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(Article begins on next page)

Dynamic Nuclear Polarization in Double Quantum Dots

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We theoretically investigate the controlled dynamic polarization of lattice nuclear spins in GaAs double quantum dots containing two electrons. Three regimes of long-term dynamics are identified, including the build up of a large difference in the Overhauser fields across the dots, the saturation of the nuclear polarization process associated with formation of so-called “dark states,” and the elimination of the difference field. We show that in the case of unequal dots, build up of difference fields generally accompanies the nuclear polarization process, whereas for nearly identical dots, build up of difference fields competes with polarization saturation in dark states. The elimination of the difference field does not, in general, correspond to a stable steady state of the polarization process.

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Understanding the non-equilibrium quantum dynamics of localized electronic spins interacting with a large number of nuclear spins is an important goal in mesoscopic physics [1–7]. These interactions play a central role in spin-based implementations of quantum information science, in that they determine the coherence properties of electronic spin quantum bits [8]. One of the promising systems for realization of spin-based qubits involves electrically-gated pairs of quantum dots in GaAs, with one electron in each quantum dot (Fig. 1b) [9]. Hyperfine interactions with lattice nuclear spins are the leading mechanism for decoherence of the electron spins, and efforts are currently being directed towards understanding these interactions [10–15], with the ultimate goal of turning the nuclear spins into a resource by controlling these interactions [16–19]. Recent experiments have successfully demonstrated a wide variety of electron-controlled nuclear spin polarization dynamics [19–22], but to date there is no unifying theoretical framework in which to understand the experimental results.

In this Letter we investigate theoretically the process of dynamic nuclear polarization (DNP) in two-electron double quantum dots. This process involves the preparation of the electronic spins in a singlet state and subsequent level crossing between the electronic singlet and triplet states with different projection of electronic angular momentum (Fig. 1a) [20]. It is accompanied by nuclear spin flips, which polarize the spins of the nuclei inside the two dots, producing an effective magnetic (Overhauser) field for the electronic spins. Experiments demonstrate that DNP strongly modifies the difference between the Overhauser fields on the two dots, which is of central importance for control over singlet-triplet qubits [19, 21]. Detailed understanding of DNP in these systems is both of fundamental interest and great practical importance for GaAs based electron spin qubits [23–26].

In what follows we develop a theoretical framework to study the non-equilibrium polarization dynamics of the nuclear spin environment. Our approach takes advantage of the large effective temperature of the nuclear spins and the short time-scale for electron spin evolution to coarse grain the electronic system’s dynamics, yielding a master equation for the nuclear spin degrees of freedom, which we solve in a semiclassical limit. Our key results may be understood by first considering three possible regimes that result from the DNP process. These include i) build-up of an effective difference field, ii) saturation in so-called “dark states,” and iii) preparation of nuclear spins in each quantum dot in states that produce identical Overhauser fields.

For example, i) in the case of two dots with unequal sizes the growth of an Overhauser difference field D_z can be understood in the following heuristic picture, which is borne out by our analytic and numerical calculations. Consider a system with a homogeneous wavefunction in the presence of both strong DNP pumping and nuclear dephasing. The size difference results in different effective hyperfine interactions $g_{\ell(r)}$ on the left(right) dot. We find that the nuclear spins have nearly equal spin flip rates on the two dots, so that the build up of the total Overhauser field S_z is proportional to $g_\ell + g_r$, while the build up of D_z is proportional to $g_\ell - g_r$. Thus, D_z tends to grow with S_z such that $D_z/S_z \rightarrow (g_\ell - g_r)/(g_\ell + g_r)$. On the other hand, ii) when the dots are identical, or nearly so, we find a second regime at strong pumping, where D_z does not grow and the polarization process shuts down the growth of S_z by driving the difference field towards a dark state [27], with $D_x = D_y = 0$. Such states are of interest for use as long-lived quantum memory. Finally, iii) electronic and nuclear degrees of freedom can be completely decoupled if two electrons are initially prepared in the singlet state, while the nuclear spins are

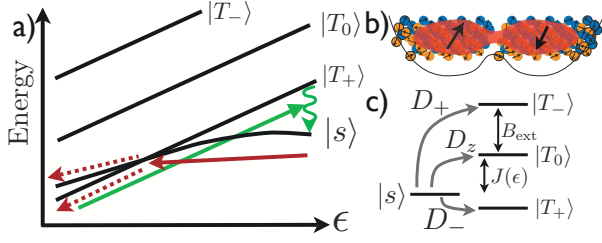


FIG. 1: a) Two-electron energy levels as a function of detuning ϵ between (1, 1) and (0, 2) singlet states. The DNP cycle is illustrated by arrows. b) A double quantum dot with two electrons interacting with a large number of lattice nuclear spins. c) Electronic energy level diagram with transitions from s to triplet states T_+, T_0, T_- driven by Overhauser fields D_-, D_z, D_+ respectively (gray arrows) and energies from external field B_{ext} and exchange splitting J between s and T_0 (black arrows). When $\mathbf{D}_\perp = 0$ electron-nuclear flip-flops are prevented, and when $\mathbf{D} = 0$, electrons and nuclei decouple.

prepared in a state with $\mathbf{D} = 0$ (Fig. 1c). In such a case, polarization stops and the dephasing time of the singlet-triplet qubit can be greatly extended. However, we have not found physical parameter regimes in which such states can be *stably* prepared.

Model – The hyperfine coupling between a localized electron in dot $d = \ell, r$ (for the left, right dot) and a nuclear spin \mathbf{I}_{kd} at \mathbf{r}_{kd} , is given by $g_{kd} = a_{\text{hf}} v_0 |\psi(\mathbf{r}_{kd})|^2$, where ψ is the electron wavefunction, v_0 is the volume per nucleus, and a_{hf} is a coupling constant. The homogeneous limit is defined by $g_{kd} = g_d$ for all k . \mathbf{S} and \mathbf{D} are defined through the collective nuclear spin operators denoting the Overhauser fields in the left (\mathbf{L}) and right (\mathbf{R}) dots $\mathbf{L} = \sum_k g_{k\ell} \mathbf{I}_{k\ell}$ and $\mathbf{R} = \sum_k g_{kr} \mathbf{I}_{kr}$ such that $\mathbf{S} = (\mathbf{L} + \mathbf{R})/2$, $\mathbf{D} = (\mathbf{L} - \mathbf{R})/2$.

For a double quantum dot with two electrons, we can write the Hamiltonian for the lowest energy (1, 1) and (0, 2) electron states, where (n, m) indicates n (m) electrons in the left (right) dot. In this subspace the effective Hamiltonian for the electron and nuclear spins takes the form $H = H_{el} + H_{\text{hf}} + H_n$, where

$$\begin{aligned} H_{el} &= \gamma_e \mathbf{B}_{\text{ext}} \cdot (\mathbf{s}_\ell + \mathbf{s}_r) + J(\epsilon) \mathbf{s}_\ell \cdot \mathbf{s}_r \\ H_{\text{hf}} &= \mathbf{S} \cdot (\mathbf{s}_\ell + \mathbf{s}_r) + \cos \theta(\epsilon) \mathbf{D} \cdot (\mathbf{s}_\ell - \mathbf{s}_r) \\ H_n &= - \sum_{k,d} \gamma_n (\mathbf{B}_{\text{ext}} + \mathbf{h}_{kd}) \cdot \mathbf{I}_{kd} \end{aligned} \quad (1)$$

here $\mathbf{s}_{\ell(r)}$ is the electron spin in the left(right) dot, γ_e (γ_n) is the electron (nuclear) gyromagnetic ratio, where we consider spin 3/2 nuclei of a single species, $\mathbf{B}_{\text{ext}} = B_{\text{ext}} \hat{z}$ is the external magnetic field, $\cos \theta(\epsilon)$ is the overlap of the adiabatic singlet state $|s\rangle$ with the (1, 1) singlet state as a function of the detuning ϵ between the (1, 1) and (0, 2) singlet states, and $J(\epsilon)$ is the splitting between $|s\rangle$ and $|T_0\rangle$ [28]. The rms values of the components of \mathbf{L}, \mathbf{R} in the infinite temperature ensemble are $\Omega_d = (\sum_k g_{kd}^2 I(I+1)/3)^{1/2}$. We

define $\Omega = \sqrt{(\Omega_\ell^2 + \Omega_r^2)/2} \approx (10 \text{ ns})^{-1}$ for typical few-electron double dot experiments, and work in units where $\Omega = -\gamma_e = \hbar = 1$. In addition to the nuclear Zeeman energy we include a “noise” term \mathbf{h}_{kd} , representing the fluctuating, local magnetic field felt by a nuclear spin at site \mathbf{r}_{kd} , which could arise from e.g. nuclear dipole-dipole and electric quadrupole interactions. We estimate the scale of the fluctuations to be such that a typical nuclear spin dephases at a rate of 1-50 kHz [28].

We find the nuclear spin evolution semiclassically by treating the nuclei and electrons as mean fields when solving for the electron and nuclear dynamics, respectively. This semiclassical approximation has been well studied in the context of central spin models and is generally reliable for extracting average quantities of high temperature, low polarization nuclear ensembles in dots with a large number of nuclei N (typically $\approx 10^6$ [28]) [10, 11].

Neglecting H_n , the nuclear spins evolve according to $\dot{\mathbf{I}}_{kd} = i[H_{\text{hf}}, \mathbf{I}_{kd}]$, giving equations of motion

$$\langle \dot{\mathbf{I}}_{kd} \rangle = \frac{g_{kd}}{2} (\langle \mathbf{s}_\ell + \mathbf{s}_r \rangle \pm \cos \theta \langle \mathbf{s}_\ell - \mathbf{s}_r \rangle) \times \langle \mathbf{I}_{kd} \rangle \quad (2)$$

where the top sign applies for $d = \ell$. We now replace $\langle \mathbf{I}_{kd} \rangle$ with \mathbf{I}_{kd} since we are treating the nuclear spins semiclassically. Consider a pulse cycle $\epsilon(t)$ of duration $T \ll 1/g_{kd} \approx \sqrt{N}/\Omega_d$. In a single cycle we can average over the fast evolution of the electrons to arrive at the coarse-grained equations [12]

$$\dot{\mathbf{I}}_{kd}(t) \approx \frac{\mathbf{I}_{kd}(t+T) - \mathbf{I}_{kd}(t)}{T} = g_{kd} \mathbf{P}_d(t) \times \mathbf{I}_{kd}(t), \quad (3)$$

$$\mathbf{P}_d(t) = \int_t^{T+t} \frac{dt'}{2T} [\langle \mathbf{s}_\ell + \mathbf{s}_r \rangle \pm \cos \theta \langle \mathbf{s}_\ell - \mathbf{s}_r \rangle], \quad (4)$$

where \mathbf{P}_d is a slowly-varying, effective Knight magnetic field felt by the nuclear spins.

We now consider the class of pulse sequences employed in Refs. [19, 21], in which the electronic system is initialized in $|s\rangle$ at large ϵ and ϵ is swept slowly through the $|s\rangle$ - $|T_+\rangle$ resonance followed by a fast return to (0, 2) and reset of the electronic state via coupling to the leads. (Fig. 1a). This results in a build up of *negative* polarization. For simplicity, we work in the limit where the electron spin flip probability per cycle is small and calculate \mathbf{P}_d to lowest order in Ω/J , Ω/B_{ext} , ΩT , and Ω/β , where $\beta^2 = \frac{1}{2} |dJ/dt|_{t=t_r}$ is the sweep rate at the resonance time t_r , i.e., $J[\epsilon(t_r)] = B_{\text{ext}}$.

To calculate $\langle \mathbf{s}_d \rangle$ we work in the Heisenberg picture. Defining $\sigma_+^m = |T_m\rangle \langle s|$, we can write $(s_\ell^\pm - s_r^\pm)/2 = (\sigma_\pm^1 - \sigma_\pm^{-1})/\sqrt{2}$ and $(s_\ell^z - s_r^z)/2 = -(\sigma_+^0 + \sigma_-^0)/2$. Since B_{ext} , J , $\beta \gg \Omega$, we can set $\langle |T_n\rangle \langle T_m| \rangle = 0$ in $\langle d\sigma_+^{m'}/dt \rangle$ to obtain the first order corrections to the electronic state:

$$\langle \dot{\sigma}_+^0 \rangle = -i\sqrt{2}v(t)D_z + iJ(t) \langle \sigma_+^0 \rangle, \quad (5)$$

$$\langle \dot{\sigma}_+^{-1} \rangle = -iv(t)D_- + i(J(t) + B_{\text{ext}}) \langle \sigma_+^{-1} \rangle, \quad (6)$$

$$\langle \dot{\sigma}_+^1 \rangle = iv(t)D_+ + i(J(t) - B_{\text{ext}}) \langle \sigma_+^1 \rangle, \quad (7)$$

where $v(t) = \cos\theta(t)/\sqrt{2}$. Since $J, B_{\text{ext}} \gg v\Omega$, Eqs. 5 and 6 can be adiabatically eliminated. To find $\langle\sigma_+^1\rangle$, we formally integrate Eq. 7 and perform a saddle point expansion about the resonance time, assuming $v(t)$ is constant in this region, to reduce it to a Landau-Zener problem [29]. From this solution we calculate the average initial spin flip probability per cycle, $p_{f0} = 2\pi v^2(t_r)\Omega^2/\beta^2$.

Putting these results into Eq. 4 gives

$$\mathbf{P}_d = \pm(\Gamma_0 \hat{z} \times \mathbf{D}_\perp - \Delta_0 D_z \hat{z} - \Delta_- \mathbf{D}_\perp), \quad (8)$$

where $\Gamma_0 = p_{f0}/\Omega^2 T$ arises from the polarization process via T_+ , $\Delta_0 = \langle 2v^2/J \rangle_c$ and $\Delta_- = \langle v^2/(J + B_{\text{ext}}) \rangle_c$ arise from electron-nuclear exchange processes via the T_0 and T_- states, respectively, $\langle \cdot \rangle_c$ indicates an average taken over one cycle, and $\mathbf{D}_\perp = D_x \hat{x} + D_y \hat{y}$.

Qualitatively, the effect of the Γ_0 term is to polarize the nuclear spins, but it also saturates the polarization by driving the nuclear spins into a dark state, $\mathbf{D}_\perp = 0$. The Δ_0 term drives the nuclear spins out of dark states, unless $D_z = 0$ as well. Without noise, states with $\mathbf{D} = 0$ are stationary during this DNP process; we refer to these as “zero states.”

Solving Eqs. 3 with \mathbf{P}_d given by Eq. 8 for an arbitrary electron wave function is a challenging many-body problem. To help treat this problem, we have developed a new numerical method, which is formally equivalent to approximating the wave function by a unique set of $M \ll N$ coupling constants g_{kd} , that well approximates the time evolution of \mathbf{L} and \mathbf{R} for a time that scales as M . A full description of this method, which was used in Fig. 2, along with a discussion of several higher order effects from finite magnetic field and adiabaticity, will be given elsewhere [29].

Unequal dots – Our results that zero states are unstable to the growth of large difference fields, in the presence of asymmetry in the size of the dots and nuclear noise (H_n), can be shown analytically in the case of a simplified model. We assume homogeneous coupling and work in the high field, large J , limit where we can set $\Delta_0 = \Delta_- = 0$ in \mathbf{P}_d . To treat the noise we first go into a frame rotating with the nuclear Larmor frequency, and assume $h_{kd}^{x,y}$ can be rotated away. We further assume that the nuclear noise can be approximated by a Gaussian, uncorrelated white noise spectrum, $\gamma_n^2 \langle h_{kd}^z(t) h_{k'd'}^z(t') \rangle_n = 2\eta \delta(t-t') \delta_{kk'} \delta_{dd'}$, where $\langle \cdot \rangle_n$ are averages over the noise [30]. These local noise processes give rise to a mean decay of the collective nuclear spin variables L_+ (R_+) and associated fluctuations $\mathcal{F}_{\ell(r)}$, defined by $\langle \mathcal{F}_d(t) \mathcal{F}_{d'}^*(t') \rangle_n = 2\Omega_d^2 \delta_{dd'} \delta(t-t')$. As a result, Eqs. 3 and 8, including H_n , give

$$\dot{L}_+ = g_\ell \Gamma_0 L_z (L_+ - R_+) / 2 - \eta L_+ + \sqrt{2\eta} \mathcal{F}_\ell, \quad (9)$$

$$\dot{L}_z = -\frac{g_\ell}{2} \Gamma_0 (L_\perp^2 - \mathbf{R}_\perp \cdot \mathbf{L}_\perp), \quad (10)$$

and similarly for \mathbf{R} . From Eq. 9, if we start in a zero state, \mathcal{F}_d will produce a fluctuation in D_\perp , and the con-

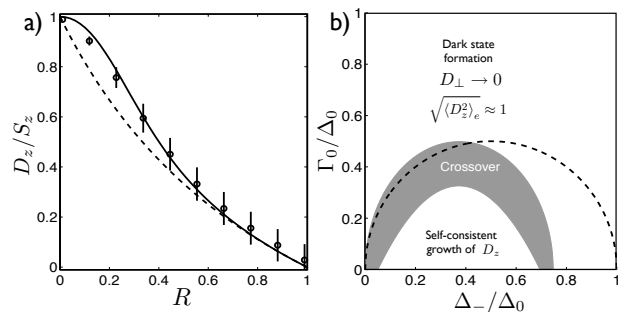


FIG. 2: a) Long time limit of D_z/S_z as the relative hyperfine coupling in the two dots, $R = g_r/g_\ell$, is varied. The solid line is Eq. 12 and the dashed line is $(1-R)/(1+R)$, obtained from a heuristic model (see text). Circles are numerical results with statistical error bars after averaging over an ensemble of 1000 initial conditions, run out to $t = 10^5/g_\ell \Gamma_0 \approx 1$ s, using an approximation to a 2D Gaussian electron wavefunction in terms of 100 coupling constants g_{kd} with noise strength $\eta/g_\ell \Gamma_0 = 10^{-3}$. b) Phase diagram for identical dots for either saturation in dark states or the self-consistent growth of difference fields as the DNP pumping rate (vertical axis) and the Knight shift from $|T_- \rangle$ (horizontal axis) are varied relative to the Knight shift from $|T_0 \rangle$. The dark grey shaded region is a numerical “crossover” regime where both effects occur depending on initial conditions and the dotted line is an analytic result from the simplified model of Eq. 13. For typical polarization cycles $\Delta_-/\Delta_0 \approx 1/4$, but $\Gamma_0/\Delta_0 \approx p_{f0} B_{\text{ext}}/\Omega^2 T$ can be tuned over a broad range.

tribution to \dot{L}_z of the form $-g_\ell \Gamma_0 L_\perp^2$ results, in the long time limit, in $L_z \ll -1$ and similarly for R_z . Thus, $|\dot{L}_z/L_z| \ll 1$ and we can treat L_z, R_z as static to find $\langle L_\perp^2 \rangle_n, \langle R_\perp^2 \rangle_n$ and $\langle \mathbf{L}_\perp \cdot \mathbf{R}_\perp \rangle_n$, which allow us to find the slow evolution of L_z, R_z . To lowest order in $1/S_z$ and $1-R$, where $R \equiv g_r/g_\ell$,

$$\langle \dot{D}_z \rangle_n = -\eta [\langle D_z \rangle_n - (1-R) \langle S_z \rangle_n] / \langle S_z \rangle_n^2, \quad (11)$$

and $\langle S_z \rangle_n = -\sqrt{\eta t}$. This growth of S_z as $t^{1/2}$ is a result of our assumption of delta correlated nuclear noise. If we assume a finite correlation time τ_c such that $\langle \mathcal{F}_d(t) \mathcal{F}_d^*(t') \rangle_n = \Omega_d^2 \exp(-|t-t'|/\tau_c)/\tau_c$, then for $g\Gamma_0 |S_z| \ll 1/\tau_c$, $|S_z| \sim t^{1/2}$, but eventually $|S_z| \sim t^{1/3}$. Integrating Eq. 11 gives $D_z/S_z \rightarrow (1-R)/2$. For general R we find, in the long time limit,

$$\frac{D_z}{S_z} \rightarrow \frac{1-R^2}{2R + \sqrt{4R^2 + (1-R)^4}}. \quad (12)$$

Fig. 2a shows good agreement between these results and numerics for an *inhomogeneous* Gaussian wavefunction.

Identical dots – For identical dots the previous arguments are no longer valid. Fig. 2b, however, shows the results of numerical simulations [29] that demonstrate the existence of a parameter regime for which there is self-consistent growth of D_z even for identical dots. Simulations were performed at each set of parameters by taking

20 different initially polarized nuclear spin configurations with $S_z = -10$, $D_z = -2$, $\eta/g_l\Delta_0$ between $10^{-2} - 10^{-4}$, and a 2D Gaussian electron wavefunction approximated with 400 values of g_{kd} . We determined which parameter values had $\langle D_z \rangle_e$ growing after $t = 10^3/g_l\Delta_0$. For $\Gamma_0/\Delta_0 > 1/2$, no self-consistent growth of D_z appears, and the system approaches a dark state. For smaller Γ_0/Δ_0 and for moderate Δ_-/Δ_0 , continued growth of D_z is observed. We find a similar boundary for unequal dots provided $|1 - R| \lesssim 0.05$.

This phase diagram for identical dots can be verified analytically in a simplified model, where the hyperfine coupling in each dot takes two values ($g_1 \gg g_2$) on two groups of spins of similar size. We assume initially $-g_2 S_z \gg g_1 |D_z| \gg 1 \gg D_\perp$ with the polarization mostly in the strongly coupled spins. To lowest order in g_2/g_1 , $\eta/g_2 D_z$, $g_1 D_z/g_2 S_z$, and D_\perp/D_z , we find [29]

$$\langle \dot{D}_z \rangle_n \propto (\Gamma_0^2 + \Delta_-^2 - \Delta_0 \Delta_-)(g_1 \langle D_z \rangle_n / g_2 \langle S_z \rangle_n)^3. \quad (13)$$

Growth of D_z requires nonzero D_\perp , but, as we show below, for large polarization and weak noise $D_\perp \sim D_z/S_z$, which implies that the growth D_z must occur self-consistently to prevent saturation. This is illustrated by Eq. 13, where the continued growth of D_z is entirely determined by the sign of $\Gamma_0^2 + \Delta_-^2 - \Delta_0 \Delta_-$. For large Γ_0 or strong DNP pumping, the sign is positive, saturation effects dominate, large difference fields are unstable and the system eventually reaches a dark state. For smaller Γ_0 , the sign is negative and coherent evolution arising from interactions with $|T_{0,-}\rangle$ allows D_z to continue growing and D_\perp remains finite. Fig. 2b shows reasonable agreement between this predicted boundary and our numerical results.

We now address the stability of the zero states in the absence of nuclear noise. For identical dots, in the homogeneous limit, Eqs. 3 and 8 give

$$\dot{D}_+ = g i (\Delta_- - i\Gamma_0) S_z D_+ - g i \Delta_0 D_z S_+, \quad (14)$$

$$\dot{D}_z = g [(\Delta_- - i\Gamma_0) D_+ S_- - c.c.] / 2i. \quad (15)$$

Near a zero state \mathbf{S} is constant since $\dot{\mathbf{S}} \sim O(D^2)$. The polarization, $g\Gamma_0 S_z$, acts as a damping term for D_+ ; consequently, for $S_z \ll -1$, $D_+ \rightarrow [\Delta_0 S_+ / (\Delta_- - i\Gamma_0)] D_z / S_z$. Together with Eq. 15 this implies $\dot{D}_z = 0$. Thus the stability matrix, $\partial \dot{D}_\mu / \partial D_\nu |_{D=0}$, has two negative eigenvalues and one zero eigenvalue. Due to this zero eigenvalue, we expect the stability of a zero state to be highly sensitive to external perturbations. We find that inhomogeneous hyperfine coupling, multiple nuclear species, the hybridization of $|s\rangle$ and $|T_0\rangle$ as discussed in Refs. [25, 26], and additional higher order corrections to \mathbf{P}_d in $1/B_{\text{ext}}$ do not, however, break this zero eigenvalue. In the absence of noise, we find numerically that for some parameters a large fraction of initial conditions result in the system spending a long time near a zero state; however,

when we include nuclear noise or higher order corrections in the inverse sweep rate, for example, zero states become repulsive on a long time scale [29]. Throughout this work we have mostly neglected nuclear spin diffusion [30] and spin-orbit coupling [18], both of which could potentially affect DNP and, in particular, the stability of zero states.

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