rangle = t\Gamma_{\text{em}}$, and the *a priori* probability for $N_{\text{ph}} = 0$ is, according to Poissonian statistics, $P_{\text{dark}}(t) = \exp(-\Gamma_{\text{em}}t)$. The waiting time distribution for photon emissions is $p_{\text{wait}}(t) = \Gamma_{\text{em}}^{-1}\exp(-\Gamma_{\text{em}}t)$ with mean $\langle t\rangle = \Gamma_{\text{em}}^{-1}$. The narrowing of ν_{kk}, Eqs. (6) and (7), leads to a decreasing Γ_{em} and an increasing $\langle t\rangle$.

With Eq. (7), we find the update rule for ν upon photon emission, $\nu' = \text{Tr}_\Lambda\mathcal{S}\rho^c/\text{Tr}\mathcal{S}\rho^c$, or

$$\nu'_{kk} = \frac{\nu_{kk}(\rho_{kk})_{XX}}{\sum_j \nu_{jj}(\rho_{jj})_{XX}}, \quad (8)$$

where ν_{kk} and $(\rho_{kk})_{XX} = \langle X|\rho_{kk}|X\rangle$ are taken before the emission. The population in the Overhauser field δh_z corresponding to the two-photon resonance $\delta h_z = \delta$ is depleted by the photon emission [Fig. 2(b), solid line].

Adaptive technique.—The stationary, isolated TLS at the two-photon resonance is in a dark state. However, the coupling to the nuclei introduces a nonzero probability for occupation of $|X\rangle$ and for photon emission. Since the detection of a photon provides information about δh_z, the photon emission does not necessarily signify a failed attempt to narrow the nuclear field distribution but can be used as an input for the next weak measurement with adjusted frequencies of the driving lasers, $\omega'_p = \omega_p + \epsilon/2$ and $\omega'_c = \omega_c - \epsilon/2$, so that the new two-photon resonance condition is $\delta h_z = \delta'$, where $\delta' = \delta + \epsilon$ while $\Delta' = \Delta$. We choose ϵ such that the new resonance with the Overhauser field lies in one of the two maxima $\delta h_z^{\max}$ formed after the photon emission; see Fig. 2(b). This situation is described by Eq. (2) with the substitution $\delta \to \delta + \delta h_z^{\max}$. The adaptive technique also works by changing only one of the laser frequencies. Right after the photon emission, the TLS is in one of the single electron states $|\uparrow\rangle$ or $|\downarrow\rangle$. Within a time $1/\Gamma$, much faster than any nuclear time scale, the system will reach the new stationary state. The photon emission from the QD can again be monitored, leading to an enhanced nuclear population at the new resonance [Fig. 2(b), dashed line], thus further narrowing the nuclear distribution. Repeating this procedure leads to a nuclear width that is limited only by the width of the EIT resonance [Figs. 2(c) and 2(d)].

Electron-spin decoherence.—The electron-spin coherence is quantified using the expectation value of the raising operator $S_+(t)$ in a state $|x_+\rangle$ that is prepared perpendicular to the total field $\mathbf{B}_{\text{tot}}$ and is freely precessing about the fluctuating nuclear field δh_z, $\langle S_+(t)\rangle \equiv \langle x_+|S_+(t)|x_+\rangle$. We obtain $\langle S_+(t)\rangle = (\hbar/2)\sum_k \nu_{kk}\exp(it\delta h_z^k)$, which we plot in Fig. 3 at various stages in an adaptive optical measurement scheme. As the off-diagonal elements $\nu_{kk'}$ for $k \neq k'$ do not enter $\langle S_+(t)\rangle$ and Eq. (4) decouples, these results are valid for any ν_0 consistent with the chosen Overhauser field probability distribution. We make a Gaussian fit $\langle S_+(t)\rangle \propto \exp(-t^2/t_0^2)$ for short times t and plot the coherence time t_0 as a function of the total waiting time in Fig. 4. This is the main result of our theoretical analysis: The repeated observation of the QD photon emission and adaptation of the laser frequencies ω_c and ω_p after each photon emission leads to a pronounced enhancement of the electron coherence time, for the realistic parameters chosen, from $t_0 = 5$ ns to ≈ 500 ns within a total observation time of 10 μs.

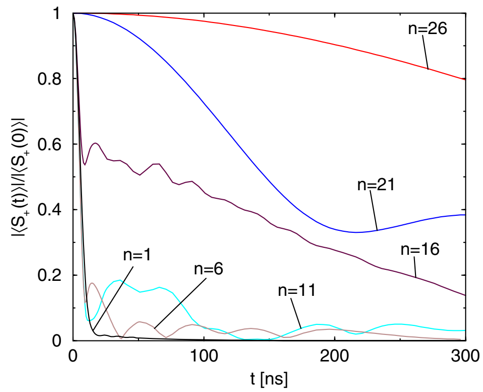

FIG. 3 (color online). Electron coherence function $|\langle S_+(t)\rangle|/|\langle S_+(0)\rangle|$ vs electronic precession time t calculated from $\nu(\delta h_z)$ in Fig. 2 after emission of the nth photon ($n = 1, 6, \ldots, 26$). The initial decay is approximately Gaussian.

Imperfect detectors.—We cannot expect to have perfect photon detectors at our disposal; therefore, we discuss here the case of a detector with efficiency $e < 1$. For an imperfect detector, Eq. (6) becomes $\dot{\rho}^c_{kk} = (\mathcal{L}_k - e\mathcal{S})\rho^c_{kk}$, reflecting that photons are detected with probability e. We have numerically analyzed the case of $e = 10\%$ (other parameters as above) and find $t_0 \approx 460$ ns after a somewhat longer preparation time $t = 50$ μs. This is still much shorter than the time after which the nuclear spin decays, around 0.01 s due to higher-order hyperfine flip-flop terms [17], but possibly longer due to Knight-shift gradient effects. Nuclear flip-flop processes occur on a time scale of ≈ 100 μs [14] but are ineffective in changing h_z in a magnetic field that enforces nuclear spin conservation and, thus, preserve h_z for short-range flip-flops while long-range flip-flops are suppressed by the Knight-shift gradient. This picture is supported by the observed slow ($\gtrsim 1$ s) decay of polarized nuclear spins in contact with donors in GaAs [23]. While a quantitative theory for the relevant time scale of nuclear spin decay due to nuclear-dipole interactions is missing, the arguments given above suggest that our picture of a slow decay is reasonable.

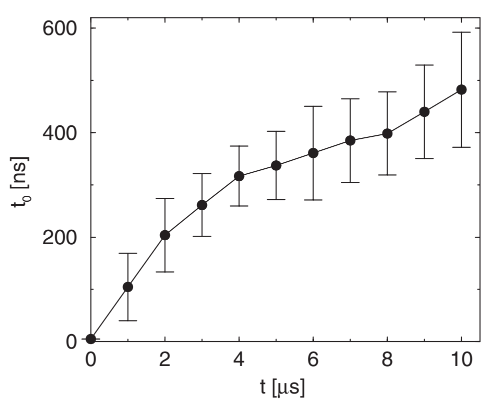

FIG. 4. Characteristic time t_0 of the initial Gaussian decay of $|\langle S_+(t)\rangle|/|\langle S_+(0)\rangle|$ in Fig. 3 as a function of the optical preparation time t, averaged over 50 numerical runs (error bars indicate the standard deviation).

Conclusions.—We find that it is possible to efficiently enhance the quantum phase coherence of an electron spin in a QD surrounded by a large ensemble of nuclear spins by a continuous weak measurement of the Overhauser field using optical excitation at a two-photon resonance of the TLS formed by $|\uparrow\rangle$, $|\downarrow\rangle$, and $|X\rangle$. An intriguing question is whether the electron-spin coherence can be enhanced by a quantum Zeno type effect to the point where it is ultimately determined by spin-orbit interaction: Since the reservoir correlation time of dominant electron-spin decoherence due to flip-flop terms of the hyperfine interaction is ~ 1 μs, this would most likely require high efficiency detection of the scattered photons.

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