

Triplet-singlet spin relaxation via nuclei in a double quantum dot: supplementary information

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I. EXPERIMENTAL SETUP

The experiment is implemented using a double quantum dot with local quantum point contact (QPC) charge sensors (Fig. 1a). These features are formed by metal gates on a GaAs/AlGaAs heterostructure containing a two-dimensional electron gas (2DEG). Voltage pulses are applied to gates L and R using a Tektronix AWG520 arbitrary waveform generator, with a rise time of ≤ 1 ns. The QPC conductances, g_{ls} and g_{rs} , are measured using standard lock-in detection with 1 nA current excitations. g_{ls} and g_{rs} are sensitive to the local charge configuration and exhibit several percent changes when an electron either enters the double dot or moves from one dot to the other. We determine the absolute number of electrons in each quantum dot using charge sensing and tune the device to a region where the left dot contains 0 or 1 electron, while the right dot contains 1 or 2. The device is cooled in a dilution refrigerator to a temperature of about 100 mK and a magnetic field $\mathbf{B}_{ext} = B\mathbf{e}_z$ is applied perpendicular (along the z -axis) to the sample plane.

II. DEFINITION OF B_{nuc}

We start by reviewing the effective magnetic field picture for nuclear spins in a single quantum dot¹, then extend it to the double-dot case. For a single electron in a single quantum dot with large orbital level spacing, $\hbar\omega \gg |g^*\mu_B B|$, we can write a spin hamiltonian for the ground orbital state of the dot, $\psi(\mathbf{r})$, and neglect higher orbital states. The included terms are the Zeeman interaction, $H_B = -g^*\mu_B \mathbf{B} \cdot \hat{\mathbf{S}}$, where $g^* = -0.44$ for GaAs and μ_B is the Bohr magneton, and the hyperfine contact interaction, $H_{HF} = Av_0 \sum_k |\psi(\mathbf{r}_k)|^2 \hat{\mathbf{I}}^k \cdot \hat{\mathbf{S}}$, where A is the hyperfine constant for GaAs, v_0 the unit cell volume, and the sum is over all N lattice sites the electron wavefunction overlaps with. We can define an effective (Overhauser) magnetic field due to nuclei,

$$\mathbf{B}_{nuc} = \frac{Av_0}{-g^*\mu_B} \sum_k |\psi(\mathbf{r}_k)|^2 \hat{\mathbf{I}}^k, \quad (1)$$

such that $H_{tot} = H_B + H_{HF} = -g^*\mu_B(\mathbf{B} + \mathbf{B}_{nuc}) \cdot \hat{\mathbf{S}}$ is the effective spin hamiltonian for a single electron quantum dot ($b_0 = \frac{A}{-g^*\mu_B} \sim 3.5$ tesla for GaAs²). At high temperatures relative to the nuclear Zeeman scale, $T \gg \hbar\gamma_n |\mathbf{B}|/k_B$, where γ_n is the gyromagnetic ratio for a given nuclear spin species, the equilibrium state of each nuclear spin is well described by an identity density matrix, $\rho_k = \mathbf{1}/(2I_0 + 1)$. For $N \gg 1$, the equilibrium expectation values

for \mathbf{B}_{nuc} are gaussian distributed with zero mean and a root-mean-square magnitude^{1,3}

$$\begin{aligned} B_{\text{nuc}} &= \sqrt{\langle |\mathbf{B}_{\text{nuc}}|^2 \rangle} = b_0 [v_0^2 \sum_{kk'} |\psi(\mathbf{r}_k)|^2 |\psi(\mathbf{r}_{k'})|^2 \langle \hat{\mathbf{I}}^k \cdot \hat{\mathbf{I}}^{k'} \rangle]^{1/2} \\ &= b_0 \sqrt{v_0 I_0 (I_0 + 1) \int d^3 r |\psi(\mathbf{r})|^4}. \end{aligned} \quad (2)$$

As the timescale for evolution of the nuclear field (set by nuclear dipole-dipole and back-action of the electron spin on the individual nuclear spins) is much longer than the precession time of the electron in the nuclear field, $\hbar/(g^* \mu_B B_{\text{nuc}})$, we assume the nuclear field to be static during the relevant electron-nuclear interaction time^{1,3}. This assumption is consistent with the kHz linewidths for solid-state NMR on GaAs². Using for $\psi(\mathbf{r})$ in Eq. (2) the ground state wave function for a parabolically confined quantum dot⁴,

$$\psi(\mathbf{r}) = \left[\frac{16}{l^{3/2}} z e^{-4z/l} \left[\frac{\exp\left(-\frac{(x^2+y^2)}{4\sigma^2}\right)}{\sqrt{2\pi\sigma^2}} \right] \right], \quad (3)$$

with lateral dimension σ and thickness l , yields an effective nuclear field $B_{\text{nuc}} = b_0 \sqrt{I_0(I_0 + 1)} \sqrt{\frac{3v_0}{8\pi l \sigma^2}} = 2.3 \text{ tesla} \sqrt{\frac{v_0}{l \sigma^2}} = 18 \text{ mT} \sqrt{\frac{\hbar\omega[\text{meV}]}{l[\text{nm}]}}$, where $\hbar\omega$ is the single-particle level spacing of the dot. Comparing to a uniform wave function of volume V ($\psi(\mathbf{r}) = V^{-1/2}$) gives an effective number of nuclear spins, $N = [v_0 \int d^3 r |\psi(\mathbf{r})|^4]^{-1} = \frac{8\pi l \sigma^2}{3v_0}$ such that $\sqrt{\langle |\mathbf{B}_{\text{nuc}}|^2 \rangle} = \frac{b_0 \sqrt{I_0(I_0 + 1)}}{\sqrt{N}}$.

The spin hamiltonian for a double dot with one electron in each dot and negligible exchange coupling ($J = 0$) is given by

$$H_{(1,1)} = -g^* \mu_B [\mathbf{B}^{(1)} \cdot \hat{\mathbf{S}}^{(1)} + \mathbf{B}^{(2)} \cdot \hat{\mathbf{S}}^{(2)}], \quad (4)$$

where $\mathbf{B}^{(i)} = \mathbf{B}_{\text{ext}} + \mathbf{B}_{\text{nuc}}^{(i)}$ is the total effective magnetic field for dot i . The eigenstates have spins aligned and anti-aligned with these two fields, $|s, s'\rangle$, with eigenenergies $E_{ss'} = -g^* \mu_B (B^{(1)} s + B^{(2)} s')$ and $s, s' = \pm \frac{1}{2}$. Eigenstates of external field (when $B_{\text{nuc}} = 0$) are denoted $|\tilde{s}, \tilde{s}'\rangle$, and are spin aligned/anti-aligned with the total external magnetic field. Without loss of generality we choose the external field to lie along the z axis.

III. FITTING FUNCTION

A. Hyperfine-driven decay

The two-step relaxation process we consider—spin mixing due to nuclei followed by inelastic decay—closely resembles processes well known in the chemical physics literature³ but not previously investigated in the context of quantum dots^{5–8} to our knowledge. In this model, energy relaxation is mediated by a process that couples only to charge, leaving the spin state unchanged. In the absence of spin selection rules, this would couple any (1,1) state to any (0,2) state with a transition rate Γ_{in} that depends on interdot coupling as well as the energy difference between the two charge states (E_{SG}). Such inelastic decay, for example due to phonons, has been studied previously in double-dot systems⁹. For the spin-mixing part, we take $\hbar\Gamma_{in}, J \ll |E_{ss'}|$ and work in the eigenbasis $|s, s'\rangle$ set by local nuclear fields. The decay rate of $|s, s'\rangle$ into the (0,2) singlet, $|G\rangle$, is given by $\Gamma_{ss'} = \Gamma_{in} |\langle s, s' | S \rangle|^2$, i.e., the overlap of $|s, s'\rangle$ with the (1,1) singlet, $|S\rangle$, times the rate of decay of $|S\rangle$ into $|G\rangle$. We remark that this model bears a strong resemblance to the model for a single quantum dot with two electrons of Ref. 5.

We now evaluate the overlap matrix elements, $|\langle s, s' | S \rangle|^2$. A single spin state $|s\rangle$ for a magnetic field $\mathbf{B} = (x, y, z)$ (and of norm $n = \sqrt{x^2 + y^2 + z^2}$) can be expressed in the basis of eigenstates of external field ($|\tilde{s}\rangle$, spin aligned with the z -axis) as

$$|s\rangle = \frac{-(2s)i(n+z)|\tilde{s}=s\rangle - (ix-2sy)|\tilde{s}=-s\rangle}{\sqrt{2n^2+2nz}}. \quad (5)$$

where s and \tilde{s} can take the values $\pm 1/2$. In the tilde-basis, $|S\rangle = (|\frac{\tilde{1}}{2}, -\frac{\tilde{1}}{2}\rangle - |-\frac{\tilde{1}}{2}, \frac{\tilde{1}}{2}\rangle)/\sqrt{2}$. The overlap elements, given a field $B_1 = (x_1, y_1, z_1)$ in dot 1 and $B_2 = (x_2, y_2, z_2)$ in dot 2, are then

$$\left| \left\langle S \left| \pm \frac{1}{2}, \pm \frac{1}{2} \right\rangle \right|^2 = \frac{[(n_1 + z_1)x_2 + x_1(n_2 + z_2)]^2 + [(n_1 + z_1)y_2 + y_1(n_2 + z_2)]^2}{8(n_1^2 + n_1z_1)(n_2^2 + n_2z_2)}, \quad (6)$$

$$\left| \left\langle S \left| \pm \frac{1}{2}, \mp \frac{1}{2} \right\rangle \right|^2 = \frac{[(n_1 + z_1)(n_2 + z_2) + x_1x_2 + y_1y_2]^2 + [-x_1y_2 + y_1x_2]^2}{8(n_1^2 + n_1z_1)(n_2^2 + n_2z_2)}. \quad (7)$$

The states with the same $|m_s|$ value have the same overlap. Averaging over all quasi-static field values, we can find $\langle \Gamma_{ss'} \rangle$, the effective decay rate for an experiment with many different realizations, each with a different field value drawn from the field distribution. As we have taken the external field to be aligned along the z -axis,

$$\begin{aligned} \langle z_i \rangle &= B, \\ \langle x_i \rangle &= \langle y_i \rangle = 0, \\ \langle (\mu_i - \delta_{\mu z} B)(\nu_j - \delta_{\nu z} B) \rangle &= \delta_{ij} \delta_{\mu\nu} B_{\text{nuc}}^2 / 3. \end{aligned}$$

We assume, appropriate to the high temperature approximations used heretofore, that the nuclear fields in dots 1 and 2 are uncorrelated; accordingly, expectation values of two separate fields factorize. This yields

$$\langle \Gamma_{\pm\frac{1}{2}, \pm\frac{1}{2}} \rangle = \Gamma_{in} \langle F \rangle \langle G \rangle \quad (8)$$

$$\langle \Gamma_{\pm\frac{1}{2}, \mp\frac{1}{2}} \rangle = \frac{\Gamma_{in}}{2} (\langle F \rangle^2 + \langle G \rangle^2) \quad (9)$$

where

$$\begin{aligned} \langle F \rangle &= \left\langle \frac{(n+z)^2}{2n^2+2nz} \right\rangle = \left\langle \frac{n+z}{2n} \right\rangle \\ \langle G \rangle &= \left\langle \frac{x^2+y^2}{2n^2+2nz} \right\rangle = \left\langle \frac{n^2-z^2}{2n(n+z)} \right\rangle = \left\langle \frac{n-z}{2n} \right\rangle \end{aligned}$$

are averages over a single dot. We have assumed both dots to be of equal size, having the same effective strength on the average. Transforming to cylindrical coordinates, the integral over a gaussian corresponding to the single-dot nuclear field distribution is

$$I = \left\langle \frac{z}{n} \right\rangle = \frac{1}{[2\pi B_{\text{nuc}}^2/3]^{3/2}} \int_{-\infty}^{\infty} dz \int_0^{\infty} r dr \int_0^{2\pi} d\theta \frac{z \exp[-\frac{r^2+(z-B)^2}{2B_{\text{nuc}}^2/3}]}{\sqrt{r^2+z^2}}. \quad (10)$$

After integration over θ, r and variable change, $u = \sqrt{3/2} \frac{z}{B_{\text{nuc}}}$,

$$I = e^{-3B^2/2B_{\text{nuc}}^2} \int_{-\infty}^{\infty} du u e^{\sqrt{6}uB/B_{\text{nuc}}} \text{Erfc}(|u|). \quad (11)$$

We note that to a good approximation, $I \simeq \frac{\langle z \rangle}{\sqrt{\langle n^2 \rangle}} = \frac{|B|}{\sqrt{B_{\text{nuc}}^2 + B^2}}$ for all external field values, B . This approximation intuitively corresponds to Zeeman-split levels broadened by the nuclear field, and breaks down for larger Γ_{in} or J values. Using this approximation, we find the effective decay rates, used in the main text, from the four eigenstates of the (1,1) charge configuration to the (0,2) singlet state to have two forms, one for the $|m_s| = 1$ states (T_+ , T_-), with a rate $\Gamma_{\pm\frac{1}{2}, \pm\frac{1}{2}} = \frac{\Gamma_{in}}{4} \frac{B_{\text{nuc}}^2}{B_{\text{nuc}}^2 + B^2}$, and the other for the $|m_s| = 0$ states (T_0 , S), with a rate $\Gamma_{\pm\frac{1}{2}, \mp\frac{1}{2}} = \frac{\Gamma_{in}}{2} [1 - \frac{B_{\text{nuc}}^2}{2(B_{\text{nuc}}^2 + B^2)}]$.

B. Thermal component

In addition to the inelastic decay from the (1,1) singlet to the (0,2) singlet, coupling to the leads allows for a spin-independent transition of either $(1,1) \rightarrow (1,2) \rightarrow (0,2)$ or $(1,1) \rightarrow (0,1) \rightarrow (0,2)$ to occur, breaking blockade and reducing the expected signal. As a thermally activated process, the rate for each should depend on the energy difference between the (1,1) state and either (1,2) or (0,1). Denoting this detuning E_T , the corresponding decay has the expected form, $\Gamma_T = \Gamma_0 e^{-E_T/k_B T}$. We note that larger (1,1) to (0,2) detuning E_{SG} corresponds to smaller E_T detuning, as the M point moves closer to the top of the triangle of Fig. 2. This functional form is consistent with the observed high detuning behavior shown in Fig. 4. Combining this decay with the previous section results, it is convenient to define $\tau_0^{-1} = \Gamma_{in}/4 + \Gamma_T$, the zero-field decay time, and $\tau_B^{-1} = \Gamma_{\pm\frac{1}{2}, \pm\frac{1}{2}} + \Gamma_T$, the decay rate of the $|m_s| = 1$ states.

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