Leakage-induced decoherence during single electron spin manipulation in a double quantum dot

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Coherent single electron spin oscillation in a double quantum dot system driven by a magnetic electron spin resonance field is studied theoretically using a Bloch-type rate equation approach. The oscillation frequency and relaxation time obtained using typical model parameters are consistent with experiment findings. The dominant decoherence mechanism is identified to be a leakage current through a Coulomb blockade barrier at a quantum dot during the spin manipulation. Nuclear field fluctuations which induce a long relaxation time are found to contribute only negligibly to the decoherence despite an earlier suggestion.

Because of a long decoherence time, the use of electron spins in solid-state systems, such as semiconductor nanostructures, is a promising approach for realizing a qubit, which is the basic unit in quantum computing and quantum information processing [1, 2, 3]. The manipulation and readout of a single electron spin are all essential. Solid-state implementations are expected to be most scalable and are under intensive research. Electrons realizing solid-state spin-based qubits are in general confined to semiconductor quantum dots where their numbers can be precisely controlled [4]. The weak coupling of electron spins to the rest of the solid allows long-lived spin states to be precisely controlled [3]. The weak coupling of electron spins in solid-state systems, such as semiconductor quantum dots, has been achieved experimentally [3]. For example, the coherent manipulation of two electron spins in a double quantum dot (DQD) system has been achieved experimentally [3]. More importantly, the coherent rotation of a single electron spin in a DQD system using electron spin resonance (ESR) was demonstrated very recently by Koppens et al. [8]. Clear evidence of coherent oscillation of the spin state was observed. There was also a gradual decay of the oscillation amplitude with a time constant of the order of 1 μs, which was attributed to the fluctuations of nuclear fields due to the host materials. This is consistent with the conventional belief that nuclear fields may lead to fast relaxation of electron spins [3, 10]. However, recent experiments have shown that nuclear field fluctuations can only induce the relevant spin transitions at a much longer time scale of 100 μs or more at large external magnetic fields [11] and hence cannot be responsible for the experimentally observed spin relaxation.

In this Letter, we derive a detailed theory for the ESR induced single spin oscillation and the associated spin-dependent quantum transport in a DQD system in order to explain the experiments in [8]. By deriving and numerically integrating a set of Bloch-type rate equations for the reduced density matrix elements for the DQD, we successfully obtain the coherent oscillations of a single electron spin when driven by an ESR field in close agreement with experiments. We further show that nuclear field fluctuations cannot account for the experimentally observed decay of the spin oscillations in contrast to the suggestion by Koppens et al. [8]. It is shown to be caused by a leakage current through a Coulomb barrier during spin manipulation. Approaches for enhancing the coherence will be discussed.

Model — The system consists of DQD connected to two electron reservoirs via tunneling barriers (see Fig. 1). We first define the Hamiltonian for only the DQD under coherent manipulation

\[ H_0 = \sum_{i,\sigma} E_i a_{i\sigma}^\dagger a_{i\sigma} + V_0 \sum_\sigma \left( a_{L\sigma}^\dagger a_{R\sigma} + H.c. \right) + \sum_i \left( U_i n_i^\dagger n_i + U_{LR} \sum_\sigma n_{L\sigma} n_{R\sigma} \right) + H_{mag}, \tag{1} \]

where \( i = L \) or \( R \) denotes the left or right quantum dot, while \( a_{i\sigma}^\dagger \), \( a_{i\sigma} \), and \( n_{i\sigma} \) are the creation, annihilation, and number operators, respectively, for electrons at dot \( i \) with spin \( \sigma \). The first four terms on the r.h.s. represent the electron orbital energy, interdot tunneling, and intra- and interdot Coulomb interactions, respectively. The last term describes the interactions with magnetic fields:

\[ H_{mag} = \sum_i g_i \mu_B \left( \mathbf{B}_{Ni} \cdot \mathbf{S}_i + B_{ext}^z S_i^z + \frac{B_{ext}^z}{2} \cos(\omega_c t) S_i^z \right), \]

where \( \mathbf{S}_i \) is the spin operator at dot \( i \), while \( g \) and \( \mu_B \) are the electron \( g \)-factor and the Bohr magneton, respectively. The nuclear magnetic field at dot \( i \) due to the host materials is denoted by \( \mathbf{B}_{Ni,i} \). It is known to fluctuate at a time scale of the order of 1 s, which is much longer than that of the relevant electron transport process. They are therefore taken as stationary random fields [12]. An external field \( B_{ext}^z \) is applied in the perpendicular direction to generate a Zeeman splitting \( g_i \mu_B B_{ext}^z \). Most interestingly, a spin at dot \( i \) is manipulated by the applied oscillating magnetic ESR field \( B_{ext}^z \cos(\omega_c t) \) when in resonance with the Zeeman splitting.
The full Hamiltonian for the complete device of DQD connected to external leads is \( H = H_{DQD} + H_{\text{leads}} + H_T \). The Hamiltonian of the leads is defined as \( H_{\text{leads}} = \sum_{\alpha=1}^{\alpha} E_{\alpha k\sigma} c_{\alpha k\sigma} c_{\alpha k\sigma}^\dagger \), where \( c_{\alpha k\sigma}^\dagger \) (\( c_{\alpha k\sigma} \)) is the creation (annihilation) operator of an electron with momentum \( k \) and spin \( \sigma \) in lead \( \alpha \) (\( \alpha = l, r \)). Finally, the tunneling coupling between the DQD and the leads is given by \( H_T = \sum_{\alpha=1}^{\alpha} (\Omega d_{\alpha l} c_{\alpha l\sigma} + \Omega d_{\alpha r} c_{\alpha r\sigma} + \text{H.c.}) \).

At least one electron is always kept in the right dot in the experiments [8] by applying appropriate gate voltages. A second electron is transported from lead \( l \) to \( r \) via the dots. The relevant electronic states for the DQD span a seven-dimensional Hilbert space. The basis set consists of the single-electron states \( | \uparrow_R \rangle, | \downarrow_R \rangle \), and five double-electron states, namely, the double-dot triplet states \( | 1 \rangle \equiv | \uparrow_L\uparrow_R \rangle, | 2 \rangle \equiv | \uparrow_L\downarrow_R \rangle, \) and \( | 3 \rangle \equiv | \downarrow_L\downarrow_R \rangle \), the double-dot singlet states \( \langle 4 | \equiv | \uparrow_L\downarrow_R \rangle - | \downarrow_L\uparrow_R \rangle \), and the single-dot singlet states \( | 5 \rangle \equiv | \downarrow_L\downarrow_R \rangle \). Single-dot triplet states are excluded due to their much higher orbital energy [14][13]. In this representation, \( H_{DQD} \) is rewritten as

\[
H_{DQD} = \sum_{\sigma} E_{\sigma R} | \sigma R \rangle \langle \sigma R | + \sum_{\beta=1}^{5} E_{\beta} | \beta \rangle \langle \beta | + \frac{g_{\mu B}}{\sqrt{2}} \left( (B_x^L i B_y^R + B_x^R i B_y^L) | 1 \rangle + (B_x^L - i B_y^R) | 2 \rangle + \text{H.c.} \right) \]

\[
+ \frac{g_{\mu B}}{\sqrt{2}} \left( (B_x^L - i B_y^R) | 1 \rangle + (B_x^L - i B_y^R) | 2 \rangle + \text{H.c.} \right) \]

\[
+ V_0 (| 4 \rangle \langle 5 | + | 5 \rangle \langle 4 |) + g_{\mu B} B_z^R (| 3 \rangle \langle 4 | + | 4 \rangle \langle 3 |)
+ \Omega_1 \cos(\omega_1 t) \left[ | 3 \rangle \langle 1 | + | 1 \rangle \langle 3 | \right] + \Omega_2 \cos(\omega_2 t) \left[ - | 4 \rangle \langle 1 | + | 1 \rangle \langle 4 | \right] + \text{H.c.},
\]

where \( B_{\alpha L} = \frac{1}{2} B_{\alpha L} + B_{\alpha L}^* \), \( B_{\alpha R} = \frac{1}{2} B_{\alpha R} + B_{\alpha R}^* \), and \( E_{\sigma R} = E_R + \frac{1}{2} g_{\mu B} \sigma R (B_{\alpha L}^* + B_{\alpha R}^* \right) \). We have also introduced energy levels given by \( E_{\sigma R} = E_R \pm \frac{1}{2} g_{\mu B} \sigma R (B_{\alpha L}^* + B_{\alpha R}^* \right) \), \( E_{\alpha L} = E_R \pm \frac{1}{2} g_{\mu B} E_\alpha \), and \( E_{\alpha R} = E_L + E_R + U_{LR} \), \( E_5 = 2E_R + U_R \). A critical term in the transport is the hopping or tunneling through the right dot. There can be a non-zero Coulomb energy barrier

\[
\Delta = E_5 - E_4 = U_R - U_{LR} - (E_L - E_R),
\]

for the second electron at the right dot if the intradot repulsion \( U_R \) dominates.

Coherent manipulation — To apply and detect the coherent rotation of an individual electron spin, the experiment cycles repeatedly through a manipulation stage and a combined readout and initialization stage (both stages last for 1 μs) [8]. In the combined readout and initialization stage, the right dot potential is pulsed low so that the right-dot barrier vanishes (\( \Delta \approx 0 \)). Also, the ESR field is turned off (\( \Omega_1 = \Omega_2 = B_z^* = 0 \)). This initializes the system at a spin blockade regime as will now be explained. When a electron enters the left dot from the lead carrying a random spin, the DQD system takes one of the four double-electron double-dot states \(| 1 \rangle, | 2 \rangle, | 3 \rangle, \) or \( | 4 \rangle \) with equal probability. If the system takes \( | 4 \rangle \), the electron readily carries out a spin-independent hop to the right dot as \( \Delta \approx 0 \) and turns the electrons into the single-dot state \(| 5 \rangle \). This transition is accounted for by the \( V_0 \) term in \( H_{DQD} \) in Eq. (2).

The electron then exits to the right lead and this completes the transport of one electron. Alternatively, starting at state \(| 3 \rangle \), the \( z \) component of the random nuclear fields induce a random relative phase between the spins. The system thus evolves into the near-degenerate state \(| 4 \rangle \) as described by the \( B_z^R \) term in \( H_{DQD} \). The electron can then be similarly transported. As a result, the second electron can always tunnel through the right dot for initial configurations with \( S_z = 0 \) [Figs. 1(a)-(b)]. In contrast, the other possible initial states \(| 1 \rangle \) and \(| 2 \rangle \) are split off in energy due to the application of a large external field \( B_z^\text{ext} \gg \sqrt{<B_z^2>} \) and thus are not coupled to \(| 3 \rangle \) or \(| 4 \rangle \). They are hence spin blocked states and stop further transport [Figs. 1(c)-(d)]. Therefore, starting from an arbitrary state, current flows in general until the DQD arrives stochastically at \(| 1 \rangle \) or \(| 2 \rangle \) and this completes the initialization stage.

During the whole manipulation stage which follows, the right dot potential is pulsed up to provide a Coulomb blockade with \( \Delta \gg 0 \). A burst of oscillating ESR field is applied for a period \( \tau \) just before the end of the manipulation stage (i.e., \( \Omega_1, \Omega_2 > 0 \)). A spin which is at resonance can be coherently rotated. The spin blocked states \(| 1 \rangle \) or \(| 2 \rangle \) hence coherently evolves into the non-spin-blocked state \(| 3 \rangle \) or \(| 4 \rangle \) via the \( \Omega_1 \) or \( \Omega_2 \) term in

\[
\text{FIG. 1: (Color online) Schematic diagram showing electron transfer in a DQD structure starting at double-dot states (a) } | \uparrow_L \downarrow_R \rangle, \text{ (b) } | \downarrow_L \uparrow_R \rangle, \text{ (c) } | \uparrow_L \uparrow_R \rangle, \text{ and (d) } | \downarrow_L \downarrow_R \rangle. \text{ Electrons with } S_z = 0 \text{ in (a) and (b) admit transport under the effects of both nuclear fields and interdot tunneling. In contrast, transport for states with } S_z = \pm 1 \text{ in (c) and (d) is forbidden due to the high Pauli exclusion energy cost of the corresponding single-dot states.}
\]
$H_{\text{DQD}}$ in Eq. (2). However, due to the Coulomb barrier $\Delta$ at the right dot, all transport is suggested to be completely suppressed \cite{3}. We will explain later that there is a non-negligible chance that electrons can tunnel through the Coulomb blockade constituting an unmeasured leakage current. Other electrons from the left lead then enter and fill the DQD again. In all cases, there is a probability $P(\tau) = \rho_{33}(\tau) + \rho_{44}(\tau)$ that the DQD ends up at a non-spin-blocked state at the end of the manipulation stage. Here, $\rho_{\alpha\beta}(\tau)$ denotes a reduced density matrix element for the DQD after manipulation.

At the subsequent read-out and initialization stage, without the Coulomb barrier ($\Delta \simeq 0$), an electron can be transported if it is left at a non-spin-blocked state after manipulation. This occurs with probability $P(\tau)$. Once an electron is transported, a random number of electrons may follow until a spin-blocked state is reached again as explained above. It is easy to show that on average one electron follows. As a result, the current detected in the readout stage is

$$I_d(\tau) = \frac{2e}{T}[\rho_{33}(\tau) + \rho_{44}(\tau)],$$

where $e$ is the electronic charge and $T = 2$ $\mu$s. In particular, at large $\tau$ when coherence cannot be maintained, $\rho_{33}(\tau) = \rho_{44}(\tau) = 1/4$ and the current reduces to $I_d(\infty) = e/T$.

Bloch-type rate equation approach — We now derive a set of Bloch-type rate equations for the reduced density matrix elements $\rho_{\alpha\beta}$ of the DQD under the coherent manipulation by an ESR pulse. We adopt Gurvitz et al.‘s approach \cite{11} in which the many-body Schrödinger equation of the system is reduced to quantum rate equations by integrating out continuum reservoir states at the large voltage bias limit. We consider the high Zeeman splitting limit in the DQD so that spin flips caused by hyperfine interactions can be neglected. The rate equations for the diagonal density matrix elements are obtained after some algebra as \cite{13}

$$\dot{\rho}_{00} = -4\Gamma_0 \rho_{00} + \Gamma_r \rho_{55},$$
$$\dot{\rho}_{\alpha\alpha} = \Gamma_0 \rho_{\alpha\alpha} - i\langle m | [H_{\text{DQD}}, \hat{\rho}] | m \rangle,$$
$$\dot{\rho}_{55} = -\Gamma_r \rho_{55} - i\langle 5 | [H_{\text{DQD}}, \hat{\rho}] | 5 \rangle,$$

for $m = 1, 2, 3$ or 4 where $\rho_{00} = \rho_{\uparrow\downarrow} + \rho_{\downarrow\uparrow}$. Here, $\Gamma_0 = 2\pi \rho_0 \Omega_a^2$ is the transition rate for electron tunneling, while $\rho_0$ and $\Omega_a$ denote, respectively, the density of states and transition amplitude at lead $\alpha$. The rate equations for the off-diagonal elements are

$$\dot{\rho}_{mn} = -\frac{\xi_{mn}}{2} \Gamma_r \rho_{mn} + (1 - \xi_{mn}) \Gamma_0 \rho_{00} - i\langle m | [H_{\text{DQD}}, \hat{\rho}] | n \rangle,$$

for $m, n = 1, 2, 3, 4$ or 5 ($m \neq n$). The coefficient $\xi_{mn}$ equals one when $m$ or $n$ is 5 and equals zero otherwise.

Any time dependence of the coefficients in Eq. (4) has been eliminated by applying a rotating wave approximation, which is well justified under the electron spin resonant condition considered here \cite{14}.

We take typical model parameters appropriate for Koppen et al.‘s experiment. Specifically, the tunneling coupling constant between the dots is $V_0 = 0.25$ $\mu$eV. The transition rates at the leads are $\Gamma_l = \Gamma_r = 10V_0$ \cite{17}. The perpendicular field $B_{\text{ext}}^z$ is chosen as 100 mT.

In the experiments, spins at only one dot is at resonance with the ESR field. We assume that this occurs at the left dot at a frequency $\omega_c = g\mu_B B_{\text{ext}}^z$ with a field amplitude $B_{\text{ext}}^z$ varying from 0.74 mT to 1.5 mT. The field at the right dot is non-resonant and is neglected ($B_{\text{ext}}^z = 0$). The magnitude of fluctuations of the nuclear fields was experimentally found to be about 2.2 mT \cite{16}. We hence put $B_{\text{NR}}^z = 2.2$ mT and $B_{\text{NL}}^z = 0$ mT but other values of comparable magnitude give similar results.

We first consider the case of complete Coulomb blockade in the manipulation stage as assumed in Ref. \cite{8} by taking a large right-dot barrier $\Delta = 100V_0$. Starting from a spin-blocked initial state, say $|1\rangle$, we simulate the quantum transport and manipulation dynamics by numerically integrating the rate equations (5) and (6). The current $I_d(\tau)$ flowing through the DQD as a function of the burst period $\tau$ is then calculated using Eq. (4), and plotted in Fig. 2(a). It is observed that $I_d(\tau)$ oscillates periodically w.r.t. $\tau$. It can be well ap-
proximated by the analytical result at complete Coulomb blockade given by \[ I_d(\tau) = \frac{e}{\hbar}[1 - \cos(\Omega_{\text{Rabi}}\tau)] \], where \( \Omega_{\text{Rabi}} = g\mu_B B^x_t/2 \). This is due to a Rabi oscillation of the spin state of the electron at resonance with the ESR field. It hence implies an oscillation between the spin-blocked and unblocked states when projected onto the singlet-triplet basis and hence an oscillation of \( I_d(\tau) \) according to Eq. (1). The oscillation frequency \( \Omega_{\text{Rabi}} \) measured from our numerical data agrees with the exact relation above [Fig. 2(b)] and in particular has a linear dependence on the amplitude of the ESR field \( B^x_t \) in agreement with experiments. In this case, we observe practically no decay of the current oscillations as the Rabi oscillations have almost constant amplitudes. This is however in sharp contrast to the gradual decay from experiments. Therefore, our result shows clearly that decay cannot be explained by the quasi-static nuclear fields in the presence of complete Coulomb blockade as have been suggested by Koppens et al. [8]. This is in fact consistent with a recent experiment [11] showing that the relevant transition between triplet-singlet spin states in DQD as induced by nuclear fields takes much longer durations of about 100 \( \mu s \) and 1 ms at external fields of 30 mT and 150 mT, respectively. Furthermore, other spin-relaxation mechanisms including spin-orbit interactions and hyperfine interactions have even longer time scales [20, 21] and similarly cannot account for the decay of the current oscillations.

Our theory agrees much better with experiments when a tunneling leakage current through the Coulomb blockade barrier is considered. Here, we assume a more realistic value for the Coulomb barrier \( \Delta = 20V_0 = 5 \) \( \mu eV \) which will be further explained below. The corresponding simulation result is shown in Fig. 3. We find that the Rabi oscillations damp gradually during the ESR burst at a rate in good agreement with experiments. A smaller \( \Delta \) would lead to an even faster decay. The oscillation period \( \Omega_{\text{Rabi}} \) depends linearly on the amplitude of the ESR field as before. The current \( I_d(\tau) \) oscillates around the limiting value \( I_d(\infty) = e/T = 80 \) fA in agreement with experiments. Finally, the Coulomb barrier \( \Delta \) for an experimental device is limited by the fact that the right dot potential cannot be pulsed arbitrarily high because one electron has to remain all the time. From Eq. 3, one can see that \( \Delta \) does not only depend on the difference between the intra- and interdot Coulomb repulsions, but also on the difference of the orbital levels for the two dots. It can be shown that the value of \( \Delta \) used here is then a reasonable one for the experiment when these two differences are comparable.

In conclusion, we have investigated the coherent rotation of a single electron spin in a DQD system driven by an ESR field. A set of Bloch-type rate equations have been derived analytically and integrated numerically. We then obtain a current oscillation as a direct consequence of a coherent Rabi oscillation of an individual electron spin state in agreement with experiments. We also verify that the frequency of the oscillation has a linear dependence on the amplitude of the ESR field. A leakage current through a Coulomb blockade barrier during the manipulation stage is identified as the dominant mechanism of decoherence. Nuclear field fluctuations which induce a relaxation time much longer than the manipulation period are shown to contribute negligibly to the decoherence despite an earlier suggestion. Our detailed quantitative theory which allows the identification of the relevant decoherence mechanism is of great importance. Based on our result, it is evident that the coherence can be improved by suppressing the leakage current. This can be achieved by enhancing the Coulomb barrier \( \Delta \) by fabricating a smaller right dot in a similar device, or by decreasing the orbital level difference via tuning the appropriate gate voltages. We hope that our results can motivate further experimental investigations.

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![Graph showing current oscillations](image-url)
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