Hyperfine Interaction-Dominated Dynamics of Nuclear Spins in Self-Assembled InGaAs Quantum Dots

Christian Latta,* Ajit Srivastava, and Atac Imamoğlu
Institute of Quantum Electronics, ETH-Zürich, CH-8093 Zürich, Switzerland
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We measure the dynamics of nuclear spins in a single-electron charged self-assembled InGaAs quantum dot with negligible nuclear spin diffusion due to dipole-dipole interaction and identify two distinct mechanisms responsible for the decay of the Overhauser field. We attribute a temperature-independent decay lasting ~100 sec at 5 T to intradot diffusion induced by hyperfine-mediated indirect nuclear spin interaction. By repeated polarization of the nuclear spins, this diffusion induced partial decay can be suppressed. We also observe a gate voltage and temperature-dependent decay stemming from cotunneling mediated nuclear spin flips that can be prolonged to ~30 h by adjusting the gate voltage and lowering the temperature to ~200 mK. Our measurements indicate possibilities for exploring quantum dynamics of the central spin model.

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Hyperfine interaction between a single quantum dot (QD) electron and the nuclear spin ensemble defined by the nanoscale confinement provides a realization of the central spin problem [1–9]. This model has attracted considerable attention recently since the correlations between the confined electron and the nuclear spin ensemble induced by hyperfine coupling constitute the principal decoherence mechanism for the electron spin [10]. It has been recognized in this context that an enhancement of electron coherence time could be achieved by preparing nuclear spins in eigenstates of the Overhauser (OH) field operator [11,12]: for this approach to be effective, it is essential to understand and characterize the dynamics of prepared (polarized) nuclear spin states.

In this Letter, we present measurements of nuclear spin dynamics in a single-electron charged self-assembled InGaAs QD. In contrast to prior work in self-assembled and gate-confined QDs [13–16], we probe nuclear spin dynamics when both the exchange coupling to a fermionic reservoir (FR) and the dipolar interaction between nuclear spins are vanishingly small. Our observations reveal a spatially limited, *temperature-independent*, nuclear spin diffusion [17,18] originating from electron mediated nuclear spin interactions. In addition, they unveil a cotunneling mediated, *temperature-dependent*, decay of the OH field approaching 10^5 s. Remarkably, the diffusion induced reduction in the OH field taking place on ~100 s time scale can be strongly suppressed by repeating the preparation cycle consisting of polarization (pump) and free evolution (wait).

The QDs in this sample are separated by a 35 nm GaAs tunnel barrier from a *n*-doped GaAs layer. A bias voltage applied between a top semitransparent Ti/Au Schottky gate and back contacts allows us to control the charging state of the QD and the relative alignment of its electronic levels with the Fermi energy of the FR [19]. High-resolution, modulation-free, resonance-scattering spectroscopy [20] was performed on a single QD in a fiber-based confocal microscope incorporated in a dilution refrigerator. The electron temperature was varied between 200 mK and 4 K while the magnetic field in the Faraday geometry was kept constant at 5 T.

We used a ''pump-probe'' technique to investigate the OH field dynamics. In the first step, the QD nuclear spins were polarized by slowly scanning a single-mode laser across the blue detuned Zeeman resonance of the neutral exciton (X^0): as was shown in Ref. [13], the magnitude of the OH field obtained in such a ''dragging'' experiment is given precisely by the detuning of the applied laser field from the bare resonance. A typical dragging process is shown in Fig. 1(a). After generating an OH field of ~20 μeV with ~40 sec of dragging, the gate voltage is abruptly changed (in <1 msec) to a value V_{wait} that results in the injection of an electron into the QD from the FR [Fig. 1(b)]. Because of the large trion (X^-) energy shift of ~5 meV, the laser field is far off resonance during the waiting time τ_{wait} in which the coupled electron-nuclear system evolves freely. As a last step, the magnitude of the remaining OH field is measured after removing the electron from the QD and rapidly scanning the laser across the transition in ~50 msec. This probing is fast enough not to cause any appreciable dynamic nuclear spin polarization and simply reveals the resonance energy at the time of the measurement. We also confirmed that no appreciable change in OH field takes place during the time needed to switch the gate voltage between V_{pump} and V_{wait}. The ''pump-wait-probe'' sequence is then repeated for different τ_{wait}. A typical OH decay curve obtained using this procedure is shown in Fig. 1(c).

In perfect agreement with earlier measurements [13,21], we found no measurable decay of the OH field for a neutral QD up to 1000 s. This result reconfirms that the OH field in

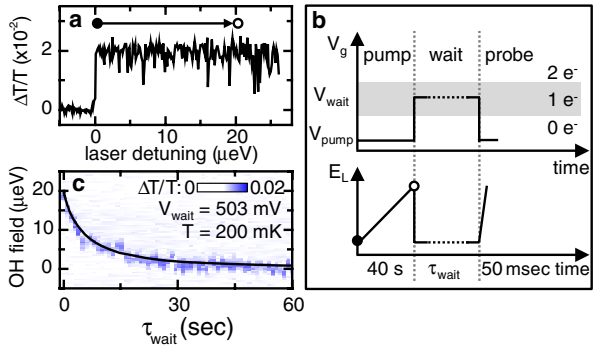

FIG. 1 (color online). (a) Build up of nuclear spin polarization by resonant optical dragging of the neutral exciton at 5 T measured using differential transmission ($\Delta T/T$). (b) Schematic of "pump-probe" technique to measure the OH field decay: (1) A nuclear spin polarization is built up at a voltage V_{pump} by slowly scanning the laser energy E_L (full circle to open circle and back). (2) Gate voltage is set to V_{wait} for a time τ_{wait} keeping the laser energy fixed. (3) Gate voltage is set to V_{pump} again, followed by a fast (50 msec) laser scan to measure the OH field. (c) A typical measurement of the OH field decay at 200 mK in the presence of a resident electron in the strong cotunneling regime. The solid black line shows the calculated result (see text).

self-assembled QDs is stationary in the absence of a confined electron [Fig. 2(b) black triangles]. A possible explanation for this observation is the presence of large and inhomogeneous quadrupolar shifts within and around the QD: recent calculations for self-assembled InAs QD find a mean biaxial strain of 8.6% with a standard deviation of 1.3% [22]yielding a quadrupolar shift of 9.8 MHz with a standard deviation of 6.3 MHz for the As atoms [23]. Outside the QD, the quadrupolar shifts vanish within a length scale of ~3 nm. As a consequence of this strong inhomogeneity in quadrupolar shifts, dipolar-interaction-induced nuclear spin diffusion both within the QD and across its boundary are suppressed. Conversely, the nontrivial OH field dynamics in the presence of an electron

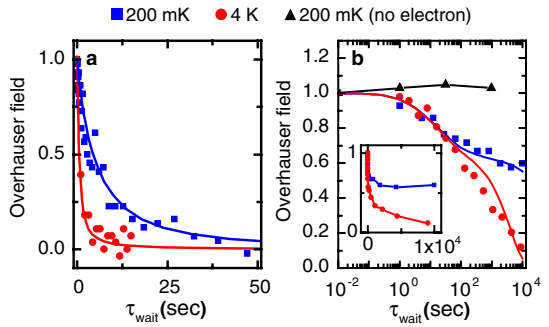

FIG. 2 (color online). (a) Decay of the OH field in a region of strong cotunneling for $V_{\mathrm{wait}} = 505$ mV and different temperatures. (b) Decay of the OH field with negligible cotunneling for the case of no resident electron (▲), a resident electron ($V_{\mathrm{wait}} =$ 530 mV) at 200 mK (■) and 4 K (●). The inset shows the same data in a linear-linear plot.

that we discuss demonstrates that the QD electron-nuclei system is a near-perfect realization of the central spin problem [7].

When we choose V_{wait} such that the single-electron charged QD is in the cotunneling regime [24] and the exchange coupling to the FR is strongest, we find that the OH field exhibits a fast decay [13,14] on the order of a few seconds. The observed decay is clearly temperature dependent [see Fig. 2(a)].

In contrast, the OH field dynamics for V_{wait} that corresponds to negligible electron cotunneling shows decay on two distinct time scales (Fig. 2). The initial decay now takes place on a time scale of ~100 s and is temperature independent. Within this time, only a fraction of the OH field decays; for a single polarization cycle the decaying fraction is approximately 50% [Fig. 2(b) inset]. This initial decay is followed by a temperature-dependent slower decay which varies from 3500 s at 4 K [FIG. 2(b) red dots] to 10^5 s at 200 mK [Fig. 2(b) blue squares].

The presence of two different OH field decay time scales with different temperature dependence points to two independent electron mediated mechanisms. To get further insight, we measured the gate-voltage dependence of the temperature-dependent decay rate across the single-electron charging plateau: in Fig. 3, squares (circles) denote the experimentally measured values of the temperature-dependent decay time at $T = 200$ mK ($T = 4$ K). The values are extracted by fitting an exponential to this decay; the full (open) squares or circles indicate that the measured rate is the faster (slower) component of the OH field decay. The solid blue (red) curves show the gate-voltage dependence of the cotunneling time at $T = 200$ mK ($T = 4$ K) scaled by a (common) constant factor. The measured temperature-dependent decay rates follow the gate-voltage and temperature dependence of the

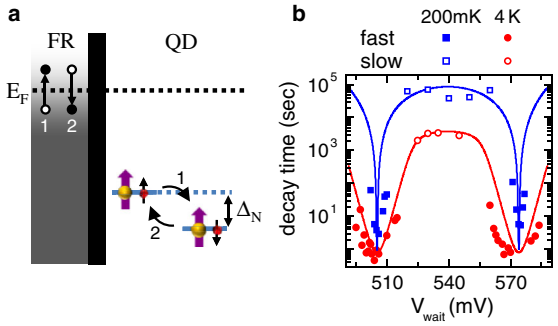

FIG. 3 (color online). (a) Schematic of the temperature-dependent decay process. A nuclear spin can be flipped without flipping the electron spin via noncollinear hyperfine interaction (see text). The energy difference $\Delta_N \ll T$ is provided (1) [absorbed (2)] via a particle-hole annihilation (excitation) in the FR. (b) Extracted decay times of the OH field mediated by cotunneling as a function of gate voltage. The solid lines are the calculated OH decay times (see text).

cotunneling rate across the charging plateau. In fact, we use the expected linear temperature dependence of the depicted cotunneling rate and the good agreement with the experimentally measured decay times to determine our electron temperature to be $T \simeq 200$ mK [25].

To investigate the temperature-independent initial decay of the OH field, we polarized the QD nuclear spins successively in four steps with a waiting time of 200 s in the presence of an electron between each step. The magnitude of the OH field at the end of each polarization cycle was kept the same. As shown in Fig. 4(b), with successive polarization we find that the initial decay of the OH field is practically eliminated. From this observation we conclude that the temperature-independent component of decay arises from "spatially limited diffusion" of nuclear spin polarization. Indeed, with such a polarization scheme, the nuclear spin polarization could diffuse within the QD in each dwell time leading to an overall increase in the polarization. To reach the same magnitude of the OH field in later steps, progressively smaller nuclear polarization is required during dragging. As the diffusion process just redistributes the excess polarization created during each step, one expects to see smaller decay of the OH field with each step, eventually leading to a complete suppression of the diffusion induced decay.

Our measurements thus indicate the presence of two qualitatively different mechanisms determining nuclear spin dynamics: temperature-dependent decay and temperature-independent diffusion of nuclear spin polarization, both mediated by the electron and leading to a decay of the OH field. To explain our findings, we use a model which includes Fermi-contact hyperfine interaction ($\hat{H}_{hyp}$) describing the coupling of the electron spin $\hat{S}$ to $\sim 10^4 - 10^5$ nuclear spins $\hat{I}^i$ of the QD host material, exchange interaction between the QD electron and the electrons in the FR ($\hat{H}_{exch}$), and an effective noncollinear

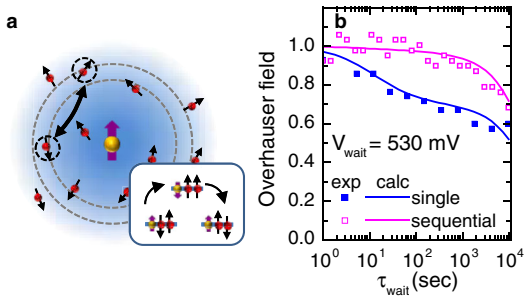

FIG. 4 (color online). (a) Schematic of the electron mediated nuclear spin diffusion: because of the inhomogeneous Knight shifts and quadrupolar fields, only nuclear spins with a small energy difference can interact which is depicted by the dashed circles. Two nuclear spins can flip without flipping the electron spin. (b) Demonstration of the spatially limited nuclear spin diffusion: By sequential polarization of the nuclear spins, the polarization can be saturated, suppressing further nuclear spin diffusion (see text for details).

dipolar hyperfine interaction ($\hat{H}_{dip}$). The total Hamiltonian can then be written as $\hat{H} = \hat{H}_0 + \hat{H}_{hyp} + \hat{H}_{exch} + \hat{H}_{dip}$ where, $\hat{H}_0 = \Delta \hat{S}_z + \sum_i \Delta_N^i \hat{I}_z^i$ is the Zeeman-Hamiltonian with $\Delta = g_e \mu_B B \approx 180$ μeV and Δ_N^i denoting the electronic and nuclear Zeeman energies, respectively. Here we have incorporated the inhomogeneous quadrupolar interaction induced energy shifts of each nucleus in Δ_N^i; since typical quadrupolar fields (< 1 T) are smaller than the external field (5 T), we have $\Delta_N^i \simeq \Delta_N = g_N \mu_N B \approx 0.1$ μeV. $\hat{H}_{exch} = \sum_{k,k'} J_{k,k'} \hat{s}_{k,k'} \cdot \hat{S}$ describes the exchange coupling between the QD electron and the FR with $\hat{s}_{k,k'}$ denoting the spin operator of the FR electrons and $J_{k,k'}$ denoting the exchange interaction strength. This interaction leads to an incoherent electron spin flip rate κ which in our QD is $\sim 10^{-7}$ μeV at 4 K in the center of the charging plateau where the cotunneling is smallest.

The Fermi-contact hyperfine interaction is given by $\hat{H}_{hyp} = \sum_i A_i [\hat{I}_z^i \hat{S}_z + \frac{1}{2}(\hat{I}_+^i \hat{S}_- + \hat{S}_+ \hat{I}_-^i)]$, where $A_i \propto |\psi(\vec{r}_i)|^2 \approx 10^{-2}$ μeV is the hyperfine constant of the ith nucleus with $\psi(\vec{r}_i)$ denoting the QD electron wave function. The first term in $\hat{H}_{hyp}$ is the OH (Knight) field seen by the electron (nuclei) [26]. For large magnetic fields used in our experiments, the flip-flop terms in $\hat{H}_{hyp}$ are ineffective due to the large difference in the electron and nuclear Zeeman energies. Eliminating these terms in Eq. (3) using a Schrieffer-Wolff transformation [27], we obtain new terms describing electron mediated spin flip of two spatially separated nuclear spins:

$$\hat{H}_{ind} = \sum_{i \neq j} \frac{2 A_i A_j}{\Delta - \Delta_N} \hat{I}_-^i \hat{I}_+^j \hat{S}_z. \quad (1)$$

This indirect, coherent long-range interaction leads to a diffusion of nuclear spin polarization within the region where the electron wave function is nonvanishing [Fig. 4(a)]. Although the total QD nuclear spin polarization does not decrease due to $\hat{H}_{ind}$, the OH field seen by the electron decays partially due to a redistribution of the nuclear spin polarization within the QD. We attribute the temperature-independent decay of the OH field to such an electron mediated diffusion.

The last term in the Hamiltonian describes an effective noncollinear hyperfine interaction between the electron and the ith nucleus with coupling constant B_i:

$$\hat{H}_{dip} = \sum_i B_i \hat{I}_x^i \hat{S}_z. \quad (2)$$

Such an effective coupling appears when quadrupolar axes of the QD nuclear spins are not parallel to the external field [28]. The temperature-dependent decay of the OH field can be explained by a second order process originating from $\hat{H}_{dip}$. The energy conservation in this irreversible nuclear spin flip process is ensured by the coupling of the QD electron to the FR [Fig. 3(a)]; the corresponding OH field decay rate is then $(B_i/\Delta_N)^2 \kappa$, which explains the

temperature and gate-voltage dependence of the decay shown in Fig. 3. Since $\Delta_N \ll T$, we expect this rate to be linearly proportional to the electron temperature T. We rule out any contribution of cotunneling assisted direct hyperfine flip-flop processes, since the corresponding rate can be shown to be 4 orders of magnitude slower than the rates that we measure in our experiments.

We model the nuclear spin dynamics using semiclassical rate equations, taking into account the diffusion and decay processes arising from $\hat{H}_{\mathrm{ind}}$ and $\hat{H}_{\mathrm{dip}}$, respectively. For simplicity, we assume a two-dimensional QD with $N = 10^4$ spin 1/2 nuclei. The rate equation describing the change in time of the probability $P_i^{\uparrow}(t)$ that the ith nucleus is in the $|\uparrow\rangle$ state is

$$\frac{dP_i^{\uparrow}(t)}{dt} = \left(\frac{B_i}{\Delta_N}\right)^2 \kappa[1 - 2P_i^{\uparrow}(t)] + \sum_j \left(\frac{2A_iA_j}{\Delta}\right)^2 \rho_{ij}[P_j^{\uparrow}(t) - P_i^{\uparrow}(t)]. \quad (3)$$

The first term on the right-hand side of Eq. (3) represents the temperature-dependent decay while the second term represents the decay induced by the spatially limited diffusion. To obtain Eq. (3), we assume that the coherent coupling of two distant nuclear spins with similar energies via $\hat{H}_{\mathrm{ind}}$ is interrupted by a pure dephasing process with rate γ_{deph}. The Lorentzian factor $\rho_{ij} = \gamma_{\mathrm{deph}}/(\delta_{ij}^2 + \gamma_{\mathrm{deph}}^2)$ describes the effective density of states for the flip-flop process between two (distant) nuclear spins with energy difference $\delta_{ij} = \Delta_N^i + A_i - \Delta_N^j - A_j$.

A possible source of γ_{deph} is the intrinsic gate-voltage fluctuations in our experimental setup; such fluctuations would influence the electron wave function giving rise to an effective broadening of the Knight field experienced by the nuclei. As the bandwidth of this noise is limited by the bandwidth of the gate in our sample (~ 50 kHz), these fluctuations should not affect the decay process which is accompanied by a nuclear spin flip and requires an energy exchange of $\sim\Delta_N \approx 0.1$ μeV. Equation (3) can be solved for a given initial distribution of nuclear polarization which we assume is proportional to $\psi(\vec{r})$ (in turn assumed to be a Gaussian); with the knowledge of $P_i^{\uparrow}(t)$ for all nuclear spins, one can easily get the OH field as $\mathrm{OH}(t) = \sum_i A_i[P_i^{\uparrow}(t) - 0.5]$. For the calculations, we used $\Delta = 174$ μeV, $\Delta_{\mathrm{N}} = 0.1$ μeV, $\sum A_i = 174$ μeV, and $B_i \sim 10^{-2}A_i$ [29]. We first fix the parameters for the case of smallest cotunneling at 4 K ($\kappa = 10^{-7}$ μeV) and then use calculated $\kappa(V_{\mathrm{wait}}, T)$ to obtain $\mathrm{OH}(t)$ for different temperatures and gate voltages. We obtain a value of ~ 2 kHz for γ_{deph} which is well below the bandwidth of the gate. The results of the calculations are plotted in Figs. 1–4 with solid lines.

Our results demonstrate that the nuclear spin dynamics in an InGaAs QD is solely determined by the coupling of each nucleus to the central electron spin. At ultralow temperatures, the OH field decay for a QD well isolated from an electron reservoir is predominantly due to intradot diffusion. By saturating the diffusion process using multiple polarization cycles and reducing the extrinsic (gate-voltage) fluctuations that enhance the diffusion rate, it should be possible to prolong the spin-echo T_2 time of the electron spin [9,30]. In addition, elimination of the dephasing of indirect interaction would open up the possibility for observation of coherent quantum dynamics of the nuclear spins upon an abrupt turn-on of the Fermi-contact hyperfine interaction.

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*Present address: Physics Department, Harvard University, 17 Oxford St., Cambridge, Massachusetts 02138, USA
clatta@physics.harvard.edu

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