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Exchange Control of Nuclear Spin Diffusion in a Double Quantum Dot

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The influence of gate-controlled two-electron exchange on the relaxation of nuclear polarization in small ensembles (N ~ 10^6) of nuclear spins is examined in a GaAs double quantum dot system. Waiting in the (2,0) charge configuration, which has large exchange splitting, reduces the nuclear diffusion rate compared to that of the (1,1) configuration. Matching exchange to Zeeman splitting significantly increases the nuclear diffusion rate.

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Precise control of single electron spins in quantum dots [1] can be used to provide a comparable degree of control of the polarization of small ensembles of nuclei, which couple to single confined electrons via the hyperfine interaction [2–5]. Ultimately, this control may provide a means of storing spin-based quantum information in nuclear ensembles [6,7]. The simplest such process is the induction of an out-of-equilibrium average polarization of the nuclear ensemble by dynamic nuclear polarization (DNP), where the “flip” of a polarized electron spin is accompanied by the “flop” of a nuclear spin [8]. In recent years, experimental studies of DNP have been extended from bulk systems [9,10] to the nanoscale [11–13], including quantum dots containing a small number of electrons [2–4,14–17]. DNP driven by spin-blocked transport can create feedback that leads to hysteretic and complex time-dependent currents [18–23], while gate-driven DNP leads to a simpler buildup of nuclear polarization, often saturating at surprisingly small levels [3,4].

This Letter reports time-dependent measurements of the induction and relaxation of DNP in a few-electron double quantum dot as a function of magnetic field and electron arrangement in the double dot. Cyclic evolution of the two-electron spin state, driven by gate pulses [3], repeatedly flops nuclear spins to create a small local DNP of order 1%. Relaxation is monitored by detecting the Overhauser field averaged over left and right dots, due to nuclear spin blockade ensures that only the (1,1) singlet returns to the nuclear diffusion rate. The mean total effective field experienced by electrons in (1,1) is Btot = B0 + Bnuc, where B0 is the external field applied perpendicular to the two-dimensional electron gas plane and Bnuc = (Bl_nuc + Br_nuc)/2 is the Overhauser field averaged over left and right dots, due to N ~ 10^6 nuclear spins. The avoided crossing between the singlet (S) and the (1,1) m_s = 1 triplet (T+) occurs at a value of 0 [thick, green arrow in Fig. 1(b)] set by the total Zeeman energy, E_tot = g mu_B Btot, where g ~ 0.4 is the electron g factor in GaAs, mu_B is the Bohr magneton, and Btot is the magnitude of Btot. The gap and width of the avoided crossing are set by E_nuc = g mu_B DeltaB_nuc, where DeltaB_nuc is the magnitude of the component of DeltaB_nuc = (Bl_nuc - Br_nuc)/2 transverse to Btot.

We probe the S-T+ resonance using the pulse sequence shown in Fig. 2(b), which first prepares (2,0)S at (P) then separates the electrons (S) for a time T before returning to (2,0) for measurement (M) for time T_M ~ 5 μs. The Pauli spin blockade ensures that only the (1,1) singlet returns to (2,0), with triplets blocked for a time T. In this way, the two-electron spin state is mapped to a charge configuration that is detected with the rf QPC. Cycling this sequence yields a feature at (M) in the (2,0) region, indicated by white lines in Fig. 1(c). Once calibrated, V_{rf} gives the probability 1-P_S that an initial singlet evolved into T+ during the separation interval T. Fitting the time-averaged function P_S(T) gives an inhomogeneous dephasing time.
FIG. 1 (color online). (a) False-color SEM image of a representative double dot with an integrated rf-QPC charge sensor. (b) Energy-level diagram of the two-electron system. The green arrow points to the $S-T_+$ resonance. (c) $V_R$ around the (2,0)-(1,1) charge transition during cycling of the probe sequence. A plane has been subtracted. The region indicated by the black dashed line is a fit to the theoretical Gaussian form [35]. (d) Singlet return probability $P_S$ as a function of separation time $\tau_S$, yielding a $T_2^* \approx 15$ ns. $B_0 = 8$ mT. (d) Singlet return probability $P_S$ as a function of separation time $\tau_S$, yielding a $T_2^* \approx 15$ ns. $B_0 = 8$ mT. (e) Singlet return probability $P_S$ as a function of gate bias $V_L$ and magnetic field $B_0$. The dashed line converts the position of the resonance in $V_L$ to $B_0$.

$T_2^* \approx 15$ ns. The dependence of the $S-T_+$ resonance position (in $V_L$, with $V_R$ fixed) on $B_0$ in the range $B_0 = 5$–18 mT, in the absence of a polarization, serves as a calibration to determine $B_\text{tot}$ when nuclear polarization is present [26].

DNP is investigated using a three-step “pump-pause-probe” sequence: The pump sequence starts from a singlet in (2,0) then moves adiabatically through the $S-T_+$ resonance, flipping an electron and flipping a nuclear spin—in principle, once per cycle at a rate of 4 MHz [3]. The “probe” sequence [Figs. 2(a) and 2(b)] also starts with a singlet in (2,0) but moves to the $S-T_+$ resonance, providing a measure of $B_\text{tot}$. A cycle rate of 200 kHz is used for the probe sequence and does not induce DNP, as seen in Fig. 2(c). Pump and probe cycles are separated by a static “pause” of duration $\Delta t$.

The pump sequence creates a steady-state DNP of order $\sim 10$ mT, which, in the absence of a pause, relaxes during the probing cycle on a time scale $\tau_R = 8$ s, found by fitting an exponential to $B_\text{tot}(t)$ [Fig. 3(c)]. Increasing $B_0$ from 8 mT to 10 mT doubles the time taken for $B_\text{tot}$ to return to $B_0$. This increase in $\tau_R$ with $B_0$ saturates above $B_0 \sim 10$ mT, so that there is little change in relaxation time at $B_0 = 15$ mT compared to the $B_0 = 10$ mT data, consistent with the measured field dependence of nuclear fluctuations [27]. We also note that at $t = 0$, $B_\text{tot}$ appears nearly independent of $B_0$. This suggests that the pump sequence ceases to produce polarization above a certain value of $B_\text{tot}$, qualitatively consistent with previous measurements [3]. The measured relaxation rate cannot account for the small steady-state polarization ($\sim 10$ mT), and we are led to conclude that there must be a significant decrease in the efficiency of the polarization cycle with increasing $B_\text{nuc}$.

The effect of pausing in (2,0) between the pump and probe sequences can be seen in Fig. 4(b), which shows that more than half the polarization remains after pausing for 30 s in (2,0)$S$ [Fig. 4(c)]. Once the probe sequence is initiated after the pause, $B_\text{tot}$ once again decays with $\tau_R \sim 8$ s. The influence of the probe sequence is examined further by introducing multiple pause intervals in (2,0), interleaved with probe cycles [Fig. 4(d)].
perpendicular to the electron gas, we approximate the constant that depends on the two-electron spin state. With these different relaxation times to a nuclear spin diffusion increases diffusion to Fig. 1(e). The red solid curve is an exponential fit. (d) Relaxation yields $R_{\alpha}^{(f)}$ in Fig. 4(f). Pausing for the duration of shown in (b), FIG. 3 (color online). (a) Energy-level diagram near the 104, PRL $A_{\alpha}$ based on an estimate of $14 \text{cm}^2 \text{s}^{-1}$, and hyperfine constant $A$, $(\nu_0$ is the unit cell volume for GaAs and $h$ is Planck’s constant). The change of this shift from site to site in the lattice is given by $\left| \psi(r_i) \right|^2 - \left| \psi(r_j) \right|^2 \sim \alpha \nabla \left| \psi(r) \right|^2|_{r_i}$, where $a$ is the lattice constant. A wave function $\psi(x, y, z) = \phi(x, y)(2\sigma_z^{-3/2} e^{-z/\sigma_z})$ gives a maximum gradient of the Knight shift of $\frac{A_{\alpha}}{h} \left| \phi(x, y) \right|^2 0.92\sigma_z^{-2}$. For a nearest-neighbor distance of like species of 0.565 nm/\sqrt{2} (like species are in a fcc lattice, $a = 0.565 \text{nm}$), we find a Knight shift gradient of 15% of the maximum Knight shift, i.e., $0.15A_{\alpha}/N = 2 \text{kHz}$. This Knight shift is comparable to the random gradient associated with the nuclear dipole-dipole field.

Alternatively, electrons can enhance diffusion via the virtual process of electron-mediated nuclear spin exchange which couples distant nuclear spins [8,25]. To estimate the strength of this process we consider the nuclear field (with $B_{\text{tot}}$ calibrated using $\left| \phi(r) \right|^2$). The pump cycle rate is for the case of pausing in (2,0), consis- tently near the electron Zeeman energy, $g\mu_B B_{\text{tot}}$, giving an effective transverse magnetic field felt by nuclear spin $i$ in a coupling $A_i$. The process is suppressed by the electron Zeeman energy, $g\mu_B B_{\text{tot}}$, which gives $\langle h \gamma_i \rangle^{-1} A_i B_{\text{tot}}^{nuc}$, where $\gamma_i$ is the gyromagnetic ratio of spin $i$. Using the specific values for our device, with $N \approx 6 \times 10^6$ nuclear spins, we find that an enhancement over the intrinsic dipolar field occurs for $B_{\text{tot}} \leq 10B_{\text{tot}}^{nuc} \sim 20 \text{mT}$. The dependence of the nuclear relaxation time $\tau_R$ on the two-electron spin state during the pause duration is shown in Fig. 4(f). Pausing for the duration of $\Delta t$ in the (2,0) state yields $\tau_R = 56 \text{s}$ [red data in Fig. 4(f)], while pausing in (1,1) yields $\tau_R = 26 \text{s}$ [green data in Fig. 4(f)]. We ascribe these different relaxation times to a nuclear spin diffusion constant that depends on the two-electron spin state. With diffusion dominated by the shortest dimension of the dot, perpendicular to the electron gas, we approximate the diffusion constant $D = \sigma_z^{-2}/\tau_R$ based on an estimate of the width of the wave function, $\sigma_z \sim 7 \text{nm}$. This gives $D \sim 1 \times 10^{-14} \text{cm}^2 \text{s}^{-1}$ for the case of pausing in (2,0), consistent with earlier optical measurements of nuclear diffusion in GaAs [10]. Activation of the probe sequence increases diffusion to $D \sim 7 \times 10^{-14} \text{cm}^2 \text{s}^{-1}$.

The presence of strongly confined electrons is expected to affect nuclear spin diffusion in two opposing ways. Suppressing diffusion, electrons couple nonuniformly with the nuclei and create an inhomogeneous Knight shift [28], lifting the degeneracy between nuclear dipoles and preventing them from flip-flopping with each other. The Knight shift gives rise to a frequency shift of order $\lambda_{i} = \frac{A_{\alpha}}{h} \left| \phi(r) \right|^2$ for nuclear spin at position $r_i$, electron wave function $\psi$, and hyperfine constant $A$, $(\nu_0$ is the unit cell volume for GaAs and $h$ is Planck’s constant). The change of this shift from site to site in the lattice is given by $\left[ \left| \psi(r_i) \right|^2 - \left| \psi(r_j) \right|^2 \right] \sim \alpha \nabla \left| \psi(r) \right|^2|_{r_i}$, where $a$ is the lattice constant. A wave function $\psi(x, y, z) = \phi(x, y)(2\sigma_z^{-3/2} e^{-z/\sigma_z})$ gives a maximum gradient of the Knight shift of $\frac{A_{\alpha}}{h} \left| \phi(x, y) \right|^2 0.92\sigma_z^{-2}$. For a nearest-neighbor distance of like species of 0.565 nm/\sqrt{2} (like species are in a fcc lattice, $a = 0.565 \text{nm}$), we find a Knight shift gradient of 15% of the maximum Knight shift, i.e., $0.15A_{\alpha}/N = 2 \text{kHz}$. This Knight shift is comparable to the random gradient associated with the nuclear dipole-dipole field.

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Electron-mediated flipping leads to an increase in diffusion with decreasing $B_{\text{tot}}$, in keeping with the $B_0$ dependence of the data shown in Fig. 3(d). Nonsecular corrections to the nuclear dipole-dipole interaction will also enhance diffusion for $B_{\text{tot}} \lesssim 1 \text{mT}$ [28]. Flipping of nuclear spins via electron cotunneling processes is an additional mechanism that can lead to decay of the polarization; however, for the gate biases used in this experiment,
FIG. 4 (color online). (a) Immediate decay in the position of the resonance during the probe sequence. (b) Decay in the position of the resonance following a pause interval of 30 s in (2,0) between the pump and probe sequences. Pausing in (2,0) suppresses hyperfine coupling. (c) Same as (b), but with the pause interval set to 45 s. (d) Decay of the resonance during probing, interleaved with multiple pause intervals. (e) Decay in the position of the resonance following a pause of 30 s in (1,1). (f) Decay of $B_{\text{tot}}$ as a function of the pause interval $\Delta t$ and for different configurations of a two-electron spin state.

this contribution is expected to be negligible. Varying the tunnel barriers to the leads while keeping the number of electrons on each dot fixed in (2,0) or (1,1) produced little variation in decay time of the polarization. At the $S-T+$ resonance, the exchange energy is effectively “canceled” by the Zeeman energy, allowing rapid flipping of electrons that readily mediate rapid exchange of nuclear spins. This is the likely explanation for the enhanced diffusion observed during the probe sequence.

For our double-dot system, the largest nuclear polarization achievable using a gate pump sequence was shown to be $\sim 1\%$ [3]. Based on our measurement of $\tau_R$, we emphasize that this maximum steady-state DNP cannot be limited by rapid diffusion of polarization out of the dots. Rather, these results indicate that the pump sequence strongly decreases in efficiency with increasing polarization. Such a scenario is consistent with the idea of dark state formation [30], in which the nuclear system is driven to a configuration where it does not interact with the electron spins used in the pump sequence. Hyperfine-mediated nuclear dynamics in quantum dots have been considered theoretically in the context of spin-preserving processes [25,31–34], but measurements of the nuclear relaxation in systems that allow for the removal of a single electron have only recently been reported [2]. For two-electron systems, the measurements presented here bring to light the role of electron exchange, which, as we have shown, can lead to a suppression of hyperfine-mediated nuclear spin diffusion.

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[26] A linear fit of $V_t(B_0)$ is used over the limited range.