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.

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 \( g_{\ell y} \) 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 \( g_x + g_z \), while the build up of \( D_z \) is proportional to \( g_y - g_z \). Thus, \( D_z \) tends to grow with \( S_z \) such that \( D_z/S_z \rightarrow (g_x - g_y)/(g_x + g_z) \). 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 \( D_z = D_y = 0 \). 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
prepared in a state with \( D = 0 \) (Fig. 1). 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 \( I_{kd} \) at \( r_{kd} \), is given by \( g_{kd} = a_{kd} v_0 |\psi(r_{kd})|^2 \), where \( \psi \) is the electron wavefunction, \( v_0 \) is the volume per nucleus, and \( a_{kf} \) is a coupling constant. The homogeneous limit is defined by \( g_{kd} = g_d \) for all \( k \). \( S \) and \( D \) are defined through the collective nuclear spin operators denoting the Overhauser fields in the left \( (L) \) and right \( (R) \) dots \( L = \sum_k g_{kd} I_{kd} \) and \( R = \sum_k g_{kr} I_{kr} \), such that \( S = (L + R)/2 \), \( D = (L - R)/2 \).

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
\[
H_{ed} = H_{ed} + H_{hid} + H_n,
\]
where
\[
H_{ed} = \gamma_e B_{ext} \cdot \langle s_l + s_r \rangle + J(\epsilon) s_l \cdot s_r
\]
\[
H_{hid} = S \cdot (s_{l} + s_{r}) + \cos \theta \langle D \cdot (s_{l} - s_{r}) \rangle
\]
\[
H_n = -\sum_{k,d} \gamma_n (B_{ext} + h_{kd}) \cdot I_{kd}
\]
where \( s_{l(r)} \) is the electron spin in the left(right) dot, \( \gamma_e (\gamma_n) \) is the electron (nuclear) gyromagnetic ratio, where we consider spin 3/2 nuclei of a single species, \( B_{ext} = B_{ext} \hat{z} \) is the external magnetic field, \( \cos \theta \langle \epsilon \rangle \) is the overlap of the adiabatic singlet state \( |s\rangle \) with the \((1,1) \) singlet state as a function of the detuning \( \epsilon \) 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 \( L, R \) in the infinite temperature ensemble are \( \Omega_d = (\sum_k g_{kd} I(I + 1)/3)^{1/2} \).

We define \( \Omega = \sqrt{(\Omega_d^2 + \Omega_s^2)/2} \approx (10 \text{ ns})^{-1} \) for typical few-electron double dot experiments, and work in units where \( \Omega = -\gamma_e = h = 1 \). In addition to the nuclear Zeeman energy we include a “noise” term \( h_{kd} \), representing the fluctuating, local magnetic field felt by a nuclear spin at site \( r_{kd} \), 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.[23]

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 \( 10^6 \)[10][11]).

Neglecting \( H_n \), the nuclear spins evolve according to
\[
\{I_{kd}\} = \frac{g_{kd}}{2} \left( |s_{l} + s_{r}\rangle \pm \cos \theta |s_{l} - s_{r}\rangle \right) \times \{I_{kd}\}, \tag{2}
\]
where the top sign applies for \( d = \ell \). We now replace \( \{I_{kd}\} \) with \( \mathcal{I}_{kd} \) 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]
\[
\mathcal{I}_{kd}(t) \approx \frac{I_{kd}(t + T) - I_{kd}(t)}{T} = g_{kd} P_d(t) \times \mathcal{I}_{kd}(t), \tag{3}
\]
\[
P_d(t) = \int_{t'}^{t+iT} dt' \left( |s_{l} + s_{r}\rangle \pm \cos \theta |s_{l} - s_{r}\rangle \right), \tag{4}
\]
where \( P_d \) is a slowly-varying, effective Knight magnetic field felt by the nuclear spins.

We now consider the class of pulse sequences employed in Refs. [13][21], in which the electronic system is initialized in \(|s\rangle \) at large \( \epsilon \) and \( \epsilon \) is swept slowly through the \(|s\rangle\text{-}T_+\rangle \) resonance followed by a fast return to \((0,2) \) and reset of the electronic state via coupling to the leads. (Fig. 1). 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 \( P_d \) to lowest order in \( \Omega/e, \Omega/B_{ext}, \Omega/T, \) and \( \omega/\beta \), where \( \beta^2 = \frac{1}{2} |dJ/d\epsilon| \Gamma_{res} \) is the sweep rate at the resonance time \( \epsilon \), i.e., \( J[e(\epsilon_r)] = B_{ext} \).

To calculate \( \{s_d\} \) we work in the Heisenberg picture. Defining \( \sigma_{z} = [T_m] \{s\} \), we can write \( \langle s_{l}^+ s_{r}^- \rangle / 2 = (\sigma_{z}^+ - \sigma_{z}^-)/\sqrt{2} \) and \( \langle s_{l}^- s_{r}^+ \rangle / 2 = -(\sigma_{z}^+ + \sigma_{z}^-)/2 \). Since \( B_{ext}, J, \beta \gg \Omega \), we can set \( \langle T_n \rangle \{T_m\} \rangle = 0 \) in \( (d\sigma_{z}^+/dt) / dt \) to obtain the first order corrections to the electronic state:
\[
\langle \dot{\sigma}_{z}^+ \rangle = -i\sqrt{2} \nu(t) D_+ + iJ(t) \langle \sigma_{z}^+ \rangle, \tag{5}
\]
\[
\langle \dot{\sigma}_{z}^- \rangle = -i\nu(t) D_+ + iJ(t) + B_{ext} \langle \sigma_{z}^- \rangle, \tag{6}
\]
\[
\langle \dot{\sigma}_{z}^+ \rangle = i\nu(t) D_+ + iJ(t) - B_{ext} \langle \sigma_{z}^+ \rangle, \tag{7}
\]
where \( \nu(t) = \cos \theta(t)/\sqrt{2} \). Since \( J, B_{ext} \gg \nu \), Eqs. 5 and 6 can be adiabatically eliminated. To find \( \langle \sigma_+ \rangle \), we formally integrate Eq. 7 and perform a saddle point expansion about the resonance time, assuming \( \nu(t) \) 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, \( p_{f0} = 2\pi \nu^2(t_\nu)/\beta^2 \).

Putting these results into Eq. 8 gives

\[
P_d = \pm \left( \Gamma_0 \hat{z} \times D_\perp - \Delta_0 D_\perp \hat{z} - \Delta_0 D_\perp \right),
\]

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

Qualitatively, the effect of the \( \Gamma_0 \) term is to polarize the nuclear spins, but it also saturates the polarization by driving the nuclear spins into a dark state, \( D_\perp = 0 \). The \( \Delta_0 \) term drives the nuclear spins out of dark states, unless \( D_s = 0 \) as well. Without noise, states with \( D = 0 \) are stationary during this DNP process; we refer to these as “zero states.”

Solving Eqs. 3 with \( P_d \) 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 \( L \) and \( R \) 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 \( \Delta_0 = \Delta_\perp = 0 \) in \( P_d \). To treat the noise we first go into a frame rotating with the nuclear Larmor frequency, and assume \( h_{k\ell}^{\perp} \) can be rotated away. We further assume that the nuclear noise can be approximated by a Gaussian, uncorrelated white noise spectrum, \( \gamma_n^2(h_{i\ell}^{\perp}(t)h_{k\ell'}^{\perp}(t'))_n = 2\eta \delta(t - t')\delta_{kk'}\delta_{\ell\ell'} \), where \( \langle \rangle_n \) 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 \( F_{i\ell}(\nu) \), defined by \( \langle F_{i\ell}(t) F_{k\ell}(t') \rangle_n = 2\Omega_0^2 \delta_{\ell\ell'}\delta(t - t') \). As a result, Eqs. 3 and 4 including \( H_n \), give

\[
\dot{L}_+ = g_\nu \Gamma_0 \left( L_+ - R_+ \right)/2 - \eta L_+ + \sqrt{2\eta} F_{\nu},
\]

\[
\dot{L}_z = -\frac{\gamma_r}{2} \Gamma_0 \left( L_+^2 - L_\perp \cdot L_\perp \right),
\]

and similarly for \( R \). From Eq. 5 if we start in a zero state, \( F_d \) will produce a fluctuation in \( L_\perp \), and the contribution to \( \dot{L}_z \) of the form \(-g_\nu \Gamma_0 L_+^2 \) results in, in the long time limit, in \( L_z \ll -1 \) and similarly for \( R_z \). Thus, \( |L_z/L_\perp| \ll 1 \) 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 \( \langle L_\perp \cdot R_\perp \rangle_n \), which allow us to find the slow evolution of \( L_z, R_z \). To lowest order in \( 1/S_z \) and \( 1 - R \), where \( R = g_r/g_\nu \),

\[
\langle \dot{L}_z \rangle_n = -\eta \langle (L_+ - (1 - R)) (S_z)_n \rangle / \langle S_z \rangle^2_n,
\]

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 \( \tau_c \) such that \( \langle F_{i\ell}(t) F_{k\ell}(t') \rangle_n = \Omega_0^2 \exp(-|t - t'|/\tau_c) \), 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 \( D_z/S_z \to (1 - R)/2 \). For general \( R \) we find, in the long time limit,

\[
\frac{D_z}{S_z} \to \frac{1 - R^2}{2R + \sqrt{4R^2 + (1 - R)^4}}.
\]

Fig. 2 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. 2, 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 \(S_z = -10, D_z = -2, \eta/g_0\Delta_0\) 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 \(t = 10^3/g_0\Delta_0\). For \(\Gamma_0/\Delta_0 > 1/2\), no self-consistent growth of \(D_z\) appears, and the system approaches a dark state. For smaller \(\Gamma_0/\Delta_0\) and for moderate \(\Delta_0/\Gamma_0\), continued growth of \(D_z\) is observed. We find a similar boundary for unequal dots provided \(|1 - R| \leq 0.05\).

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 \(-g_2S_z \gg g_1/|D_z| \gg 1 \gg D_{\perp}\) with the polarization mostly in the strongly coupled spins. To lowest order in \(g_2/g_1, \eta/g_2D_z, g_1D_z/g_2S_z\), and \(D_{\perp}/D_z\), we find [20]

\[
\langle D_z \rangle_n \propto (\Gamma_0^2 + \Delta_0^2 - \Delta_0\Delta_-)(g_1 \langle D_z \rangle_n/g_2 \langle S_z \rangle_n)^3. \tag{13}
\]

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] which remained constant of \(D_z\) is entirely determined by the sign of \(\Gamma_0^2 + \Delta_0^2 - \Delta_0\Delta_-\). For large \(\Gamma_0\) 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 \(\Gamma_0\), 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. [2] 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

\[
\dot{S}_z = g_i(\Delta_+ - g_0S_z)D_+ - g_i^2D_z S_+, \tag{14}
\]

\[
\dot{D}_z = \frac{g_i(\Delta_+ - g_0)D_+S_- - c.c.}{2i}. \tag{15}
\]

Near a zero state \(S\) is constant since \(\dot{S} \sim O(D^2)\). The polarization, \(g_i\Gamma_0S_z\), acts as a damping term for \(D_+\); consequently, for \(S_z \ll -1, D_+ \rightarrow |\Delta_0S_z/(\Delta_+ - g_0)|D_z/S_z\). Together with Eq. [13] this implies \(D_z = 0\). Thus the stability matrix, \(\partial D_p/\partial D_z|_{D_z=0}\), 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. [22][23], and additional higher order corrections to \(P_2\) in \(1/B_{ext}\) 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 [24]. 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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[8] L. A. Pappalardo, Pappalardo, NSF, the Physics Frontier Center, and the ARO.