PHYSICAL REVIEW B **82**, 041311(R) (2010)

Phase-sensitive probes of nuclear polarization in spin-blockaded transport

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(Received 2 June 2010; published 29 July 2010)

Spin-blockaded quantum dots provide a unique setting for studying nuclear-spin dynamics in a nanoscale system. Despite recent experimental progress, observing phase-sensitive phenomena in nuclear spin dynamics remains challenging. Here we point out that such a possibility opens up in the regime where hyperfine exchange directly competes with a purely electronic spin-flip mechanism such as the spin-orbital interaction. Interference between the two spin-flip processes, resulting from long-lived coherence of the nuclear-spin bath, modulates the electron-spin-flip rate, making it sensitive to the transverse component of nuclear polarization. In a system repeatedly swept through a singlet-triplet avoided crossing, nuclear precession is manifested in oscillations and sign reversal of the nuclear-spin pumping rate as a function of the waiting time between sweeps. This constitutes a purely electrical method for the detection of coherent nuclear-spin dynamics.

DOI: 10.1103/PhysRevB.82.041311 PACS number(s): 73.63.Kv, 72.25.Pn, 76.70.Fz

Due to their long coherence times, nuclear spins offer unique opportunities to study quantum dynamics. While conventional techniques using rf fields probe macroscopic groups of spins, currently there is wide interest in the local dynamics of nanoscale groups of spins. In particular, semiconducting quantum dots have emerged as a platform for investigating the intriguing quantum many-body dynamics of coupled electron and nuclear spins.[1–8] Many experimental achievements in this field rely on the phenomenon of spin blockade, in which electrons must flip their spins in order to pass through the system in accordance with the Pauli exclusion principle.[9] Such spin-blockaded transport is sensitive to the nuclear-polarization-dependent Overhauser field, which controls the energy splittings between electronic spin states. This sensitivity affords an exciting opportunity to electrically control and detect the states of nuclear spins.[10,11]

In this Rapid Communication, we propose a method which can be used to electrically probe *coherent* nuclear phenomena, such as Larmor precession or Rabi oscillations, which are not easily probed by the Overhauser effect alone. We identify a regime where transport is sensitive to all three vector components of the local nuclear polarization, due to coherent interplay between hyperfine coupling and an electron-only spin coupling such as the spin-orbit interaction.[12,13] As we show, in this regime a variety of phenomena which directly reveal coherent nuclear dynamics can be observed. Recently observed oscillations of the nuclear pumping rate controlled by nuclear Larmor precession[14] indicate that this unexplored regime is now within experimental reach.

Here we investigate these phenomena in a two-electron double quantum dot in a uniform magnetic field. The two-electron singlet and $m=\pm 1$ triplet states are coupled via the difference of hyperfine fields due to transverse nuclear polarization in the two dots, $\Delta\mathbf{B}_{\mathrm{nuc},\perp}$.[10,11,15] Without an additional coupling, the electron-spin-flip rate depends only on $|\Delta\mathbf{B}_{\mathrm{nuc},\perp}|$ and not on its orientation in the XY plane.[15]

The situation becomes considerably more interesting when singlet-triplet transitions can occur due to either the spin-orbit or the hyperfine interaction (see Fig. 1). In this case, we find that the probability of electron-spin flip depends on *the angle* θ of the nuclear polarization measured in the XY plane relative to a fixed axis determined by the spin-orbit interaction. Such a dependence makes electron transport sensitive to *the phase* of nuclear-spin precession. Below, we analyze this phenomenon for a Landau-Zener-type process, occurring when the electronic system is swept through a singlet-triplet level crossing, as employed, e.g., in Refs. 14 and 16 and depicted in Fig. 1(b). To make contact with experiment, we focus, in particular, on resulting signatures in nuclear-spin polarization. Strikingly, we find robust oscillations in the nuclear-spin pumping rate which result from nuclear precession and *survive even after averaging over the distribution of random initial nuclear-spin states.* Note that other types of electron-only coupling such as Zeeman coupling to the nonuniform field of a micromagnet[17] can produce analogous effects.

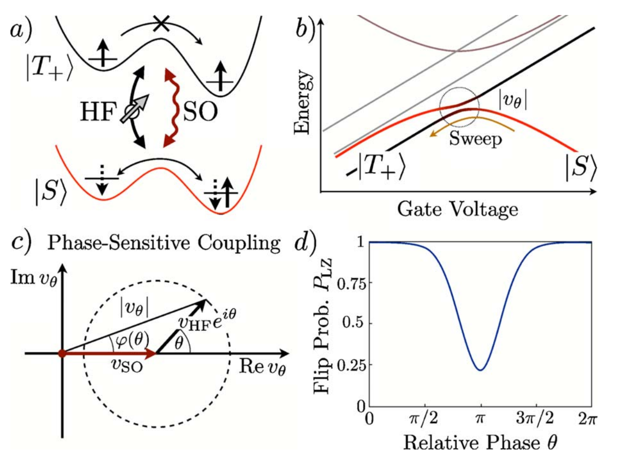

FIG. 1. (Color online) Coherent interplay of hyperfine and spin-orbit-mediated transitions. (a) Interdot tunneling in the triplet state is suppressed by Pauli exclusion but can be mediated by the hyperfine and spin-orbit interactions which do not conserve electron spin. (b) When exchange energy compensates Zeeman energy, the singlet and the triplet levels $|S\rangle$ and $|T_+\rangle$ exhibit an avoided crossing with splitting $|v_\theta|=|v_{\mathrm{SO}}+v_{\mathrm{HF}}e^{i\theta}|$. (c) Behavior near the S-T_+ crossing, controlled by $|v_\theta|$, is sensitive to the relative phase θ between spin-orbit and hyperfine matrix elements. (d) Phase-dependent transition probability, Eq. (3), with $v_{\mathrm{SO}}=0.4\sqrt{\hbar\beta}$ and $v_{\mathrm{HF}}=0.6\sqrt{\hbar\beta}$.

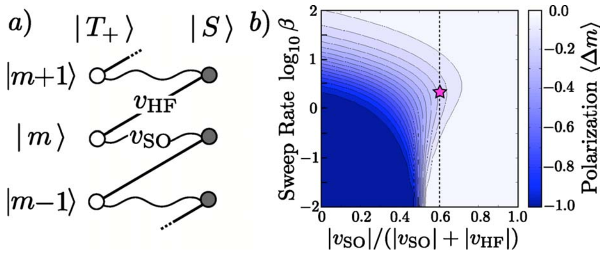

FIG. 2. (Color online) Quantum walk model of electron-nuclear-spin dynamics near the S-T_+ crossing, Eq. (4). (a) Unit cells are labeled by m, the z component of total nuclear spin. Intracell and intercell hopping correspond to spin-orbit and hyperfine transitions with matrix elements $v_{\rm SO}$ and $v_{\rm HF}$, respectively. (b) Expected change in nuclear polarization $\langle \Delta m\rangle$, Eq. (9). The star indicates the optimal sweep rate β_* along the dashed line.

The origin of the spin-angle dependence can be understood heuristically by analyzing the avoided crossing that opens near the degeneracy of triplet and singlet levels $|T_+\rangle$ and $|S\rangle$ in nonzero magnetic field, circled in Fig. 1(b). Due to the spin-orbit interaction, tunneling between dots is accompanied by a rotation of electron spin[18] which introduces a nonzero spin-flip amplitude. In addition, the hyperfine interaction between electron and nuclear spins allows electronic transitions accompanied by nuclear-spin flips, described by the effective Hamiltonian $H_{\rm HF}=A^+|S\rangle\langle T_+|+A^-|T_+\rangle\langle S|$ with $A^{\pm}=\Sigma_{i=1}^{N_{\rm nuc}} g_i I_i^{\pm}$. The magnitude and sign of each coupling constant g_i depends on the location of nucleus i (positive in dot 1 and negative in dot 2).

Taking advantage of the fact that the number of nuclei interacting with the electrons is very large, $N_{\rm nuc}\approx \mathcal{O}(10^6)$, we note that the commutator $[A^+,A^-]$ is typically smaller than A^+A^- by a factor of order $1/N_{\rm nuc}^{1/2}$. This allows us to treat A^+ as a nearly classical variable, $A^+\approx v_{\rm HF}e^{i\theta}$. Here $v_{\rm HF}$ is proportional to the magnitude of the transverse hyperfine difference field $\Delta\mathbf{B}_{{\rm nuc},\perp}$ and θ describes its orientation in the XY plane.

Together, the spin-orbit and hyperfine pieces provide a matrix element between singlet and triplet states

$$v_\theta \equiv \langle S|H|T_+\rangle = v_{\rm SO}+v_{\rm HF}e^{i\theta}. \qquad (1)$$

Evolution in the $\{|S\rangle,|T_+\rangle\}$ subspace near the level crossing is described by the 2×2 Hamiltonian

$$H_{ST_+}=\begin{pmatrix} \frac{1}{2}\Delta & v_\theta^* \\ v_\theta & -\frac{1}{2}\Delta \end{pmatrix}, \quad \Delta(t)=\varepsilon_{T_+}(t)-\varepsilon_S(t), \qquad (2)$$

where ε_{T_+} and ε_S are the energies of the diabatic $|S\rangle$ and $|T_+\rangle$ states. The level detuning $\Delta(t)$ can be controlled by electrostatic gates and/or magnetic field.

We consider the case where the system is initialized to the "(0,2)" singlet state $|S\rangle$ with both electrons residing in the right dot (large positive Δ), and then swept through the avoided crossing to the "(1,1)" charge regime with one electron on each dot [see Fig. 1(b)]. For a constant sweep rate $\beta=|d\Delta/dt|$, the electron-spin flip in such a model is interpreted as a Landau-Zener transition occurring with probability [see Fig. 1(d)]

$$P_{\rm LZ}=1-\exp(-2\pi|v_{\rm SO}+v_{\rm HF}e^{i\theta}|^2/\hbar\beta), \qquad (3)$$

where $P_{\rm LZ}$ is the probability to remain in the ground state. The explicit dependence on the phase θ of nuclear polarization, which enters through the matrix element v_θ, shows the singlet/triplet transition probability's sensitivity to the transverse nuclear polarization *vector*.

This model provides a useful heuristic for understanding electron-spin dynamics, in particular, for situations where the nuclear-spin state is characterized by a well-defined azimuthal angle θ. However, to understand the behavior with more general initial states and to account for the effects of backaction on the nuclei, we must examine the quantum many-body dynamics of coherently coupled electron and nuclear spins. In particular, we are interested in dynamical nuclear polarization (DNP) which is generated by nuclear precession around the in-plane component of electron spin polarization. By solving this problem below, we will further justify the form of Eq. (1) and will obtain the electron and nuclear-spin-flip rates.

A key new feature here is the relaxed selection rule governing nuclear pumping: due to spin-orbit coupling, electron-spin flips may occur *with* or *without* a compensating nuclear-spin flip. Although these two processes apparently lead to orthogonal final states, long-lived coherence of the nuclear-spin bath allows interference between different transition *sequences* (see Fig. 2). Moreover, a *single electron* can change the total nuclear polarization by an amount Δm that can be *larger than 1, and of either sign*.

At this point, it is convenient to map the problem onto the bipartite one-dimensional (1D) quantum walk shown in Fig. 2(a), where each unit cell is labeled by m, the z projection of the total nuclear spin $I^z=\Sigma_i I_i^z$. In doing so, $[I^z,A^{\pm}]$ remains nonzero. Intracell hopping between internal states T and S, characterized by $\Delta m=0$, describes a spin-orbit transition occurring with amplitude $v_{\rm SO}$. Intercell hopping, characterized by $\Delta m=\pm 1$, describes a hyperfine transition that occurs with the amplitude $v_{\rm HF}=\langle T,m-1|H_{\rm HF}|S,m\rangle$. Initially, we consider sweeps which are fast compared to the nuclear Larmor period, and so neglect the nuclear Zeeman energy.

Because $N_{\rm nuc}$ is large, the value of $v_{\rm HF}$ will change very little during the course of a few sweeps, and we treat it here as a constant. However, the initial values of $v_{\rm HF}$ may differ from one run to another with a statistical distribution of the form $p(v)\propto v e^{-v^2/s^2}$, where s is a constant. Additionally, because the typical values of total nuclear spin will be large, of order $\sqrt{N_{\rm nuc}}$, we may take the ladder of allowed values of m to be infinite while the total number of sweeps is not too large.

The state of the system evolves according to

$$i\hbar\dot{\psi}_m^T = v_{\rm SO}\psi_m^S + v_{\rm HF}\psi_{m+1}^S + \tfrac{1}{2}\Delta(t)\psi_m^T,$$

$$i\hbar\dot{\psi}_m^S = v_{\rm SO}\psi_m^T + v_{\rm HF}\psi_{m-1}^T - \tfrac{1}{2}\Delta(t)\psi_m^S. \qquad (4)$$

At time t_0, the system is initialized to the singlet state with polarization $m=m_0$: $\psi_m^S=\delta_{m,m_0}$, $\psi_m^T=0$. The expected change in polarization,

$$\langle \Delta m \rangle = \sum_m (m - m_0) P_m; \quad P_m = |\psi_m^S|^2 + |\psi_m^T|^2, \tag{5}$$

is determined by the probabilities $\{P_m\}$ to find the nuclear spin with z-projection m in the final state at time t_F after the sweep. For convenience we take the (nonclassical) initial nuclear state to be an eigenstate of I^z but the framework can be applied for more general states.

In the Fourier representation $|\psi_m\rangle = \frac{1}{2\pi}\oint d\theta e^{-im\theta}|\psi_\theta\rangle$, where $|\psi_m\rangle$ and $|\psi_\theta\rangle$ are two-component spinors $(\psi_m^T, \psi_m^S)^{\rm T}$, and $(\psi_\theta^T, \psi_\theta^S)^{\rm T}$, the dynamics for different θ components decouple. We obtain

$$i\hbar \frac{d}{dt}|\psi_\theta\rangle = H_{ST_+}|\psi_\theta\rangle, \tag{6}$$

where H_{ST_+} is given in Eq. (2). Here the parameter θ plays the role of the azimuthal angle of $\Delta \mathbf{B}_{\rm nuc,\perp}$ in the classical problem, Eq. (1). Indeed, a state $\psi_\theta^{S,T} \propto \delta(\theta - \theta_0)$ is a coherent state concentrated near θ_0.

To calculate $\langle \Delta m \rangle$, we make use of the relation $m|\psi_m\rangle = \frac{i}{2\pi}\oint d\theta \frac{d}{d\theta}(e^{-im\theta})|\psi_\theta\rangle$. Integrating by parts to move the derivative onto $|\psi_\theta\rangle$, we find

$$\langle \Delta m \rangle = \frac{1}{2\pi i}\oint d\theta \langle \psi_\theta | \frac{d}{d\theta}|\psi_\theta\rangle, \tag{7}$$

where without loss of generality we have taken $m_0=0$.

To evaluate the expression in Eq. (7), we must solve for the two-level dynamics of $|\psi_\theta\rangle$ under the time-dependent Hamiltonian (2). We can write the evolution operator as

$$U(\theta) = \begin{pmatrix} a_\theta & b_\theta e^{i\varphi_0(\theta)} \\ -b_\theta^* e^{-i\varphi_0(\theta)} & a_\theta^* \end{pmatrix}, \quad |a_\theta|^2 + |b_\theta|^2 = 1, \tag{8}$$

where $\varphi_0(\theta) = \arg[v_\theta]$, with v_θ defined in Eq. (1) [see also Fig. 1(c)]. With this parametrization, we have $a_\theta = a_{-\theta}$, and $b_\theta = b_{-\theta}$.

The initial state $|\psi_\theta^{(0)}\rangle = (0,1)^{\rm T}$ evolves into $|\psi_\theta\rangle = (b_\theta e^{i\varphi_0(\theta)}, a_\theta^*)^{\rm T}$. Because a_θ and b_θ are *even* functions of θ, the only term that contributes to $\langle \Delta m \rangle$ is that generated when the derivative acts on $e^{i\varphi_0(\theta)}$ in Eq. (7), giving

$$\langle \Delta m \rangle = -\frac{1}{2\pi}\oint d\theta \left(\frac{d\varphi_0}{d\theta}\right)|b_\theta|^2, \tag{9}$$

where $|b_\theta|^2$ is the singlet-triplet transition probability in the θ channel. Comparing this result to that for the net electron-spin-flip probability, given by $P_{\rm el} = \frac{1}{2\pi}\oint |b_\theta|^2 d\theta$, we note that, unlike the situation where only the hyperfine interaction is present, here there is no simple relationship between the electron and nuclear-spin-flip rates.

The analysis leading to Eq. (9) is valid for *arbitrary* time dependence $\Delta(t)$, and, in particular, can even describe cases involving multiple S-T crossings. Remarkably, for very slow sweeps the change in polarization becomes sharply quantized. Indeed, since $|b_\theta|^2 \to 1$ in the adiabatic limit, the integral in Eq. (9) is equal to a *winding number*: $\langle \Delta m \rangle = -\frac{1}{2\pi}\oint d\varphi_0$. Thus $\langle \Delta m \rangle = -1$ or 0 if v_θ does or does not wind around the origin as θ traverses the Brillouin zone, depending on the relative magnitudes of $v_{\rm HF}$ and $v_{\rm SO}$.[19] The sign is negative because electron transitions from S to T_+ flip nuclear spins from up to down.

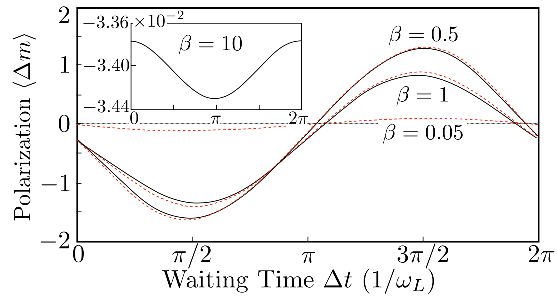

FIG. 3. (Color online) Polarization due to a round trip through the S-T_+ crossing with $v_{\rm HF}=0.6$, $v_{\rm SO}=0.4$, $\Delta_0=50$, and precession $\Delta\theta = \omega_L \Delta t$ between forward and return sweeps, Eq. (13). Here ω_L is the nuclear Larmor frequency and Δt is the waiting time. Dashed lines show $\langle \Delta m \rangle$ calculated using $\varphi_0(\theta)$ and the dynamical phase, Eq. (14), neglecting Stokes phase. The oscillation phase shifts by $\pi/2$ for faster sweeps where $\frac{d\varphi_{\rm ad}}{d\theta}$ is small (see inset). Units are chosen with $\hbar=1$.

The quantity $\langle \Delta m \rangle$ exhibits interesting dependence on the sweep speed, which can be analyzed most straightforwardly for linear sweeps $\Delta(t) = -\beta t$, when $|b_\theta|^2$ is given by the Landau-Zener formula (3). Due to the absence of dynamics for very fast sweeps, and since $\langle \Delta m \rangle = 0$ for very slow sweeps in the region $|v_{\rm SO}| > |v_{\rm HF}|$, the polarization efficiency is a *nonmonotonic* function of sweep rate [see Fig. 2(b)]. In the limit of weak hyperfine interaction, $|v_{\rm HF}| \ll |v_{\rm SO}|$, expanding the exponential in Eq. (3), we find

$$|\langle \Delta m \rangle| \approx (v_{\rm HF}/v_{\rm SO})^2 \lambda e^{-\lambda}, \quad \lambda = 2\pi v_{\rm SO}^2/\hbar\beta. \tag{10}$$

This expression attains its maximum value $|\langle \Delta m \rangle|_{\max} = v_{\rm HF}^2/(e v_{\rm SO}^2)$ at the optimal sweep rate $\beta_* = 2\pi v_{\rm SO}^2/\hbar$.

Next we consider a generalization to multiple sweeps and show that dynamical polarization is sensitive to nuclear Larmor precession between sweeps. We focus on a sequence of two identical gate sweeps, where each individual sweep is short on the scale of the nuclear Larmor time, but with a waiting time Δt between sweeps long enough to allow nuclear-spin precession. Individual sweeps may pass through the S-T crossing one or more times. Assuming that only one nuclear species contributes to the hyperfine field, the change in nuclear polarization due to the two sweeps is given by

$$\langle \Delta m \rangle = \oint \frac{d\theta}{2\pi i}\langle \mathcal{U}^{-1}\partial_\theta \mathcal{U}\rangle, \quad \mathcal{U} = U(\theta + \Delta\theta) W U(\theta), \tag{11}$$

where $U(\theta)$ is the evolution matrix for a single sweep, given by Eq. (8), W is the electronic evolution operator for the waiting interval Δt, the expectation value is taken over the state $|\psi_\theta^{(0)}\rangle = (0,1)^{\rm T}$, and $\Delta\theta = \omega_L \Delta t$ is the precession angle with ω_L the nuclear Larmor frequency. Due to the periodicity of $U(\theta)$ in θ, Eq. (8), $\langle \Delta m \rangle$ exhibits periodic oscillations in $\Delta\theta$ which are a direct manifestation of coherent nuclear polarization dynamics.

The analysis is simple when the detuning $|\Delta|$ is very large in the interval between sweeps. Then $W \approx \exp(i\chi\sigma_3)$, where the phase χ is large and very sensitive to any fluctuations in the waiting time Δt or other parameters of the system. The resulting randomness of χ completely suppresses coherence in the electronic state between sweeps while nuclei remain coherent. For demonstration, below we choose the particular protocol $\Delta(t)=\beta(t_n-t)$, for $|t-t_n|<\tau$, and $\Delta=\infty$ otherwise, where $n=1,2$, and $\tau\ll\Delta t=t_2-t_1$ with t_1 and t_2 the times of passage through the avoided crossing.

For this experimentally relevant case, it is helpful to think of the matrix products defining $\mathcal{U}$ and $\mathcal{U}^{-1}$ in Eq. (11) as sums over amplitudes associated with all possible histories of S-T transitions. Fast electron decoherence between sweeps suppresses the contribution of terms arising from nonidentical histories in $\mathcal{U}$ and $\mathcal{U}^{-1}$, leaving behind a sum of classical *probabilities* of all trajectories. Starting with the singlet initial state $(0,1)^{\mathrm{T}}$, and summing over all final states, we find

$$\langle\Delta m\rangle = \left[-(1,1)\partial_\eta \oint \frac{d\theta}{2\pi} M(\theta+\Delta\theta)M(\theta)(0,1)^{\mathrm{T}}\right]_{\eta=0},$$

$$M(\theta) \equiv \begin{pmatrix} 1-p(\theta) & p(\theta)e^{\eta(d\varphi/d\theta)} \\ p(\theta)e^{-\eta(d\varphi/d\theta)} & 1-p(\theta) \end{pmatrix}, \qquad (12)$$

where η is an auxiliary variable introduced for bookkeeping and $p(\theta)=|b_\theta|^2$ is the transition probability in the θ channel for a single sweep. Here the phase $\varphi(\theta)$ is the sum of the geometric part $\varphi_0(\theta)=\arg[v_\theta]$, the Stokes phase $\varphi_s(\theta)$, see, e.g., Ref. 20, and the dynamical phase $\varphi_{\mathrm{ad}}(\theta)=\frac{1}{\hbar}\int_{-\tau}^{\tau}\times E(\beta t,\theta)dt$ associated with adiabatic evolution on a single branch of the two-level spectrum of Hamiltonian H_{ST_+}, Eq. (2), with $E(\Delta,\theta)=-\sqrt{(\Delta/2)^2+|v_\theta|^2}$. Equation (12) then gives

$$\langle\Delta m\rangle = -\oint \frac{d\theta}{2\pi}\left\{\frac{d\varphi}{d\theta}p(\theta)+\frac{d\varphi'}{d\theta}[1-2p(\theta)]p'(\theta)\right\}, \qquad (13)$$

where $p'(\theta)=p(\theta+\Delta\theta)$, $\varphi'(\theta)=\varphi(\theta+\Delta\theta)$.

The two terms in Eq. (13) count the contributions from the two sweeps. The factor $1-2p(\theta)$ accounts for the fact that a transition from S to T on the first sweep reverses the direction of nuclear-spin flips in the second sweep. Oscillations and sign reversal of $\langle\Delta m\rangle$ as a function of the precession angle $\Delta\theta$ are clearly visible in Fig. 3, where we have used direct numerical integration to find the transition amplitudes $b(\theta)$ needed to evaluate the expression in Eq. (13).

The dynamical phase contributes to $\langle\Delta m\rangle$ through

$$\frac{d\varphi_{\mathrm{ad}}}{d\theta} = -\int_{-\tau}^{\tau}\frac{dt}{\hbar}\frac{\partial E(\beta t,\theta)}{\partial\theta} = \frac{4v_{\mathrm{SO}}v_{\mathrm{HF}}}{\hbar\beta}\sin\theta\,\sinh^{-1}\frac{|\Delta_0|}{2|v_\theta|}, \qquad (14)$$

where $d\varphi_{\mathrm{ad}}/d\theta$ grows logarithmically with the range of detunings $2\Delta_0$ spanned by the sweep. Note that $\partial E/\partial\theta$ is the group velocity in the Bloch band corresponding to the 1D quantum walk, Fig. 2(a).

This contribution describes the DNP buildup at times long after the level crossing. The in-plane component of electron spin maintains a nonzero average value due to the presence of the spin-orbit interaction. While this remnant electron spin polarization can be small, its influence on nuclear polarization accumulates over a long time, as indicated by the $1/\beta$ dependence. As a result, this contribution to DNP typically dominates over that of the geometric phase $\varphi_0(\theta)$, see Fig. 3.

Equation (11) can be readily extended to arbitrary series of nonidentical sweeps. If we take into account classical noise in the detuning $\Delta(t)$ during each sweep, the primary effect is to flatten the function $p(\theta)$, causing it to saturate toward the value 1/2 for all θ. Because the qualitative form of $p(\theta)$ is preserved, however, oscillations resulting from the geometric phase contribution $d\varphi_0/d\theta$ survive under quite general circumstances.

Resonant effects at the nuclear Larmor frequency also can be seen directly in the time dependence of electron transport properties. As the transverse polarization precesses in time, the matrix element v_θ traces out the dashed circle shown in Fig. 1(c), leading to a modulation of $|v_\theta|$ at the nuclear Larmor frequency. This effect can be detected as an ac modulation of conductance (current) in the system or as a correlation between the electron-spin-flip probabilities on successive sweeps through the avoided crossing. Such measurements would constitute a purely electrical detection of nuclear-spin dynamics.

In summary, we have shown that the coherent evolution of nuclear spins in quantum dots can be observed through oscillations and sign reversal of the nuclear-spin pumping rate, which occur due to the coherent interplay between hyperfine and spin-orbit couplings. Recent observations indicate that this interesting regime is now within experimental reach, opening a variety of possibilities to explore many-body spin dynamics in the solid state.

We thank H. Bluhm, H.-A. Engel, S. Foletti, M. B. Hastings, J. J. Krich, E. I. Rashba, and A. Yacoby for helpful discussions, and acknowledge support from W. M. Keck Foundation Center for Extreme Quantum Information Theory and NSF under Grants No. DMR-0906475 and No. DMR-0757145.

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