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Inhomogeneous nuclear spin flips: Feedback mechanism between electronic states in a double quantum dot and the underlying nuclear spin bath

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We discuss a feedback mechanism between electronic states in a two-electron double quantum dot and the underlying nuclear spin bath. We analyze two pumping cycles for which this feedback provides a force for the Overhauser fields of the two dots to either equilibrate or diverge. Which of these effects is favored depends on the g factor and Overhauser coupling constant of the material. The strength of the effect increases with the ratio of Overhauser coupling to electron exchange energy and also increases as the external magnetic field decreases.

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Hyperfine interaction with the host nuclei in nanoscale GaAs systems, while relatively weak, can nevertheless limit the electron coherence time and thereby complicate strategies to implement quantum information and quantum computing schemes in these systems.[1–4] Conversely, ever-increasing control of angular-momentum transfer between electrons and nuclei in a range of materials enables numerous applications precisely because of the environmental isolation of the nuclear system. These include applications to quantum information processing employing NMR.[5] From the perspective of fundamental physics, experiments on few-electron systems with controllable coupling to the nuclear many-body system uncover a fascinating arena of new phenomena with ramifications for theoretical physics and engineering.[6]

Experiments on double quantum dots with two electrons have uncovered an intriguing phenomenon called the "Pauli blockade"[7] in which two electrons with parallel spins are forbidden from combining in one dot by the exclusion principle. Recent transport and gate pulsing experiments[2,8,9] have exploited this condition to promote and study a spin "flip-flop" process in which angular momentum is exchanged with the local nuclei. A variety of behaviors of the nuclear state, as hyperfine flip flops are repeated, has been observed[8,9] and numerically predicted,[10,11] with reports that such repeated flip flops can cause the Overhauser fields of the two dots either to equalize[8] or to diverge.[9] In these studies a central issue concerns under which circumstances the nuclear spin flops will lead to equilibration of the Overhauser difference field between the two dots and under which it will lead to its divergence. Here we describe a force which, depending on the initial electron state as well as the product of the g factor g and the Overhauser coupling constant A,[12] works to decrease or increase this difference field. We assume that gA is negative, as it is for GaAs. We describe two pulse sequences which differ in the choice of the initial electron state and which consequently have a force tending to cause the Overhauser fields in the two dots to equilibrate or to diverge.

Electronic States of the Double Dot with $N=2$. We calculate the electronic states of the two-electron ($N=2$) double dot within the Hund-Mulliken formalism[13,14] developed for the hydrogen molecule. We focus on the regime in the charge stability diagram[15] where the charge states $(N_L,N_R)=(1,1)$ and (0,2) are close to degeneracy, with N_L, N_R the numbers of electrons on the left, right dots. Typically, in this method, eigenstates of total spin, singlets and triplets, are employed as basis states. However, since we wish to study the inhomogeneous Overhauser effect due to different effective magnetic fields in the two dots, we choose a basis which diagonalizes, at the single-particle level, the z component of this inhomogeneous field and in which the spatial dependence of nuclear spin flips induced by electronic spin "flops" is transparent. The basis is: $\{\xi_n\}\equiv\{|R_\uparrow R_\downarrow\rangle,|L_\uparrow R_\downarrow\rangle,|L_\downarrow R_\uparrow\rangle,|L_\uparrow R_\uparrow\rangle\}$, where L and R indicate the orbital states of the left and right dots and the arrows denote spin direction.[16] Two remaining states of the Hund-Mulliken model, $|L_\downarrow R_\downarrow\rangle$ and $|L_\uparrow L_\downarrow\rangle$ are not relevant to our analysis, the former due to high Zeeman energy and the latter being far away in the charge stability diagram. Note that $|R_\uparrow R_\downarrow\rangle$ is the standard $S(0,2)$ state and $|L_\uparrow R_\uparrow\rangle$ is the standard $|T^+\rangle$ state. The hyperfine Hamiltonian for two electrons is properly written as

$$H_{hf}=\frac{vA}{\hbar^2}\sum_m^M[\delta(\mathbf{r}_1-\mathbf{R}_m)\mathbf{S}_1\cdot\mathbf{I}_m\otimes\mathbf{1}+\mathbf{1}\otimes\delta(\mathbf{r}_2-\mathbf{R}_m)\mathbf{S}_2\cdot\mathbf{I}_m] \tag{1}$$

where $\mathbf{r}_i$ and $\mathbf{S}_i$ are operators in the subspace of electron i (first quantized representation) and m is summed over a total of M nuclei (typically $M\sim10^6$); and where v is the volume per nucleus. We assume, for simplicity, a single nuclear species with spin 1/2. With all nuclei polarized, GaAs has an Overhauser field of approximately $5.3T$.[17] Taking this maximum field as a constraint leads to an average coupling constant $A=270\ \mu$eV.

The standard Hund-Mulliken Hamiltonian applied to a double quantum dot includes single-particle energies and gate voltages [i.e., "detuning" ε (Ref. 15)] for the two dots, tunnel coupling γ, Zeeman energy $E_Z\equiv g\mu_B B_{ext}$, with μ_B the Bohr magneton and B_{ext} the external magnetic field, and Coulomb matrix elements.[13] Including now terms from H_{hf} we get (upper triangle of Hermitian matrix shown),

$$H = \begin{array}{c} \begin{matrix} |R_\uparrow R_\downarrow\rangle & |L_\uparrow R_\downarrow\rangle & |L_\downarrow R_\uparrow\rangle & |L_\uparrow R_\uparrow\rangle \end{matrix} \\ \begin{pmatrix} E_C - \varepsilon & I_z^{LR} - I_z^{RR}\langle L|R\rangle + \gamma & I_z^{LR} - I_z^{RR}\langle L|R\rangle + \gamma & I_+^{RR}\langle L|R\rangle - I_+^{LR} \\ & I_z^{LL} - I_z^{RR} & V_x & I_+^{RR} \\ & & I_z^{RR} - I_z^{LL} & I_+^{LL} \\ & & & I_z^{RR} + I_z^{LL} + E_Z \end{pmatrix} \end{array}, \tag{2}$$

where we have taken the orbital energies of L and R to be zero for simplicity, and we include only two Coulomb terms: the charging energy $E_C \equiv V_{RRRR} - V_{RLRL}$ and the exchange matrix element $V_x \equiv V_{LRRL}$.[18] Equation (2) is written to leading order in the overlap $\langle L|R\rangle$. Higher order terms $(\mathcal{O}(|\langle L|R\rangle|^2))$ occur due to the normalization of the basis states.[13] The matrix elements of H in this electronic basis remain operators in the Hilbert space of the nuclear coordinates,[19]

$$\vec{I}^{\alpha\beta} \equiv v\frac{A}{2\hbar}\sum_{m=1}^{M}\psi_\alpha^*(\mathbf{R}_m)\psi_\beta(\mathbf{R}_m)\vec{I}_m \tag{3}$$

where $\alpha, \beta \in \{L, R\}$. Previous work has generally ignored the transition term I_+^{LR}, which we see from Eq. (2) can lead to a direct transition between $|R_\uparrow R_\downarrow\rangle$ and $|L_\uparrow R_\uparrow\rangle$ and causes a spin flip where the two wavefunctions overlap, in this case *in the barrier*. While such terms could be experimentally important for large B_{ext}, i.e., where $|R_\uparrow R_\downarrow\rangle$ and $|L_\uparrow R_\uparrow\rangle$ undergo an avoided crossing deep in the (0,2) regime, we will, in this Rapid Communication, concentrate on spin-flip-flop processes occurring entirely in the left or right dot. We therefore consider the simpler Hamiltonian with the terms proportional to the L-R overlap omitted.

Nuclear spin-flip location. The crucial feature of Eq. (2) is that the $|L_\uparrow R_\uparrow\rangle$ state is coupled to $|L_\uparrow R_\downarrow\rangle$ via a term which flips a nuclear spin in the right dot (I_+^{RR}) and it is coupled to $|L_\downarrow R_\uparrow\rangle$ by a term that flips a nuclear spin in the left dot (I_+^{LL}). In the *absence* of flip-flop coupling to the $|L_\uparrow R_\uparrow\rangle$ state, the upper left 3×3 matrix in Eq. (2) has an electronic ground state, which we denote as

$$|\Psi\rangle = a(\varepsilon)|R_\uparrow R_\downarrow\rangle + b(\varepsilon)|L_\uparrow R_\downarrow\rangle + c(\varepsilon)|L_\downarrow R_\uparrow\rangle. \tag{4}$$

As shown in Fig. 1, at large (positive) ε, $|\Psi\rangle \to |R_\uparrow R_\downarrow\rangle \equiv S(0,2)$ and at large negative ε, $|\Psi\rangle$ becomes an unequal superposition of $|L_\uparrow R_\downarrow\rangle$ and $|L_\downarrow R_\uparrow\rangle$. Even when $V_x > |\langle I_z^{RR} - I_z^{LL}\rangle|$, the inhomogeneous Overhauser effect will produce a preference for either the $|L_\uparrow R_\downarrow\rangle$ or the $|L_\downarrow R_\uparrow\rangle$ component of Ψ, with the electron down spin preferentially located on the dot with smaller I_z.

As noted above, pulsing experiments[8,9,15] employ the avoided crossing between Ψ and $|L_\uparrow R_\uparrow\rangle$, which is opened up by the hyperfine coupling, to transfer units of angular momentum to the nuclear system. The initial state can be chosen to be Ψ or $|L_\uparrow R_\uparrow\rangle$. This choice, along with the sign of gA, determines the sign of angular-momentum transfer (see double arrow in Fig. 1). The position of the avoided crossing, $\tilde{\varepsilon}$, is determined by the energy of $|L_\uparrow R_\uparrow\rangle$ (see Fig. 1) which is determined by B_{ext}. Insofar as $b(\tilde{\varepsilon}) \neq c(\tilde{\varepsilon})$, a transition *from* Ψ *to* $|L_\uparrow R_\uparrow\rangle$ will preferentially induce a nuclear spin flip down on the side with the larger I_z. This tends to equilibrate the values of I_z^{RR} and I_z^{LL}. Conversely, a transition *from* $|L_\uparrow R_\uparrow\rangle$ *to* Ψ will preferentially cause nuclear spins to flip *up*, but still on the side with the larger I_z, thus leading to a tendency for $|I_z^{LL} - I_z^{RR}|$ to grow.

In the full system, the nuclear state evolves under an inhomogeneous Knight shift due to the interaction with the electron spin during the time the electrons occupy the $|L_\uparrow R_\uparrow\rangle$ state and also includes the effects of the $|L_\downarrow R_\downarrow\rangle$ state. We have considered the elastic limit of the angular-momentum exchange at the avoided crossing; phonon-assisted coupling will inevitably increase the transfer rate.[20] Further, direct dipole-dipole interaction of the nuclei will lead to spin diffusion on longer time scales. Full inclusion of these effects, which greatly enlarge the parameter space of the problem and are at the heart of the experimental picture, is beyond the scope of this work. Here we simply show an important feedback mechanism by which a spin-flip-flop process dependent only on the $I_\pm$ operators can influence the Overhauser difference field $I_z^{LL} - I_z^{RR}$.

For the purpose of focusing on the physics of the feedback mechanism we here assume that the electronic system is initialized in either Ψ or $|L_\uparrow R_\uparrow\rangle$ and driven through the avoided crossing, and it is this process only which alters the nuclear spin state. In the following, we will consider only loading in the Ψ state, though $|L_\uparrow R_\uparrow\rangle$-loading effects follow easily.

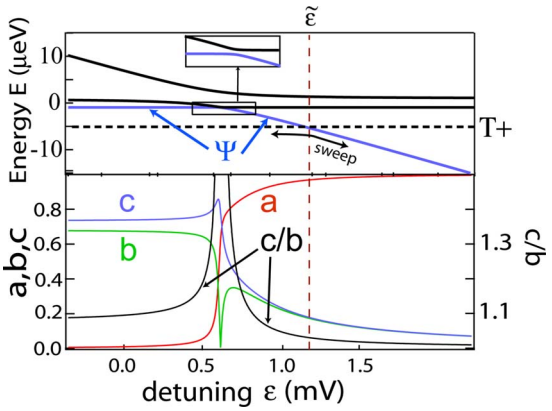

FIG. 1. (Color online) Top: electronic states near the (1,1) to (0,2) stability diagram transition. Crossing of Ψ and $|T^+\rangle$ at $\tilde{\varepsilon}$ becomes *avoided crossing* in presence of transverse Overhauser field gradient. Bottom: overlap of the Ψ state with (a) $S(0,2)$, with (b) $|L_\uparrow R_\downarrow\rangle$, and with (c) $|L_\downarrow R_\uparrow\rangle$. Parameters: $B_{ext}=0.2T$, $\gamma=1.2$ μeV, $\Delta=1000$, $E_C=0.6$ meV, and $V_x=1$ μeV.

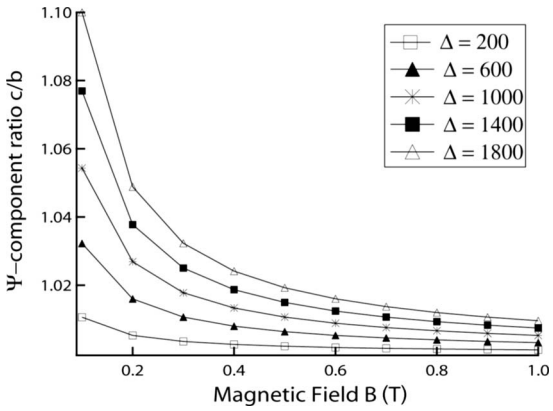

FIG. 2. The wave-function ratio $r \equiv c/b$ evaluated at the Ψ-$|T^+\rangle$ crossing point, $\tilde{\varepsilon}$, as a function of B_{ext} for various values of the nuclear spin z component difference Δ. $r(\tilde{\varepsilon})$ is monotonically increasing with Δ and decreasing with B_{ext}.

Nuclear States. To further analyze the Hamiltonian, Eq. (2), it is helpful to introduce a simplified basis for the nuclear states in which all of the nuclei are either in the left or right dot and all within a given dot interact equally with the electron. In this "box model" $|\psi_L(\mathbf{r})|^2$ is taken as a constant within a spherical "box" of some volume, $\mathcal{V}$. The wave functions then factor out of the sum in Eq. (3) and the squares of the total angular momenta I_α^2 are conserved,[21] where $\vec{I}_\alpha \equiv (vA/\mathcal{V})\Sigma_{m\in\alpha}\vec{I}_m$, and where $\alpha \in \{L,R\}$. Thus, the electrons essentially interact with two composite nuclear spins, one on the left and one on the right. The nuclear state basis is $|I_L,I_R,I_{Lz},I_{Rz}\rangle$, where $I_\alpha(I_\alpha+1)$ is the eigenvalue of $(\vec{I}^{\alpha\alpha})^2$ and $I_{\alpha z}$ is the eigenvalue of $I_z^{\alpha\alpha}$. Finally, for fixed I_L,I_R, it is convenient to transform to the basis of $\Delta \equiv I_{Lz}-I_{Rz}$ and $s \equiv I_{Lz}+I_{Rz}$.

The strength of the preference for left or right dot spin flip depends on the ratio $r \equiv c/b$ at $\tilde{\varepsilon}$. This depends on Δ and on B_{ext}. For example, smaller B_{ext} results in smaller $\tilde{\varepsilon}$, where the ratio c/b increases (for $\Delta > 0$) as shown in Fig. 1. Exactly *how* large c/b can get depends on V_x which, in the example of Fig. 1, we have set to 1 μeV.[22]

In Fig. 2 we plot the value of $r(\tilde{\varepsilon})$ as a function of B_{ext} for various values of Δ. The key point is that $r(\tilde{\varepsilon})$ increases monotonically with Δ, and it also decreases monotonically with B_{ext} (and hence $\tilde{\varepsilon}$). Interestingly, because the $|R_\uparrow R_\downarrow\rangle$ state is coupled equally to $|L_\uparrow R_\downarrow\rangle$ and $|L_\downarrow R_\uparrow\rangle$, the value of b/c is independent of γ.

The flip-flop process naturally also depends on the rate at which ε is swept since the ε variation must be sufficiently slow in order to be adiabatic[15] and remain on the lower branch of the Ψ-$|L_\uparrow R_\uparrow\rangle$ avoided crossing. More generally, the character of the state evolution can be examined as a Landau-Zener tunneling problem.[23]

The evolution of the full nuclear state is complex and the experimental manifestations of that evolution are ambiguous. Nevertheless, as a possible baseline for more detailed studies of the nuclear evolution, we describe a simple, incoherent model which results in narrowing of the distribution of Δ.

If we assume that the system is in the well-defined state $|\Psi\rangle \otimes |I_L,I_R,I_{Lz},I_{Rz}\rangle$ and the detuning is moved quickly to $\tilde{\varepsilon}$ and held there for short time τ, we can compute, to leading order in τ, the probability for a nuclear spin to flip in the right dot as

$$\Gamma_R(I_{Lz},I_{Rz} \to I_{Lz},I_{Rz}-1) \equiv \Gamma_R(s,\Delta \to s-1,\Delta+1) = \frac{\tau^2}{\hbar^2}|\langle I_{Lz},I_{Rz}-1|I_-^{RR}|I_{Lz},I_{Rz}\rangle|^2 = \frac{A^2\Omega_{R-}^2\tau^2}{4\hbar^2}|b|^2 \tag{5}$$

where we have suppressed the I_L, I_R dependence for brevity and where the matrix elements of the ladder operators are given by the well-known formulas: $\Omega_{\alpha\pm} \equiv \langle I_\alpha, I_{\alpha z}\pm 1|I_\pm|I_\alpha,I_{\alpha z}\rangle = \sqrt{I_\alpha(I_\alpha+1)-I_{\alpha z}(I_{\alpha z}\pm 1)}$. Equation (5) is nothing more than the hyperfine-induced transition probability from $|\Psi\rangle \otimes |I_{Lz},I_{Rz}\rangle$ to $|L_\uparrow R_\uparrow\rangle \otimes |I_{Lz},I_{Rz}-1\rangle$. Similarly, the flip probability in the left dot is proportional to the c component of Ψ

$$\Gamma_L(s,\Delta \to s-1,\Delta-1) = \frac{A^2\Omega_{L-}^2\tau^2}{4\hbar^2}|c|^2. \tag{6}$$

Now, assuming a *probability distribution* $W(s,\Delta)$ for the nuclear state (at fixed I_L,I_R), the condition for W to be stationary in its dependence on Δ can be written [cf. Fig. 3(b)],

$$W(s+1,\Delta-1)\Gamma_L(s+1,\Delta-1) = W(s,\Delta)\Gamma_R(s,\Delta),$$

$$W(s,\Delta-1) = W(s,\Delta)\frac{\Omega_{R-}^2(s,\Delta)}{\Omega_{L-}^2(s,\Delta+1)}\frac{|b(\Delta)|^2}{|c(\Delta-1)|^2}, \tag{7}$$

where we have assumed that $W(s) \approx W(s+1)$ and we have used the fact that b and c depend very weakly on s (only through the s-dependence of $\tilde{\varepsilon}$).

Recursion relation Eq. (7) can be solved iteratively and the influence of the narrowing force evaluated. In Fig. 3 we have plotted $W(\Delta)$ computed with the ratio Ω_{R-}/Ω_{L-} set to unity to show only the narrowing from the inhomogeneous Overhauser effect described here with the same electronic parameters as in Fig. 1, and with $I_L=I_R=1000$ (Ref. 24); including the Ω's induces more narrowing. For comparison

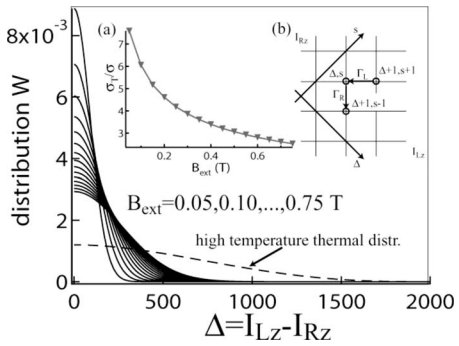

FIG. 3. (main) Reduced distribution $W(\Delta,s)$, calculated from Eq. (7) (solid lines), for $V_x=1$ μeV as a function of Δ for various $B_{ext}=0.05,0.10,\ldots,0.75$ T (lower fields have narrower W); and thermal $W(\Delta)$ (dashed), averaged over s, all with $I_L=I_R=10^3$. Inset (a) narrowing factor $\sigma_T/\sigma(B_{ext})$ versus B_{ext}. Inset (b) Illustration of $I_{Lz}-I_{Rz}$ plane. Δ and s are the diagonal coordinates, with $\Delta \equiv I_{Lz}-I_{Rz}$.

we show the $T \to \infty$ thermal distribution of Δ, averaged over s, also for $I_L = I_R = 1000$. Inset (a) shows the ratio of the root mean square (rms) Δ in the thermal distribution, σ_T, to the rms Δ with the narrowing force at varying B_{ext}, $\sigma(B_{ext})$. A substantial narrowing of $W(\Delta)$ results from the inhomogeneous Overhauser effect.

Discussion. According to the mechanism described here, repeated loading of the Ψ state and pulsing to $|L_\uparrow R_\uparrow\rangle$ should initially drive Δ toward zero ($I_{Lz} = I_{Rz}$). However, nothing in our model forces polarization to stop once $\Delta = 0$, and continued pulsing should ultimately drive $I_{Lz} \to -I_L$ and $I_{Rz} \to -I_R$ or, equivalently, $\Delta = I_R - I_L$ and $s = -I_L - I_R$. The box model approximation breaks down before this polarization is reached, as the hyperfine coupling is not uniform for all of the nuclei. A fuller treatment of the inhomogeneous hyperfine problem is required to determine the evolution of the nuclear system on such timescales.[23]

Pulsing in the opposite direction ($|L_\uparrow R_\uparrow\rangle \to \Psi$) should drive the larger of I_{Lz}, I_{Rz} to grow faster than the smaller, increasing $|\Delta|$. In our model, however, ultimately both spins will fully polarize, and again a fuller treatment is required.

The experimental situation is clouded by indirect access to the nuclear polarizations as well as uncertainty of the influence of other processes on the nuclear spin state. Reilly *et al.*[8] reported a narrowing of $W(\Delta)$ with $|R_\uparrow R_\downarrow\rangle$-loading experiments. However, more recent work[9] presents a complex picture where pulsing in *either* direction produces growth of Δ. We conclude that mechanisms beyond the feedback described here, in particular those affecting the inhomogeneous spin flips *within* each dot, are required for a full understanding of the problem. We note that a recent article by Yao[25] discusses a model similar to that described herein.

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[19] We have also used the identity: $\mathbf{S}\cdot\mathbf{I}_m = S_z I_{mz} + [S_- I_{m+} + S_+ I_{m-}]/2$.

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