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Self-Sustaining Dynamical Nuclear Polarization Oscillations in Quantum Dots

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Early experiments on spin-blockaded double quantum dots revealed robust, large-amplitude current oscillations in the presence of a static (dc) source-drain bias. Despite experimental evidence implicating dynamical nuclear polarization, the mechanism has remained a mystery. Here we introduce a minimal albeit realistic model of coupled electron and nuclear spin dynamics which supports self-sustained oscillations. Our mechanism relies on a nuclear spin analog of the tunneling magnetoresistance phenomenon (spin-dependent tunneling rates in the presence of an inhomogeneous Overhauser field) and nuclear spin diffusion, which governs dynamics of the spatial profile of nuclear polarization. The proposed framework naturally explains the differences in phenomenology between vertical and lateral quantum dot structures as well as the extremely long oscillation periods.

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The coupling of electron and nuclear spin dynamics is responsible for a wide variety of intriguing transport phenomena in semiconductor devices. Electron-nuclear spin exchange is crucial in systems such as spin-blockaded quantum dots, where transport is highly sensitive to spin selection rules [1–7]. Furthermore, the nuclear spins produce a hyperfine (Overhauser) field that shifts the electronic Zeeman energy. This field, which can reach a few tesla for fully polarized nuclei, can have dramatic consequences for transport in double quantum dots (DQDs), where discrete levels may be shifted into or out of resonance [4,8–10]. The combination of these two effects—electron-nuclear spin exchange, which polarizes nuclear spins, and subsequent backaction on energy-dependent spin-flip rates—gives rise to interesting nonlinear dynamical effects such as multistability, hysteresis, and intermittency [8–15].

Perhaps the most striking phenomenon observed in DQDs is the appearance of spontaneous, stable current oscillations under the application of a dc source-drain bias [8,16]. The oscillations occur with very long periods ranging from seconds to hundreds of seconds. These time scales are 10^7–10^9 times longer than the microscopic time scales associated with single electron transit, $(1\ \mathrm{pA})/e \sim 100$ ns, and are also considerably longer than the time scales arising from coherent oscillation mechanisms [17]. The oscillations are also accompanied by long transients when the source-drain bias is changed. After many years of experiments by different groups, the oscillations have only ever been seen in *vertical* DQDs; they have never been observed in gate-defined lateral DQDs.

The strong influence of nuclear magnetic resonance on the oscillations indicates that nuclear spin dynamics play a key role [8]. However, despite wide interest in the problem, a viable mechanism has thus far remained elusive. Here we present a straightforward mechanism which produces oscillations with similar phenomenology. The mechanism relies on nuclear spin diffusion [18,19] and on spin-dependent tunneling rates [20] which are controlled by the spatial profile of the Overhauser field diffusing into the barrier.

Nuclear spin diffusion, being a slow process, introduces the correct time scale into the dynamics. The time scale is set by the process of nuclear polarization diffusing from the dot into the barrier. The corresponding length scale in vertical DQDs is set by the combination of barrier and quantum well half-widths, which is typically a few tens of nanometers. For typical diffusion parameters [18,19] this translates into a time scale on the order of 10 s, consistent with the observed oscillation periods [8,16].

While the mechanism is robust for vertical DQDs, the phenomenology is expected to be quite different for lateral

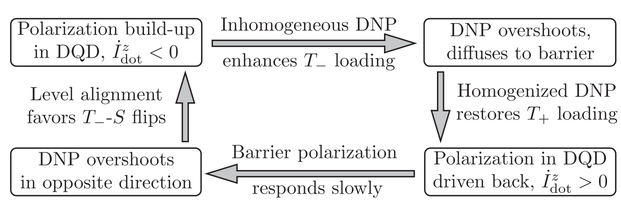

FIG. 1. Mechanism of dynamical nuclear polarization (DNP) oscillations in a spin-blockaded double quantum dot. Polarization is driven on a short time scale by resonant hyperfine transitions inside the DQD. Spin injection in the presence of an inhomogeneous Overhauser field leads to a polarization overshoot in the dot. Nuclear spin diffusion homogenizes the Overhauser field on a much longer time scale. Spin-flip transition rates inside the DQD adapt but lead to an overshoot in the opposite direction.

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DQDs. First, because spin diffusion is isotropic in the semiconductor material surrounding the DQD, the nuclear polarization reaching the barrier is much weaker for lateral DQDs than for vertical DQDs. Second, the dot-barrier distance is over 10 times that for vertical DQDs, leading to a 10^2–10^3-fold increase in diffusion times. These times, ranging from tens of minutes to several hours, can exceed spin relaxation times. In this case, nuclear polarization generated in a dot would not reach the barrier, rendering the feedback effect via polarization-dependent tunneling ineffective. This is consistent with the observation that oscillations are frequently observed in vertical structures but never in the lateral structures.

Schematically, oscillations arise as described in Fig. 1. An initial imbalance of hyperfine spin-flip rates for up and down electron spins leads to a fast buildup of nuclear polarization inside the DQD. The resulting inhomogeneity of the Overhauser field between the DQD and its surroundings *enhances* the probability of injecting the spins with the dominant hyperfine rate. This causes the polarization inside the dot to "overshoot." On a much longer time scale, nuclear spin diffusion homogenizes the Overhauser field. As the spin-injection probabilities react accordingly, the balance of hyperfine transition rates inside the DQD reverses and starts to drive the nuclear polarization in the dot back toward zero. In a similar way, the polarization inside the dot again overshoots and then the cycle repeats.

Here we describe the coupled electron and nuclear spin dynamics through a model which, in our opinion, strikes an appropriate balance between simplicity and completeness in capturing the behavior of the essential degrees of freedom. We will write a set of dynamical equations for two polarization variables, one representing the polarization within the DQD, and one representing the polarization under the tunnel barrier to the source lead. The intradot polarization variable is driven by hyperfine spin-flip processes with electron spins within the DQD. Polarization is then transferred to the barrier via spin diffusion with a large time constant. The delayed reaction of the barrier polarization variable to the intradot spin dynamics leads to oscillations as outlined above.

A key to our mechanism is the difference in probabilities for spin-up and spin-down electrons to tunnel into the quantum dot when it is empty [20]. Naively, one might expect the respective tunneling rates to differ due to the application of a homogeneous Zeeman field, since the final state energies are different. However, in this gedankenexperiment, spin-up and spin-down electrons would tunnel under identical Zeeman-shifted barriers. Provided that the dot levels are set far below the chemical potential of the lead E_F, and that the g factor is homogeneous, the rates are not imbalanced by a homogeneous field.

A very different situation arises for an inhomogeneous Overhauser field. For demonstration, consider the case shown in Fig. 2(a), where the nuclear polarization is large

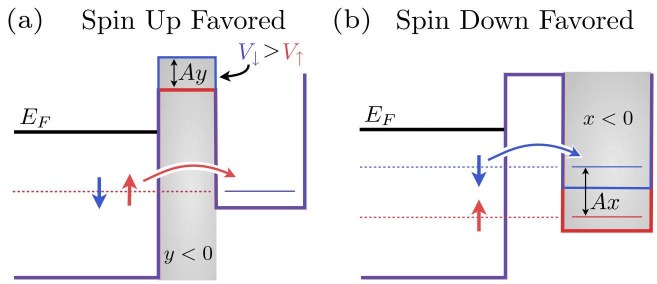

FIG. 2 (color online). Spin-dependent tunneling due to inhomogeneous Overhauser field. (a) When nuclear polarization is localized only under the barrier, $x = 0$, $y \neq 0$, up and down spins are subjected to different barriers ($B = 0$ for illustration). (b) When nuclear polarization is localized inside the dot, $x \neq 0$, $y = 0$, up and down spins tunnel in at different relative energies.

and negative under the barrier, and zero outside. Here the Overhauser field locally *increases* the Zeeman energy under the barrier, effectively creating a higher barrier for down spins, and a lower barrier for up spins. In this situation, an empty dot is more likely to be filled by a spin-up electron than by a spin-down electron. The tunneling rates can also be imbalanced by nuclear polarization localized inside the DQD, which alters the tunneling energies. Here, negative polarization favors tunneling of down spins [see Fig. 2(b)].

It is interesting to note the similarity between this effect and the phenomenon of tunneling magnetoresistance (TMR) [21,22]. In both cases transport is dominated by tunneling through a barrier, and spin polarization is used as a knob to control tunneling rate. While in TMR the spin polarization is due to magnetization in the regions surrounding the barrier, in our case the dominant effect is due to underbarrier nuclear spin polarization. The discovery of TMR has had important consequences for magnetic memory applications. One can envision that some of these ideas can be transposed to DQD systems.

We now consider sequential electron transport through a spin-blockaded double quantum dot connected to leads with an applied dc source-drain bias, as, e.g., in Refs. [8,11,12]. In the two-electron spin-blockade regime, "(1, 1)" orbital configurations with one electron in each dot and a "(0, 2)" configuration with both electrons in the second dot have nearly the same electrostatic energies; see Fig. 3(a). In the (1, 1) configuration, where overlap between electrons is negligible, all four spin states (one singlet and three triplet states) are nearly degenerate in energy. For the (0, 2) configuration, however, only the spin singlet configuration is allowed due to the Pauli exclusion principle (the single dot orbital level spacing is assumed to be much larger than the applied bias). Interdot tunneling hybridizes the (1, 1) and (0, 2) singlet states, producing the states labeled $|S\rangle$ and $|S'\rangle$ in Fig. 3.

Tunneling out of the double dot occurs from the (0, 2) singlet state, which is coupled to the drain lead. Through

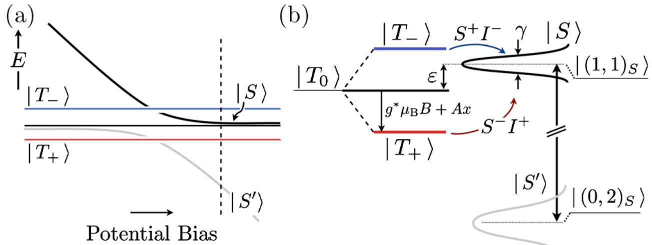

FIG. 3 (color online). Two-electron energy levels involved in spin-blockaded transport (adapted from Ref. [23]). (a) As a function of potential bias, which controls the asymmetry of the double well potential, the (1, 1) and (0, 2) singlet states exhibit an anticrossing. (b) Energy levels at large detuning, indicated by the dashed vertical line in (a). The singlet levels are broadened due to the coupling of the (0, 2) state to the drain lead. Hyperfine-assisted transitions from $|T_\pm\rangle$ to $|S\rangle$ provide a source for the nuclear polarization x within the DQD.

hybridization, both singlet states $|S\rangle$ and $|S'\rangle$ acquire finite lifetimes, reflected in their broadened line shapes [Fig. 3(b)]. When only spin-conserving tunneling is taken into account, the triplet states remain decoupled from the drain. Therefore the rate-limiting step which controls current through this system is the decay of the long-lived triplet states through resonant hyperfine-assisted transitions to the singlet states $|S\rangle$ and $|S'\rangle$, or higher order processes which may also break the conservation of spin within the double dot [6]. Hyperfine assisted transitions from $|T_\pm\rangle$ to $|S\rangle$ and $|S'\rangle$ transfer angular momentum from electron to nuclear spins, thus driving the nuclear polarization dynamics.

Here we focus on the regime of large detuning where $|S'\rangle$ is far separated in energy from the triplet states and can be ignored in the calculation of hyperfine-assisted triplet-singlet transitions. We seek a coupled set of dynamical equations in two polarization variables. The first variable represents the net polarization of the DQD. We define the fractional polarization in the DQD as $x = (N_+ - N_-)/(N_+ + N_-)$, where N_+ (N_-) is the number of nuclear spins oriented along (against) the external field. For a typical device, $N \equiv N_+ + N_- \approx 10^6$. The second variable, which we denote by y, represents the fractional polarization within the tunnel barrier connecting the source lead to the first dot.

The intradot polarization x controls feedback through the Overhauser shift of the electronic triplet levels, which can bring these levels into or out of resonance with the singlet. The energies $\varepsilon_\pm$ of the triplet states $|T_\pm\rangle$, relative to the energy of $|S\rangle$, are given by

$$\varepsilon_\pm = \varepsilon \pm g^* \mu_B B \pm Ax, \tag{1}$$

where ε is the singlet-triplet detuning [$\varepsilon < 0$ in Fig. 3(b)], g^* is the effective g factor ($g^* \approx -0.44$ in GaAs), μ_B is the Bohr magneton, B is the magnetic field strength, and $A \sim 100$ μeV is the hyperfine coupling constant.

Each time an electron decays from $|T_+\rangle$ or $|T_-\rangle$ to $|S\rangle$ via hyperfine exchange, one nuclear spin is flipped from down to up or up to down, respectively. The probability for an electron that enters the dot to cause a positive (negative) increment to the nuclear polarization during its escape is determined by the probability f_+ (f_-) that the electron entered into the state $|T_+\rangle$ ($|T_-\rangle$), and by the probability that the electron escapes via the hyperfine exchange process rather than by alternative nuclear-spin-independent mechanisms [11,23]. The hyperfine spin-flip probabilities are determined by the ratios $W^{\rm hf}_\pm/(W^{\rm hf}_\pm + W^*)$, where $W^{\rm hf}_\pm$ is the hyperfine decay rate of $|T_\pm\rangle$ and W^* describes the net effect of various nonhyperfine processes (spin-orbit coupling, spin exchange with the leads, and cotunneling, etc.).

In our model, we assume that all nuclear spin flips due to hyperfine exchange with the electrons occur *within* the DQD. Therefore, the dot polarization x receives kicks (with magnitude $1/N$) on the time scale of single electron hopping through the dot, 100 ns to 1 μs, while the barrier polarization y has no dynamics on this small time scale. On a much longer time scale, nuclear polarization may diffuse from the dot region into the barrier region, providing a source for y.

Mathematically, it is simplest to analyze the regime where $W^* \gg W^{\rm hf}_\pm$. Here the total current, i.e., the effective frequency of electrons passing through the double dot, is determined by W^*. Additionally, the hyperfine decay probabilities reduce to $W^{\rm hf}_\pm/W^*$. The dependence on W^* cancels from the nuclear polarization *rate*, which depends on products of attempt frequencies and spin flip-probabilities, leaving behind contributions proportional to the hyperfine rates $W^{\rm hf}_\pm$ weighted by the loading probabilities $f_\pm$:

$$\dot{x} = (f_+ W^{\rm hf}_+ - f_- W^{\rm hf}_-)/N - 2\Gamma_D(x - y), \tag{2}$$

$$\dot{y} = -2\Gamma_D y + \Gamma_D x, \tag{3}$$

where $\Gamma_D \sim 0.1$ s^{-1} is the inverse of the time constant for diffusion from the dot to the barrier. The hyperfine spin-flip rates $W^{\rm hf}_\pm$ are given by Fermi's golden rule [11]:

$$W^{\rm hf}_\pm = \frac{A^2}{N} \frac{(1 \mp x)\gamma}{\varepsilon_\pm^2 + \gamma^2}, \tag{4}$$

where γ is the decay rate of $|S\rangle$ due to its coupling to the drain. We account for the dependence of the loading probabilities $f_\pm$ on the Overhauser field inhomogeneity in a lowest-order expansion in x and y:

$$f_\pm = \frac{1}{4}[1 \pm \eta(x - y)], \tag{5}$$

where η controls the sensitivity of the loading probabilities to a polarization gradient. The factors of 2 in front of Γ_D in Eqs. (2) and (3) account for the fact that polarization diffuses in both directions (up and down).

Under what conditions might we expect to find oscillations in the flow defined by Eqs. (2) and (3)? Typically, oscillations are found when the linearized system has the form of an "unstable spiral":

$$\dot{u} = \alpha u + v, \qquad \dot{v} = -\mu u + \beta v, \tag{6}$$

with $\alpha + \beta > 0$ and $(\alpha - \beta)^2 - 4\mu < 0$. These conditions ensure that the eigenvalues are complex, with positive real part. Comparing with Eq. (3), we see that $\dot{y} \sim x$, with a *positive* coefficient of x due to the fact that polarization preserves its sign as it flows into the barrier. Therefore, we need the coefficient of y in Eq. (2) to be *negative*. Substituting expression (5) for $f_\pm$ into Eq. (2), this gives a condition $\eta > 4\Gamma_D/W_0$, where W_0 is the hyperfine spin-flip rate at the unstable fixed point.

Going further, we can expand Eqs. (2) and (3) in the deviations $\tilde{x}$ and $\tilde{y}$ from the (unstable) fixed point of the nonlinear system. Notably, because y only appears to linear order in the original expressions, only $\tilde{y}$-independent or $\tilde{y}$-linear terms show up in the expansion. In general, all other terms appear:

$$\dot{\tilde{x}} \approx c_{10}\tilde{x} + c_{01}\tilde{y} + \cdots, \qquad \dot{\tilde{y}} = \Gamma_D\tilde{x} - 2\Gamma_D\tilde{y}, \tag{7}$$

where the dots represent higher order terms $c_{20}\tilde{x}^2 + c_{11}\tilde{x}\,\tilde{y} + c_{30}\tilde{x}^3 + c_{21}\tilde{x}^2\tilde{y} + \cdots$. Comparing to Eq. (6), we see that we need $c_{10} > 2\Gamma_D > 0$ to ensure a positive real part of the eigenvalues, and $c_{01} < 0$, $(c_{10} + 2\Gamma_D)^2 < 4|c_{01}|\Gamma_D$ to ensure a negative discriminant. These considerations lead to the oscillatory regime shown in Fig. 4.

Using the vast separation of time scales between the hyperfine spin-flip driven polarization dynamics and the slow diffusion processes, we explore another avenue of analysis. Assuming that the barrier polarization y is constant on the time scale of changes in the dot polarization, we examine the fixed points of the resulting quasi-one-dimensional dynamical system (2). This regime can be straightforwardly analyzed by mapping out the x nullclines, defined by the zero-growth condition $\dot{x}=0$. Figure 4 shows the stable (white) and unstable (red, dark) x nullclines, superimposed on the velocity field map of the full system (arrows indicate direction of the polarization velocity, and the color scale indicates its magnitude).

Pictorially, oscillations occur through fast horizontal motion (i.e., rapid changes of the dot polarization x), followed by slow drift along the stable x nullclines [blue looplike trajectory in Fig. 4(a)]. The slow drift accounts for nuclear spin diffusion from the DQD to the barrier. Corresponding time traces are shown in Fig. 4(b). In a realistic model, current depends on the polarization values in the two dots in a complicated way. Here we demonstrate the time dependence of current by plotting a related quantity, the average hyperfine transition rate, $\bar{W}^{\rm hf} = \frac{1}{2}(W_+^{\rm hf} + W_-^{\rm hf})$. Similar to the current measured in experiments, this quantity displays highly nonsinusoidal behavior. The oscillation period is dominated by the length of the excursions along the nullclines. As a result, the period grows with the oscillation amplitude, also consistent with experiment [8].

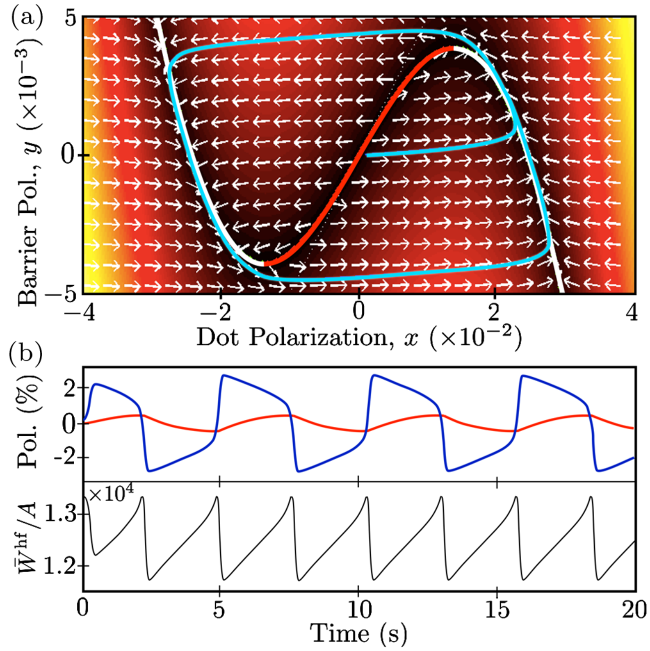

FIG. 4 (color online). Polarization dynamics in the oscillatory regime. (a) Velocity field, with arrows indicating the direction of the flow defined by Eqs. (2) and (3). Bright (dark) background color indicates fast (slow) velocity. The white and red curves show the stable and unstable x nullclines, respectively. The limit cycle trajectory shown in blue arises from a two stage process: alternating fast DNP buildup between nullclines and slow drift along the nullclines due to nuclear spin diffusion (see text). (b) Time traces of the limit cycle trajectory, from numerical solution of Eqs. (2) and (3). Upper panel: Dot polarization x (blue, dark) and barrier polarization y (red, light). Lower panel: Average hyperfine transition rate, approximately demonstrating the highly nonsinusoidal oscillatory time dependence of current. Parameter values: $\varepsilon/A = -0.0025$, $\gamma/A = 0.075$, $B = 0$, $\eta = 0.2$, $N = 10^5$, $\Gamma_D/A = 10^{-12}$.

In summary, we have identified a straightforward physical mechanism which can produce stable oscillations of dynamical nuclear polarization in spin-blockaded DQDs. The mechanism relies on nuclear spin diffusion into a tunnel barrier and is active only in vertical DQDs. The dependence of spin-injection probabilities and spin diffusion times on barrier width provides a clear experimental signature of this mechanism. Persistent oscillations can serve as a new probe of nuclear spin diffusion and spin dynamics in vertical structures.

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