Coherent oscillations of current due to nuclear spins

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We propose a mechanism for very slow coherent oscillations of current and nuclear spins in a quantum dot system, that may qualitatively explain some recent experimental observations. We concentrate on an experimentally relevant double dot setup where hyperfine interaction lifts the spin blockade. We study the dependence of the magnitude and period of the oscillations on magnetic field and anisotropy.

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There are significant experimental and theoretical efforts aimed to utilize and manipulate the electron spin in the context of electronic transport; these are commonly referred to as spintronics.^{1,2} Some of them involve nuclear spins as well. Many efforts concern GaAs semiconductor structures where the hyperfine interaction between electron and nuclear spin is relatively strong.³ Furthermore, the nuclear spin relaxation times are much longer than the time scales related to electron dynamics.^{4,5} This time scale separation has facilitated experiments where a quasistationary polarization of the nuclear system was achieved and its effect on the electronic transport was observed.^{6–8} Recent experiments implement interesting ways of controlling the interaction of electron and nuclear spin,^{9,10} whereby the coherent oscillations between “up” and “down” polarized nuclear systems have been observed.¹¹

Transport experiments with quantum dots allow for a detailed study and control of the quantum dot energy spectrum, both in the regime of linear transport and in the nonlinear regime of excitation spectroscopy.^{12,13} They reveal interesting regimes where unusual mechanisms of electron transport are the dominant ones. For instance, in the Coulomb blockade regime the direct charging of the quantum dot is forbidden by energy conservation and the residual current is due to cotunneling.^{14,15} The effect of Coulomb blockade on the nuclear spin-relaxation time was considered in Ref. 16. Another regime is the so-called *spin blockade* where the direct electron transfers are blocked by virtue of spin conservation.¹⁷ Spin blockade may be achieved in various ways, e.g., with spin polarized leads.¹⁸ Recent experiment realizes the spin blockade in a double dot system, where the absence of transitions between spin-singlet and spin-triplet states in the dots explains the current rectification observed.¹⁹ Any spin-flip mechanism facilitates these transitions, giving rise to a small residual current.

The same group has recently reported²⁰ an unexpected and unusual result. They have observed coherent oscillations of the residual current in this regime with a period in the range of seconds. This extremely long time scale together with the fact that the oscillation period and amplitude can be modified by resonant excitation of the nuclear spins strongly suggests that the origin of the oscillations may be traced to the hyperfine interaction.²⁰

In this paper we propose a concrete mechanism for these oscillations that can at least qualitatively explain the observation made in Ref. 20. The effect comes about from the dynamics of nuclear spins driven by hyperfine interaction

with electron spin. The configuration of the nuclear spins can be presented with two effective magnetic fields acting on the electron spin in the two dots. The difference of these fields lifts the spin blockade thereby affecting the average current and electron spin. These fields are quasistationary at the scale of successive electron transfers. They precess around the external magnetic field (z axis) with frequency 10^7 Hz, albeit this precession does not manifest themselves in current oscillations. The oscillations result from slow *nutations* of the fields. These nutations arise from small deflection of electron spin from the z axis, the deflection being induced by the fields.

The dynamics of the nuclear-spin fields appears to be far from chaotic so that the period and magnitude of the oscillations strongly depend on initial conditions of the nuclear-spin system. Therefore the comparison with experiment may be only qualitative. In reality, the relaxation of nuclear spin would lead to stabilization of the oscillations with a certain amplitude and period. However, such stabilizing mechanisms would manifest itself at time scale much longer than the period. This is why we do not consider them in the present model.

For details of the setup we refer the reader to Ref. 19. In the regime of interest, the double dot can be in three distinct charge configurations. A charge configuration is characterized by (N_L, N_R) , $N_L(N_R)$ being the number of electrons in the left (right) dot. Transitions from (0,1) to (1,1) and from (0,2) to (0,1) are relatively fast involving electron tunneling either from the left or to the right lead with the rates $\Gamma_{L,R}$. The bottleneck of the transport cycle are transitions between (1,1) and (0,2). In the (0,2) configuration both electrons share the same orbital state, this makes it a nondegenerate spin singlet. In contrast to this, the (1,1) configuration comprises four possible states, grouped into spin singlet and spin triplet. The transitions between the (1,1) singlet and (0,2) occur with the rate Γ_i that is determined by the tunneling amplitude between the dots and a relevant mechanism of inelastic scattering, e.g., phonons. These transitions do not require any spin flip. The transitions between the (1,1) triplet states and (0,2) singlet are forbidden by spin conservation: this causes the spin blockade.

The hyperfine interaction with nuclei induces mixing of the singlet and triplet states in the (1, 1) configuration. The part of the total Hamiltonian which contains the electron- and nuclear-spin operators reads

$$H_s = \hat{\mathbf{K}}_L \cdot \hat{\mathbf{S}}_L + \hat{\mathbf{K}}_R \cdot \hat{\mathbf{S}}_R + E_Z \cdot (\hat{S}_L^z + \hat{S}_R^z) + \Delta_{ST}(\hat{\mathbf{S}}_L + \hat{\mathbf{S}}_R)^2/2. \quad (1)$$

Here $\hat{\mathbf{S}}_{L,R}$ are operators of electron spin in each dot.²⁶ The effect of nuclear spins is combined into two effective fields $\hat{\mathbf{K}}_{L,R}$. In each dot

$$\hat{\mathbf{K}}_{L,R} = \hbar \sum_k \gamma_{L,R;k} \hat{\mathbf{I}}_k; \quad \gamma_{L,R;k} = \hbar^{-1} A |\psi_{L,R}(\mathbf{R}_k)|^2 \quad (2)$$

where $\hat{\mathbf{I}}_k$ being operators of nuclear spin at $\mathbf{R}_k$, the summation goes over all nuclei and the precession frequencies γ_k are set by an envelop of electron wave function and hyperfine constant A . The third term in Eq. (1) represents the Zeeman splitting E_Z in the external magnetic field while the fourth term represents the exchange splitting between the singlet and triplet, whose magnitude is determined by Δ_{ST} . We adopt a semiclassical approximation of the effective fields $\mathbf{K}_{L,R}$ replacing them by classical variables.^{21–23} This approximation is justified by the large number of nuclei in the dots, $N_{QD} \gg 1$. The third and fourth terms in Eq. (1) include the full spin only and therefore split the states onto singlet and three Zeeman-split triplet components. The mixing of these states is proportional to the difference of two effective fields $\mathbf{K}_A \equiv \mathbf{K}_L - \mathbf{K}_R$.

The mixing thus lifts spin blockade. We assume that this mixing is the only mechanism of the residual current. This assumption is not crucial since alternative mechanisms, those include co-tunneling and nonnuclear spin flips, would only produce an extra dc current background for nuclear-induced current oscillations.

We solve the problem in two steps. First, we solve for the density matrix of electron states assuming stationary $\mathbf{K}_{L,R}$. The output of the calculation are the average current and the average electron spin $\langle \mathbf{S}_{L,R} \rangle$ in terms of $\mathbf{K}_{L,R}$. Second, we use this output to derive equations for the dynamics of $\mathbf{K}_{L,R}$ and subsequently analyze this dynamics. This approach relies on the time-scale separation: the fields $\mathbf{K}_{L,R}$ should not change at the time scale of successive electron transfers. The transfer rate can be estimated as $(E_Z \simeq \Delta_{ST})(K_A/\Delta_{ST})^2 \Gamma_i$. The small factor (K_A/Δ_{ST}) is the ratio of the mixing and energy difference between singlet and triplet states and quantifies the suppression of the current in the spin blockade regime. The fastest nuclear-spin motion that changes $\mathbf{K}$ is the precession around external magnetic field with frequency $\omega_{NMR} \simeq 10^7$ Hz. This results in the condition $\Gamma_i \gg (\Delta_{ST}/K_A)^2 \omega_{NMR}$ for the validity of our approach. As we see below, the current oscillations are much slower with a typical period of the order of $\gamma^{-1}(\Delta_{ST}/K_A)$, where $\gamma = (\gamma_L + \gamma_R)/2$. The precession frequency and typical magnitude of effective field can be estimated²³ as $\gamma \simeq E_n/N_{QD}$, $K \simeq E_n/\sqrt{N_{QD}}$, where $E_n \simeq 0.135$ meV in GaAs and $N_{QD} \simeq 10^6$ for the quantum dots in question. The exchange splitting $\Delta_{ST} \propto 10^{-5} - 10^{-4}$ eV as estimated in Ref. 20. This gives $\gamma^{-1}(\Delta_{ST}/K_A) \simeq 0.01 - 0.1$ sec.

To make the first step, we describe the evolution of the electron system with Bloch equations for the density-matrix approach. There are seven quantum states involved in the

transport [$|s\rangle$ for the singlet in the (0,2) configuration, $|+\rangle$ and $|-\rangle$ for the two doublet components in the (1,0) configuration, $|0\rangle$ for the singlet, and $|1\rangle$, $|2\rangle$, $|3\rangle$ for triplet states in the (1,1) configuration, those correspond to $S_z = 1, 0, -1$, respectively] so that the full density matrix comprises of 49 elements. However, we can disregard most of the nondiagonal elements, i.e., the ones between states corresponding to different charge configurations, of the matrix except those between four states of the (1,1) configuration. So we end up with 19 equations only, 7 “diagonal” and 12 “off-diagonal.” Here we present three; this suffices to illustrate the overall structure:

$$\frac{d\rho_{11}}{dt} = \frac{\Gamma_L}{2} \rho_{++} + \frac{1}{2i} (K_A^+ \rho_{10} - \text{c.c.}) \quad (3a)$$

$$\begin{aligned} \frac{d\rho_{00}}{dt} = & -\Gamma_i \rho_{00} + \frac{\Gamma_L}{4} (\rho_{++} + \rho_{--}) + \frac{1}{2i} (-K_A^+ \rho_{10} + K_A^z \rho_{20} \\ & + K_A^- \rho_{30} - \text{c.c.}) \end{aligned} \quad (3b)$$

$$\begin{aligned} \frac{d\rho_{10}}{dt} = & -i \left((\Delta_{ST} + E_Z) - i \frac{\Gamma_i}{2} \right) \rho_{10} + \frac{1}{2} [-i K_A^- (\rho_{11} - \rho_{00}) \\ & + i K_A^z \rho_{12} + i K_A^+ \rho_{13}], \end{aligned} \quad (3c)$$

where $K_A^\pm = (K_A^x \pm i K_A^y)/\sqrt{2}$. Note that the inelastic rate Γ_i does not appear in Eq. (3a); the same is true for the other triplets, but it is present in Eq. (3b) for the singlet since only it is directly coupled to ρ_{ss} . Also, Eqs. (3a) and (3b) are coupled via the off-diagonal matrix element and the coupling is determined by $\mathbf{K}_A$, which clearly shows that if the two effective fields are equal, i.e., $\mathbf{K}_A = 0$, the transport is blocked. The average electron spins in each dot and the current can be readily obtained from the stationary solution $\hat{\rho}^{st}$ of Eq. (3): $\langle \mathbf{S}_{L,R} \rangle = \text{Tr}\{\hat{\rho}^{st} \hat{\mathbf{S}}_{L,R}\}$, $I = e \Gamma_R \rho_{ss}^{st}$. We do not need to present the cumbersome full solutions for the average spin here. Instead, we assume $\Gamma_{in} \ll \Gamma_{L,R}, \Delta_{ST}/\hbar$ and note that in the zeroth order in K_A/Δ_{ST} the average spin is in the z direction and $\langle x_B \rangle \equiv E_Z/\Delta_{ST}$

$$\langle S_{L,R}^z \rangle = -\frac{2E_Z}{\Delta_{ST}} S; \quad S^{-1} \equiv 2(1 + x_B^2) + \left(\frac{|K_A^+|^2}{K_A^z} \right). \quad (4)$$

With the same accuracy the current reads

$$\frac{I}{e} = \left(\frac{|K_A^+|^2}{\Delta_{ST}} \right) 4S \Gamma_i. \quad (5)$$

In addition, we need the deflections of $\langle \mathbf{S}_{L,R} \rangle$ from the z direction. They arise in the next order in K_A/Δ_{ST} and are antiparallel in opposite dots:

$$\begin{pmatrix} \langle S_{L,R}^x \rangle \\ \langle S_{L,R}^y \rangle \\ \langle S_{L,R}^z \rangle \end{pmatrix} = \pm \frac{S}{2\Delta_{ST}} \begin{pmatrix} K_A^x \\ K_A^y \\ \frac{|K_A^+|^2}{K_A^z} \end{pmatrix}. \quad (6)$$

Now we perform the second step and describe the dynamics of the effective fields. To simplify, we will assume the same electron precession frequencies for all nuclei in each

dot: $\gamma_{L,R;k} = \gamma_{L,R}$. The advantage of this model is a closed system of equations for the effective fields:

$$\frac{d}{dt} \mathbf{K}_\alpha = \gamma_\alpha \langle \mathbf{S}_\alpha \rangle \times \mathbf{K}_\alpha + \gamma_{\text{GaAs}} \mathbf{B} \times \mathbf{K}_\alpha \quad (7)$$

($\alpha=L,R$) that describes precession of these fields, their moduli $|\mathbf{K}_\alpha|$ being a constant of motion. The main precession is around external magnetic field with the frequency $\gamma_{\text{GaAs}}|B|$. However, this precession is irrelevant since it does not change the K_α^z and the angle ϕ between the projections of $\mathbf{K}_L$ and $\mathbf{K}_R$ onto the xy plane. These three slow variables actually determine the average spin and current, and the evolution equations for those,

$$\frac{d}{dt} K_\alpha^z = \gamma_\alpha (\langle S_\alpha^x \rangle K_\alpha^x - \langle S_\alpha^y \rangle K_\alpha^y) \quad (8)$$

$$\frac{d}{dt} \phi = \gamma_L \langle S_L^z \rangle - \gamma_R \langle S_R^z \rangle,$$

do not contain $\gamma_{\text{GaAs}} B \gg \gamma_{L,R} \langle S_{L,R} \rangle$. These three equations have an extra integral of motion, $K_S^z \equiv (K_L^z \gamma_R + K_R^z \gamma_L) / 2\gamma$. Moreover, the equation for two remaining variables appears to be of a Hamiltonian type. In dimensionless variables $k \equiv K_A^z / K_L$, $\tau = t \gamma K_L / \Delta_{\text{ST}}$ the equations read

$$\frac{d}{d\tau} \begin{pmatrix} k \\ \phi \end{pmatrix} = \frac{k^2}{(b - \cos \phi) Y} \begin{pmatrix} \frac{\partial}{\partial \phi} \\ -\frac{\partial}{\partial k} \end{pmatrix} \mathcal{L}, \quad (9)$$

where

$$\mathcal{L}(k, \phi) = X(k) + Y(k) \cos \phi, \quad (10)$$

$$X(k) = -\frac{1 + k_R^2 - 2(k_S^z)^2}{2} k + 2\epsilon x_B k^2 + \frac{k^3}{4},$$

$$Y(k) = k \sqrt{[1 - (k_S^z + k/2)^2][k_R^2 - (k_S^z - k/2)^2]},$$

$$b(k) = \frac{(\frac{7}{4} + 2x_B^2)k^3 + \frac{k}{2}[1 + k_R^2 - 2(k_S^z)^2]}{Y(k)},$$

and we introduced the notations $k_R, k_S^z \equiv K_R / K_L, K_S^z / K_L$ and $\epsilon = (\gamma_L - \gamma_R) \Delta_{\text{ST}} / 2\gamma K_L$. We also assume here that the asymmetry of electron precession frequencies is small, $\delta\gamma \equiv |\gamma_L - \gamma_R| \ll \gamma_{L,R}$. One would expect this for the experiment in question since the two dots are nominally identical.

The $\mathcal{L}$ is evidently yet another constant of motion that depends on the initial condition of the nuclear system. The solution of $\mathcal{L} = L$, if it exists, determines a closed orbit in the (k, ϕ) phase space and the system experiences periodic motion along this orbit. This motion manifests itself in the current oscillations by virtue of Eq. (5). The period and magnitude of the oscillations do depend on the initial conditions L, k_R, k_S^z . If L is close to 0, the period even diverges. There is, however, some regular dependence on the asymmetry ϵ and the external magnetic field that enters through parameter x_B , see Fig. 1. This dependence can be summarized as follows. The period T depends on asymmetry. If $\epsilon x_B \lesssim 1$, T

FIG. 1. The period as a function the integral of motion L for various values of x_B . The asymmetry is $\epsilon=0.1$, $k_R=0.9$, and $k_S^z=0.0$. The inset shows the period as a function of x_B for fixed values of L .

$\approx \gamma^{-1} \Delta_{\text{ST}} / K_L$ for $x_B \lesssim 1$ and $T \approx \gamma^{-1} E_Z^2 / \Delta_{\text{ST}} K_L$ for $x_B \gg 1$. In the opposite case of relatively large asymmetry $\epsilon x_B \gg 1$, $T \approx (x_B \delta\gamma)^{-1}$ for $x_B \lesssim 1$ and $T \approx x_B (\delta\gamma)^{-1}$ for $x_B \gg 1$. Also, the amplitude of the oscillations relative to the average current is of the order of 1 for $x_B \lesssim 1$ and decreases as x_B^{-2} for $x_B \gg 1$.

The initial values of $\mathbf{K}_\alpha$ that determine the actual magnitude and period of the oscillations are distributed according to Gaussian statistics²³ with average squares corresponding to average squares of total nuclear spins in the dot. In Fig. 2 the current, see Eq. (5), is plotted for various initial conditions but fixed $\epsilon=0.1$ and $x_B=1.6$. We note apparent anharmonicity of the oscillations; this feature has been stressed in Ref. 20.

One might think that the periodic oscillations we obtained in our approach is an artefact of an oversimplified model for nuclear dynamics in use. Recent work emphasizes the importance of the fact that precession frequency γ_k varies from nucleus to nucleus.²⁴ This issue can be addressed within the semiclassical approach used here. To implement such an approach numerically, we separate the nuclear-spin system into

FIG. 2. The current shown as a function of time for $\epsilon=0.1$, $x_B=1.6$, but for different initial conditions, i.e., different L . The inset shows current for $L=-0.14$ in the case of $N_b=1$ and 25, where N_b is the number of blocks. Note that the long period oscillations are still present for $N_b \gg 1$.

$N_b \gg 1$ blocks where the γ_k 's are the same within each block but differ from block to block. The number of nuclear spins in each block remains large so that the nuclear dynamics can be described in terms of effective fields $\mathbf{K}_{L,R}^{(b)}$. This results in $6N_b$ evolution equations similar to Eqs. (8). Intuitively, one expects such complicated dynamics to be chaotic, so that the memory about initial conditions is lost after some time $\approx \gamma^{-1}$. This would be really dreadful for the mechanism discussed, so we have performed extensive numerical simulations to check this circumstance.²⁵ To summarize the results, the dynamics is not chaotic, the memory about initial conditions persists, and the nuclear system exhibits regular oscillations, typically with several periods. To illustrate this fact, we present a typical result in the inset of Fig. 2. It shows the regular long-period motion and extra fast oscillations on the time scale of γ^{-1} .

In conclusion, we propose a mechanism whereby the transport via a double quantum dot induces slow regular nutations of the nuclear-spin system, these nutations are seen in the transport current. We model the concrete experimental situation²⁰ and our estimations of the typical frequency, anharmonic shape of the oscillation predicted, and sensitivity to magnetic field shown correspond to the observations made in Ref. 20. More research on relevant nuclear-spin-relaxation mechanisms is needed for detailed comparison with the experiment. On the other hand, the mechanism presented is sufficiently general and works in any conditions where the hyperfine interaction provides the main mechanism of spin blockade lifting.

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¹*Semiconductor Spintronics and Quantum Computation*, edited by D. D. Awschalom, D. Loss, and N. Samarth (Springer-Verlag, Berlin, 2002).

²S. A. Wolf, D. D. Awschalom, R. A. Buhrman, J. M. Daughton, S. von Molnár, M. L. Roukes, A. Y. Chtchelkanova, and D. M. Treger, *Science* **294**, 1488 (2001).

³D. Paget and V. L. Berkovits, *Optical Orientation* (North-Holland, Amsterdam, 1984).

⁴D. Paget, G. Lampel, B. Sapoval, and V. I. Safarov, *Phys. Rev. B* **15**, 5780 (1977).

⁵D. Paget, *Phys. Rev. B* **25**, 4444 (1982).

⁶G. Salis, D. T. Fuchs, J. M. Kikkawa, D. D. Awschalom, Y. Ohno, and H. Ohno, *Phys. Rev. Lett.* **86**, 2677 (2001).

⁷G. Salis, D. D. Awschalom, Y. Ohno, and H. Ohno, *Phys. Rev. B* **64**, 195304 (2001).

⁸K. R. Wald, L. P. Kouwenhoven, P. L. McEuen, N. C. van der Vaart, and C. T. Foxon, *Phys. Rev. Lett.* **73**, 1011 (1994).

⁹J. H. Smet, R. A. Deutchmann, F. Ertl, W. Wegscheider, G. Abstreiter, and K. von Klitzing, *Nature (London)* **415**, 281 (2002).

¹⁰R. K. Kawakami, Y. Kato, M. Hanson, I. Malajovich, J. M. Stephens, E. Johnston-Halperin, G. Salis, A. C. Gossard, and D. D. Awschalom, *Science* **294**, 131 (2001).

¹¹T. Machida, T. Yamazaki, K. Ikushima, and S. Komiyama, *Appl. Phys. Lett.* **82**, 409 (2003).

¹²L. P. Kouwenhoven, C. M. Marcus, P. L. McEuen, S. Tarucha, R. M. Westervelt, and N. S. Windgreen, *Mesoscopic Electron Transport* (Kluwer, Dordrecht, 1997), p. 105.

¹³L. P. Kouwenhoven, D. G. Austing, and S. Tarucha, *Rep. Prog. Phys.* **64**, 701 (2001).

¹⁴D. V. Averin and Y. V. Nazarov, *Phys. Rev. Lett.* **65**, 2446 (1990).

¹⁵S. De Franceschi, S. Sasaki, J. M. Elzerman, W. G. van der Wiel, S. Tarucha, and L. P. Kouwenhoven, *Phys. Rev. Lett.* **86**, 878 (2001).

¹⁶Y. B. Lyanda-Geller, I. L. Aleiner, and B. L. Altshuler, *Phys. Rev. Lett.* **89**, 107602 (2002).

¹⁷D. Weinman, W. Häussler, and B. Kramer, *Phys. Rev. Lett.* **74**, 984 (1995).

¹⁸M. Ciorga, M. Pioro-Ladriere, P. Zawadzki, P. Hawrylak, and A. S. Sachrajda, *Appl. Phys. Lett.* **80**, 2177 (2002).

¹⁹K. Ono, D. G. Austing, Y. Tokura, and S. Tarucha, *Science* **297**, 1313 (2002).

²⁰K. Ono and S. Tarucha, *Phys. Rev. Lett.* **92**, 256803 (2004).

²¹M. I. D'yakonov and V. Y. Kachorovskii, *Fiz. Tekh. Poluprovodn. (S.-Peterburg)* **20**, 178 (1986); [*Sov. Phys. Semicond.* **20**, 110 (1986)].

²²I. A. Merkulov, A. L. Efros, and M. Rosen, *Phys. Rev. B* **65**, 205309 (2002).

²³S. I. Erlingsson and Y. V. Nazarov, *Phys. Rev. B* **66**, 155327 (2002).

²⁴A. V. Khaetskii, D. Loss, and L. Glazman, *Phys. Rev. Lett.* **88**, 186802 (2002).

²⁵S. I. Erlingsson and Y. V. Nazarov, *Phys. Rev. B* **70**, 205327 (2004).

²⁶The spin operator are $\hat{S}_\alpha = \sum_{\eta, \gamma} \hat{\sigma}_{\eta\gamma} d_{\alpha\eta}^\dagger d_{\alpha\gamma}$ where $\hat{\sigma} = (\hat{\sigma}_x, \hat{\sigma}_y, \hat{\sigma}_z)$ are the Pauli matrices and $d_{\alpha\eta}^\dagger$ creates an electron with spin η in dot α .