Coherent spin rotations in open driven double quantum dots

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We analyze coherent spin rotations in a dc biased double quantum dot driven by crossed dc and ac magnetic fields. In this configuration, spatial delocalization due to interdot tunneling competes with intradot spin rotations induced by the time dependent magnetic field, giving rise to a complicated time dependent behavior of the tunneling current. When the Zeeman splitting has the same value in both dots and spin flip is negligible, the electrons remain in the triplet subspace performing coherent spin rotations and current does not flow. This electronic trapping is removed either by finite spin relaxation or when the Zeeman splitting is different in each quantum dot. In the last case, we will show that by applying a resonant bichromatic magnetic field, the electrons become trapped in a coherent superposition of states and electronic transport is blocked. Then, manipulating ac magnetic fields allows one to drive electrons to perform coherent spin rotations which can be unambiguously detected by direct measurement of the tunneling current.

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I. INTRODUCTION

The accurate tunability of time dependent fields has allowed the access and manipulation of quantum systems by the resonant illumination of atoms, finding interesting effects such as the possibility of trapping the atom in a nonabsorbing coherent superposition (*dark state*) which is known as coherent population trapping.[1–3] This effect has been applied to nonconducting states in quantum dots (QDs)—also known as *artificial atoms*—for spinless electrons,[4,5] having revealed several advantages for practical issues such as electronic current switching[4] or decoherence probing.[6]

Great interest has recently focused on the coherent control of electron spin states in the search of candidates for qubits. Within this scope, optical trapping of localized spins has been treated in self-assembled quantum dots[7] and achieved in diamond defects.[8] Electron spin states in QDs have been proposed as qubits because of their long spin decoherence and relaxation times.[9,10] The controlled rotation of a single electron spin is one of the challenges for quantum computation purposes. In combination with the recently measured controlled exchange gate between two neighboring spins, driven coherent spin rotations would permit universal quantum operations. Recently, experimental and theoretical efforts have been devoted to describe electron spin resonance (ESR) in single[11] and double quantum dots (DQDs).[12,13] There, an ac magnetic field, B_{ac}, with a frequency resonant with the Zeeman splitting Δ induced by a dc magnetic field, B_{dc}, drives electrons to perform spin coherent rotations which can be perturbed by electron spin flip induced by scattering processes such as spin orbit or hyperfine interactions. These are manifested as a damping of the oscillations. In particular, hyperfine interaction between electron and nuclei spins induces flip-flop transitions and an effective Zeeman splitting which adds to the one induced by B_{dc}.[12,14,15] The ESR mechanism also allows one to access spin-orbit physics in the presence of ac electric fields[16,17] or vibrational degrees of freedom in nanomechanical resonators.[18]

In the experiments of Ref. 12, fast electric field switching was required in order to reach the Coulomb blockade regime and to manipulate the spin electron system. In the present work we analyze theoretically a simpler configuration, easier to perform experimentally than the one proposed in Ref. 12, which does not require one to bring the double occupied electronic state in the right dot to the Coulomb blockade configuration and which consists of conventional tunnel spectroscopy in a DQD under crossed dc and ac magnetic fields, *without additional electric pulses*.

The main purpose of this paper is to analyze the spin dynamics and the tunneling current and to propose how to trap electrons in a DQD performing coherent spin rotations by a resonant ac magnetic field which can be unambiguously detected by conventional tunneling spectroscopy measurements. We also show how to trap electrons by means of resonant bichromatic magnetic fields in the case where the Zeeman splitting is different in both QDs (as it usually happens in the presence of hyperfine interaction).

We consider a DQD in the *spin blockade* regime,[19] i.e., interdot tunneling is suppressed due to the Pauli exclusion principle[20] as the electrons in the DQD have parallel spins. This effect may be lifted by the rotation of the electrons spin, under certain conditions, by the introduction of crossed B_{dc} and B_{ac}. Then, when B_{ac} is resonant with the Zeeman splitted level, the electrons both rotate their spins within each QD and tunnel, performing spatial oscillations between the left and right QD. The electronic current through such a system performs coherent oscillations which depend nontrivially on both the ac intensity and the interdot coupling. We will see that, when the effective B_{dc} is homogeneous through the sample, current is quenched since the system is coherently trapped in the triplet subspace (*dark subspace*) in spite of the driving field. However, a finite current may flow as a consequence of spin relaxation processes. If Δ is different within each QD (it can be due to an inhomogeneous B_{dc}, different g factors, or the presence of hyperfine interaction[14] with different intensity within each QD), B_{ac} is resonant only in one of them and the trapping is lifted. Then, off-resonance dynamics of the other electron should in principle affect the total dynamics of the system and it should be included in a theoretical description not restricted to the rotating wave approximation[21] which is valid just at resonance. Finally we

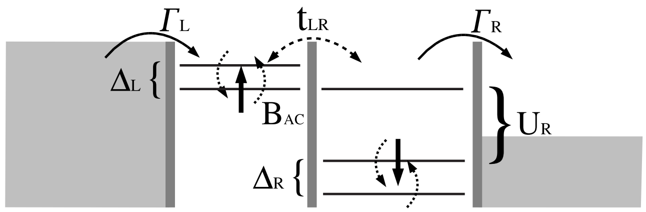

FIG. 1. Schematic diagram of the DQD in the presence of crossed dc and ac magnetic fields.

will show that it is possible to trap the electrons also in this configuration, where Δ is different within each QD, by applying a bichromatic B_{ac}, such that each frequency matches the Zeeman splitting in each QD.

II. MODEL

Our system consists on two weakly coupled QDs connected to two fermionic leads, described by the model Hamiltonian:

$$\hat{H}(t) = \hat{H}_0 + \hat{H}_{LR} + \hat{H}_T(t) + \hat{H}_{leads}, \quad (1)$$

where $\hat{H}_0=\Sigma_{i\sigma}\varepsilon_i\hat{c}^\dagger_{i\sigma}\hat{c}_{i\sigma}+\Sigma_iU_i\hat{n}_{i\uparrow}\hat{n}_{i\downarrow}+V\hat{n}_L\hat{n}_R$ describes the uncoupled DQD, $\hat{H}_{LR}=-\Sigma_\sigma(t_{LR}\hat{c}^\dagger_{L\sigma}\hat{c}_{R\sigma}+\text{H.c.})$ is the interdot coupling, and $\hat{H}_T=\Sigma_{l\epsilon\{L,R\}k\sigma}(\gamma_l\hat{d}^\dagger_{lk\sigma}\hat{c}_{l\sigma}+\text{H.c.})$ gives the tunneling between the DQD and the leads, described by $\hat{H}_{leads}=\Sigma_{lk\sigma}\varepsilon_{lk}\hat{d}^\dagger_{lk\sigma}\hat{d}_{lk\sigma}$, where ε_i is the energy of an electron located in dot i and U_i (V) is the intradot (interdot) Coulomb repulsion. For simplicity, we disregard the Heisenberg exchange interaction.[15,20] Finite exchange would slightly split the interdot singlet-triplet energy separation without modifying qualitatively the results presented here. The chemical potentials of the leads, μ_i, are such that only two electrons (one in each dot) are allowed in the system: $\varepsilon_i<\mu_i-V<\varepsilon_i+U_i$ and $\mu_i<\varepsilon_i+2V$. In this configuration, the spin blockade is manifested when a bias voltage is applied such that the state with two electrons in the right dot (the one which contributes to the current) is in resonance with those with one electron in each dot. The current is then quenched when the electrons in each QD have the same spin polarization and Pauli exclusion principle avoids the interdot tunneling.[20] We now introduce a magnetic field with a dc component along the Z axis (which breaks the spin degeneration by a Zeeman splitting $\Delta_i=g_iB_{z,i}$) and a circularly polarized ac component in the perpendicular plane XY that rotates the Z component of the electron spin when its frequency satisfies the resonance condition, $\omega=\Delta_i$:

$$\hat{H}_B(t) = \sum_i [\Delta_iS_z^i + B_{ac}(S_x^i\cos\omega t + S_y^i\sin\omega t)], \quad (2)$$

where $\mathbf{S}_i=(1/2)\Sigma_{\sigma\sigma'}c^\dagger_{i\sigma}\sigma_{\sigma\sigma'}c_{i\sigma'}$ are the spin operators of each dot (see Fig. 1).

The dynamics of the system is given by the time evolution of the reduced density matrix elements, whose equation of motion, within the Born-Markov approximation,[22] reads

$$\dot{\rho}_{ln}(t) = -i\langle l|[H_0+H_{LR}+H_B(t),\rho]|n\rangle + \sum_{k\neq n}(\Gamma_{nk}\rho_{kk}-\Gamma_{kn}\rho_{nn})\delta_{ln} - \Lambda_{ln}\rho_{ln}(1-\delta_{ln}), \quad (3)$$

where the first term in the right-hand side accounts for the coherent dynamics within the double quantum dot. Γ_{ln} are the transition rates from state $|n\rangle$ to $|l\rangle$ including those induced by the coupling to the leads—being $\Gamma_i=2\pi|\gamma_i|^2$ when they occur through lead $i\epsilon\{L,R\}$—and the eventual spin scattering processes [introduced phenomenologically by the spin relaxation rate, T_1^{-1} (Ref. 23)]. Decoherence appears due to the term $\Lambda_{ln}=\frac{1}{2}\Sigma_k(\Gamma_{kl}+\Gamma_{kn})+T_2^{-1}$, $T_2=0.1T_1$ being the intrinsic spin decoherence time. The evolution of the occupation probabilities is given by the diagonal elements of the density matrix. In our configuration, the states relevant to the dynamics are $|0,\uparrow\rangle$, $|0,\downarrow\rangle$, $|T_+\rangle=|\uparrow,\uparrow\rangle$, $|T_-\rangle=|\downarrow,\downarrow\rangle$, $|\uparrow,\downarrow\rangle$, $|\downarrow,\uparrow\rangle$, $|S_R\rangle=|0,\uparrow\downarrow\rangle$. This latest state is the only one that contributes to tunneling to the right lead, so the current is given by

$$I(t) = 2e\Gamma_R\rho_{S_R,S_R}(t). \quad (4)$$

Each coherent process is described by a *Rabi-like* frequency. For instance, in the case of two *isolated* spins, one in each QD, which are in resonance with B_{ac} ($\Delta_L=\Delta_R$), the oscillation frequency is $\Omega_{ac}=2B_{ac}$, see Appendix A 1. On the other hand, the interdot tunneling events can be described by the resonance transitions between the states $|\uparrow,\downarrow\rangle$, $|\downarrow,\uparrow\rangle$, and $|S_R\rangle$, whose populations oscillate with a frequency $\Omega_T=2\sqrt{2}t_{LR}$, as shown in Appendix A 2.

A. $\Delta_L=\Delta_R$

We consider initially the case where B_{dc} is homogeneous, so that $\Delta_R=\Delta_L$ and both spins rotate simultaneously. Then, the dynamics of the system is properly described in terms of the dynamics of the total spin of the DQD. B_{ac} acts only on the states with a finite total magnetic moment: $|T_\pm\rangle$ and $|T_0\rangle=\frac{1}{\sqrt{2}}(|\uparrow,\downarrow\rangle+|\downarrow,\uparrow\rangle)$, while the interdot tunneling, that does not change the spin, is only possible between $|S_R\rangle$ and $|S_0\rangle=\frac{1}{\sqrt{2}}(|\uparrow,\downarrow\rangle-|\downarrow,\uparrow\rangle)$. Therefore in the absence of spin relaxation, spin rotation and interdot hopping are independent processes so any eventual singlet component will decay by tunneling to the contacts. This produces a finite current in the transitory regime which drops to zero for longer times. This process is independent of B_{ac}, which is manifested in the frequency of the current oscillations, Ω_T, cf. Fig. 2(a). Thus for large enough times($t\gg\Gamma_i^{-1}$), transport is canceled and one electron will be confined in each QD. The electrons will be coherently trapped in the interdot triplet subspace, $T_\pm$, T_0 (dark subspace) and behave as an isolated single particle of angular momentum $S=1$ performing coherent spin rotations with a frequency Ω_{ac} [Fig. 2(b)].

A finite spin relaxation time mixes the dynamics of the singlet and the triplet subspaces, so that interdot tunneling is allowed and finite current appears, cf. Fig. 3(a). The shorter the spin relaxation time, the larger is the singlet-triplet mixing and therefore, the higher is the current, cf. Fig. 4(a), up to relaxation times fast enough to dominate the electron dynamics ($T_1^{-1}\gg\Omega_{ac}$). In this case, ESR is not effective in order to

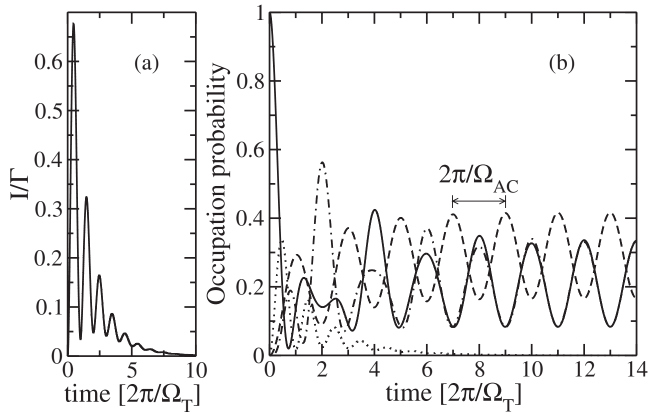

FIG. 2. (a) $I(t)$ for initial state $|\uparrow,\downarrow\rangle$ in the absence of spin relaxation for $\Delta_L=\Delta_R=\Delta$ and $\Omega_{ac}=\Omega_T/2$. (b) The corresponding occupation probabilities: $|\uparrow,\downarrow\rangle$ (solid line), $|\downarrow,\uparrow\rangle$ (dash-dotted line), $|0,\uparrow\downarrow\rangle$ (dotted line), and $|\uparrow,\uparrow\rangle$ and $|\downarrow,\downarrow\rangle$ (dashed line). Parameters ($e=\hbar=1$): $\Gamma_L=\Gamma_R=\Gamma=10^{-3}$ meV, $T_{1(2)}^{-1}=0$, Ω_T $=11.2$ GHz and holding for the rest of the plots (in meV): $\varepsilon_L=1.5$, $\varepsilon_R=0.45$, $\Delta=0.026$ ($B_{dc}\sim 1T$), $U_L=1$, $U_R=1.45$, $V=0.4$, $\mu_L=2$, and $\mu_R=1.1$.

rotate the spins and spin blockade is recovered, cf. Fig. 4(b). Since both spin rotations and spatial delocalization are resonant processes, this singlet-triplet mixing produces complicated dynamics in the current that shows oscillations with a frequency that depends both on the interdot coupling and the ac field intensity, cf. Fig. 4(c). When B_{ac} increases, the frequency of the current oscillations increases but not linearly due to the interplay with the hopping. This effect is small for long spin relaxation times.

B. $\Delta_L\neq\Delta_R$

However, if one introduces an inhomogeneous B_{dc}, so that only one of the electrons is in resonance with B_{ac} (for instance, $\omega=\Delta_R\neq\Delta_L$), the total spin symmetry is broken and then the electron in each QD behaves differently. In fact, the states $|\downarrow,\uparrow\rangle$ and $|\uparrow,\downarrow\rangle$ have different occupation probabilities and interdot hopping induces the delocalization of the individual spins. This populates the state $|S_R\rangle$ and a finite current appears showing a double peak whose position shifts following the inhomogenity, cf. Fig. 3(b). This double peak may be the origin of the under-resolved structure measured in Ref. 12. By tuning the Zeeman splittings difference, the current presents an antiresonance of depth ~ 0.1 nA near $\Delta_L=\Delta_R$, cf. Fig. 5(a), pretty similar to the coherently trapped atom spectrum in quantum optics.[3] As expected, taking one electron slightly out of resonance, the frequency of the current oscillation is modified in comparison with the double resonance situation. If one electron is far enough from resonance, the frequency of the current oscillation becomes roughly half of the value as it would be the case for the rotation of one electron spin, cf. Fig. 5(b). Otherwise, the off-resonant electron modifies the Rabi frequency for spin rotations in a more complicated way depending on B_{ac}, t_{LR}, and how much both dynamics are mixed (which is related to $\Delta_L-\Delta_R$), cf. Fig. 5(c). The limiting case when Δ_L and Δ_R are very different and only the electron in the right QD is affected effectively by B_{ac} is analyzed in Appendix A 3.

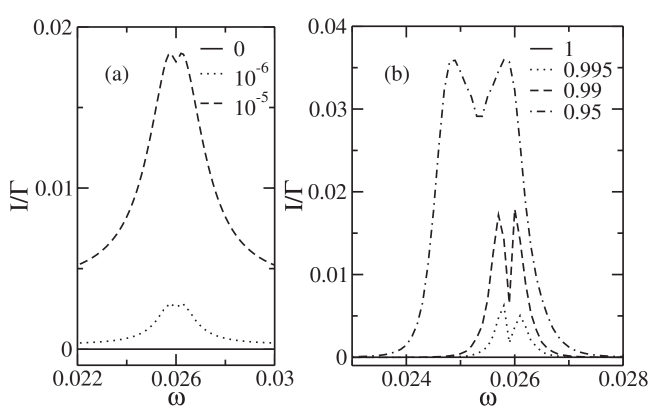

FIG. 3. Effect of (a) finite spin relaxation rates, T_1^{-1} and (b) the Zeeman inhomogenity, Δ_L/Δ_R, on the stationary current when tuning the frequency of the magnetic field. In (a), $\Delta_L=\Delta_R$; in (b), T_1 $=0$. (Same parameters as in Fig. 2 but $\Gamma=10^{-2}$ meV).

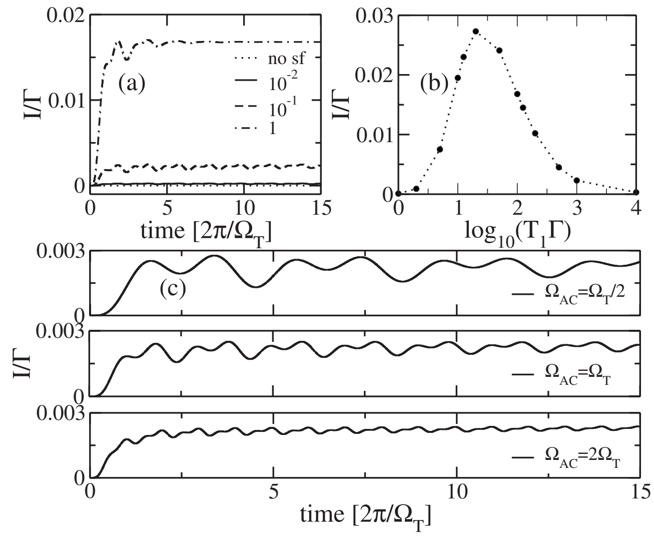

FIG. 4. (a) $I(t)$ for different spin-flip times (in μs), with Ω_{ac} $=\Omega_T=11.2$ GHz and $\Delta_L=\Delta_R=\Delta$. The initial state here is $|\uparrow,\uparrow\rangle$, then, for $T_1^{-1}=0$, there is no mixing of the triplet and singlet subspaces and therefore no current flows through the system. Spin relaxation processes contribute to populate the singlet, producing a finite current. (b) Stationary current as a function of spin relaxation time. For long T_1, electrons remain in the dark space. As T_1 decreases, I begins to flow, being again suppressed for short enough T_1, as discussed in the text. (c) $I(t)$ for different ratios between the ac field intensity and the interdot hopping, i.e., between Ω_{ac} and Ω_T, with $T_1\sim 0.1$ μs. (Same parameters as in Fig. 3.)

III. BICHROMATIC FIELD

There is a way for trapping the system in a dark state even for different Zeeman splittings by introducing a bichromatic B_{ac} with a different frequency that also brings into resonance the electron in the left QD:

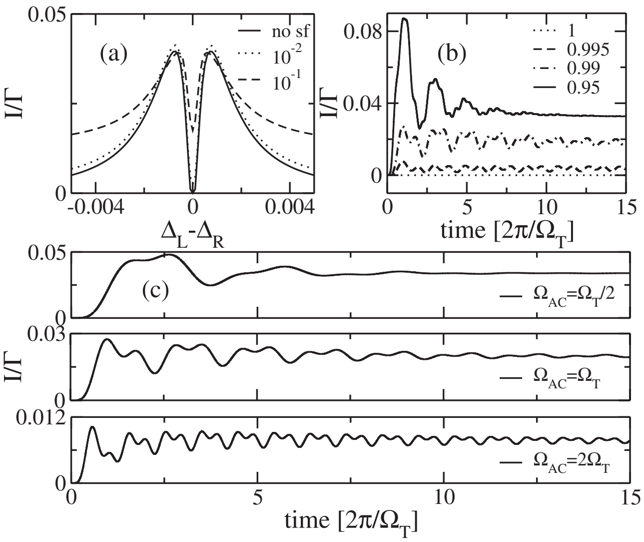

FIG. 5. (a) Dependence of the stationary current on the dc field inhomogeneity $\Delta_L-\Delta_R$ for different relaxation times (in μs). The quenching of the current for $\Delta_L=\Delta_R$ is lifted by spin relaxation. (b) $I(t)$ for different values of Δ_L/Δ_R when the electron in the right QD is kept in resonance, in the absence of relaxation. A crossover to the one electron spin resonance is observed by increasing the difference between Δ_L and $\Delta_R=\Delta$. (c) Dependence of the current oscillations on B_{ac} for $\Delta_L=0.99\Delta_R$ and $T_1^{-1}\sim 0.1$ μs. Same parameters as in Fig. 4.

$$\hat{H}_B^{(2)}(t)=\sum_{\substack{i=L,R\\ j=1,2}}[\Delta_i\hat{S}_z^i+B_{ac}(\hat{S}_x^i\cos\omega_j t+\hat{S}_y^i\sin\omega_j t)], \quad (5)$$

with $\omega_1=\Delta_L$ and $\omega_2=\Delta_R$. Then, each electron is resonant with one of the field frequencies. In this case, as Δ_i is different in both QDs, $|T_0\rangle$ mixes with $|S_0\rangle$ and a finite current flows until the electrons fall in the superposition: $|S_2\rangle=\frac{1}{\sqrt{2}}(|\uparrow,\uparrow\rangle-|\downarrow,\downarrow\rangle)$ which is not affected by the magnetic field but for off-resonant oscillations that can be averaged out. In effect, if the nonresonant terms are disregarded, Eq. (5) is reduced to $\hat{\tilde{H}}_{B,0}^{(2)}(t)=\Sigma_i[\Delta_i\hat{S}_z^i+B_{ac}(\hat{S}_x^i\cos\Delta_i t+\hat{S}_y^i\sin\Delta_i t)]$ and $\hat{\tilde{H}}_{B,0}^{(2)}|S_2\rangle=0$. Then the population of the states $|\uparrow,\downarrow\rangle$, $|\downarrow,\uparrow\rangle$, and $|S_R\rangle$ and, therefore, the current drop to zero, see Fig. 6(a). This transport quenching also allows one to operate the system as a *current switch* by tuning the frequencies of the ac fields [Fig. 6(b)] and the preparation of the system in a concrete superposition to be manipulated.

The application of a bichromatic magnetic field provides a direct measurement of the Zeeman splittings of the dots by tuning the frequencies until the current is brought to a minimum as in Fig. 6(b). Then, by *switching* one of the frequencies off and tuning the Zeeman splitting by an additional B_{dc} in one of the dots, the antiresonance configuration of Fig. 5(a) could be achieved. In this case, electrons in both QDs perform coherent spin rotations, as shown in Fig. 2(b).

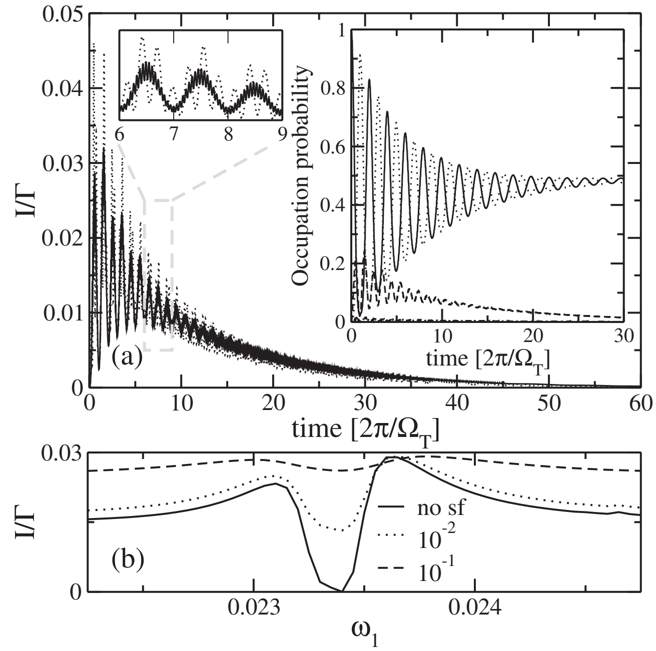

FIG. 6. (a) Transient current in the presence of a bichromatic B, when $\omega_{1(2)}=\Delta_{L(R)}$ for $\Delta_L=\Delta_R/2$ (solid) and $\Delta_L=0.9\Delta_R$ (dotted) and $T_1^{-1}=0$. Left inset: detail of the current oscillation. In the case where $\Delta_L=\Delta_R/2$, I oscillates with Ω_T and it presents faster oscillations overimposed (more important for $\Delta_L=0.9\Delta_R$) coming from the effect of each frequency on its off-resonance electron. Right inset: occupation probabilities for $\Delta_L=\Delta_R/2$: $|\uparrow,\uparrow\rangle$ (solid line), $|\downarrow,\downarrow\rangle$ (dotted line), $|\uparrow,\downarrow\rangle\sim|\downarrow,\uparrow\rangle$ (dashed line), and $|0,\uparrow\downarrow\rangle$ (dash-dotted line, remaining very close to zero). The occupation of $|0,\uparrow\downarrow\rangle$ drops to zero, and therefore I drops as well. At long times the electrons fall in a coherent superposition of $|\uparrow,\uparrow\rangle$ and $|\downarrow,\downarrow\rangle$. (b) Stationary current as a function of ω_1 when $\omega_2=\Delta_R$, for different relaxation rates, T_1^{-1}. I drops at $\omega_1=\Delta_L$. ($\Delta_R=\Delta$, $\Gamma=10^{-3}$ meV, and $\Omega_T=1.12$ GHz.)

IV. CONCLUSIONS

In summary, we present the complete electron spin dynamics in a DQD, in the spin blockade regime, with up to two extra electrons, where crossed dc and ac magnetic fields and a dc voltage are applied. In the experimental setup that we propose, different Rabi oscillations (due to the ac magnetic field and the interdot tunneling) compete: The time dependent magnetic field produces coherent spin rotations between spin up and down states while resonant interdot hopping allows the spatial delocalization of the electrons. We show how the interplay between coherent oscillations coming from the interdot tunnel and those due to B_{ac} gives rise to a nontrivial electron dynamics which strongly depends on the ratio between the different Rabi frequencies involved. We show as well that if Δ has the same value for the left and the right QD, electrons remain performing coherent spin rotations in the $S=1$ subspace and current is quenched. This electron trapping is removed by spin relaxation or inhomogeneous B_{dc} and finite current flows. *Measuring the current will allow one to control coherent spin rotations* and also to extract information on the spin relaxation time. We propose as well how to block the current by a bichromatic magnetic field in a DQD where the effective Zeeman splitting is different within each dot (and where current would otherwise flow due to singlet-triplet mixing). We demonstrate that the bichromatic field induces spin blockade in this configuration and that the system evolves to a stationary superposition of states, thus serving for spin rectification and state preparation.

Then, our results show that tunneling spectroscopy experiments in DQDs under tunable mono- and bichromatic magnetic fields allow one to drive electrons to perform coherent spin rotations which *can be unambiguously detected* by measuring the tunneling current. We also show how to induce spin blockade in DQDs with different Zeeman splittings by means of a bichromatic magnetic field.

ACKNOWLEDGMENTS

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APPENDIX: CLOSED SYSTEM

In this appendix, we present some simple cases that describe the purely coherent dynamics (i.e., for $\Gamma_L=\Gamma_R=0$ and $T_1^{-1}=0$) involved in the description presented above.

1. TWO ISOLATED ELECTRONS SPIN RESONANCE

We consider first the case where each electron is isolated in one quantum dot. This system is described by the Hamiltonian $\hat{H}(t)=\hat{H}_0+\hat{H}_B(t)$ (as written in Sec. II) and the basis $|1\rangle=|\uparrow,\uparrow\rangle$, $|2\rangle=|\downarrow,\uparrow\rangle$, $|3\rangle=|\uparrow,\downarrow\rangle$, and $|4\rangle=|\downarrow,\downarrow\rangle$. We obtain the equations of motion for the reduced density matrix elements from the Liouville equation $\dot{\rho}(t)=-i[H(t),\rho(t)]$. After a variable transformation: $\rho'_{12,24,34}=e^{-i\omega t}\rho_{12,24,34}$ and $\rho'_{14}=e^{-i2\omega t}\rho_{14}$, they can be written as

$$\dot{\rho}_1 = B_{ac}\Im(\rho'_{21}+\rho'_{31}),$$

$$\dot{\rho}_2 = B_{ac}\Im(\rho'_{12}+\rho'_{42}),$$

$$\dot{\rho}_3 = B_{ac}\Im(\rho'_{43}+\rho'_{13}),$$

$$\dot{\rho}_4 = B_{ac}\Im(\rho'_{34}+\rho'_{24}) \tag{A1}$$

for the diagonal terms, and

$$\dot{\rho}'_{12} = -\frac{i}{2}B_{ac}(\rho_2-\rho_1+\rho_{32}-\rho'_{14})+i(\Delta_L-\omega)\rho'_{12},$$

$$\dot{\rho}'_{13} = -\frac{i}{2}B_{ac}(\rho_3-\rho_1+\rho_{23}-\rho'_{14})+i(\Delta_R-\omega)\rho'_{13},$$

$$\dot{\rho}'_{14} = -\frac{i}{2}B_{ac}(\rho'_{24}+\rho'_{34}-\rho'_{12}-\rho'_{13})+i(\zeta-2\omega)\rho'_{14},$$

$$\dot{\rho}_{23} = -\frac{i}{2}B_{ac}(\rho'_{43}-\rho'_{21}+\rho'_{13}-\rho'_{24})-i\eta\rho_{23},$$

$$\dot{\rho}'_{24} = -\frac{i}{2}B_{ac}(\rho_4-\rho_2-\rho_{23}+\rho'_{14})+i(\Delta_R-\omega)\rho'_{24},$$

$$\dot{\rho}'_{34} = -\frac{i}{2}B_{ac}(\rho_4-\rho_3-\rho_{32}+\rho'_{14})+i(\Delta_L-\omega)\rho'_{34} \tag{A2}$$

for the coherences, where $\zeta=\Delta_L+\Delta_R$ and $\eta=\Delta_L-\Delta_R$.

The set of equations (A1) and (A2) can be solved by doing the Laplace transform, $\mathcal{L}\dot{\rho}=z\rho-\rho(0)$ and considering the initial condition $\rho_1(0)=1$. If the effect of the magnetic field is the same for both electrons, that is, they suffer the same Zeeman splitting, $\Delta_L=\Delta_R=\Delta$, the probability of finding only one of the electrons flipped, $P_f=\rho_2+\rho_3$ is

$$P_f=\frac{2B_{ac}^2}{\Theta^4}\left(\frac{B_{ac}^2}{4}\sin^2\Theta t+\delta^2\sin^2\frac{1}{2}\Theta t\right), \tag{A3}$$

where $\Theta^2=B_{ac}^2+\delta^2$ and $\delta=\Delta-\omega$. In the resonant case, $\delta=0$:

$$P_f=\frac{1}{2}\sin^2 B_{ac}t. \tag{A4}$$

Therefore the Rabi frequency for this configuration is

$$\Omega_{ac}=2B_{ac}, \tag{A5}$$

twice the one found for the single electron case.[24]

On the other hand, if $\Delta_L\neq\Delta_R$, the resonance condition holds only for one of them. Then, there is a superposition of different oscillations which results in a complicated dynamics when $|\eta|\ll\zeta$.[25]

2. ELECTRON DELOCALIZATION

Let us now consider the closed system in the absence of magnetic field, which can be described by the Hamiltonian $H=H_0+H_{LR}$. The interdot coupling term, H_{LR}, induces electron tunneling between both dots, involving the states $|1\rangle=|\uparrow,\downarrow\rangle$, $|2\rangle=|\downarrow,\uparrow\rangle$, and $|3\rangle=|0,\uparrow\downarrow\rangle$. Then, the Liouville equation is given by

$$\dot{\rho}_1=-2t_{LR}\Im\rho_{31},$$

$$\dot{\rho}_2=2t_{LR}\Im\rho_{32},$$

$$\dot{\rho}_3=2t_{LR}\Im(\rho_{31}-\rho_{32}),$$

$$\dot{\rho}_{12}=it_{LR}(\rho_{32}+\rho_{13}),$$

$$\dot{\rho}_{13}=it_{LR}(\rho_3-\rho_1+\rho_{12})-i(\varepsilon_L-\varepsilon_R+V-U_R)\rho_{13},$$

$$\dot{\rho}_{23}=-it_{LR}(\rho_3-\rho_2+\rho_{21})-i(\varepsilon_L-\varepsilon_R+V-U_R)\rho_{23}. \tag{A6}$$

By solving the set of equations (A6) under the condition $\varepsilon_L-\varepsilon_R=U_R-V$, where interdot tunneling is resonant, we obtain the occupation of the state $|0,\uparrow\downarrow\rangle$, which is given by

$$\rho_3=\frac{1}{2}\sin^2\sqrt{2}t_{LR}t. \tag{A7}$$

Thus the Rabi frequency is modified with respect to the single electron case [$\Omega_{1e}=2t_{LR}$ (Ref. 24)]:

$$\Omega_T = 2\sqrt{2}t_{LR}. \tag{A8}$$

3. MIXING OF SPATIAL DELOCALIZATION AND SPIN ROTATION

As discussed in the text for the open system, if $\Delta_L \neq \Delta_R$, the current shows a coherent oscillation that depends on both the intensity of the ac magnetic field and the interdot hopping.

Here we consider a simple case that presents both coherent processes—spin rotation and interdot delocalization—by considering very different Zeeman splittings in each QD. Then, only one of the electrons is in resonance with the ac field: $\Delta_L = \omega$ and only three states contribute to the dynamics: $|1\rangle = |\uparrow,\uparrow\rangle$, $|2\rangle = |\downarrow,\uparrow\rangle$, and $|3\rangle = |0,\uparrow\downarrow\rangle$, resulting in the set of equations:

$$\dot{\rho}_1 = B_{ac}\Im\rho_{21},$$

$$\dot{\rho}_2 = B_{ac}\Im\rho_{12} + 2t_{LR}\Im\rho_{32},$$

$$\dot{\rho}_3 = -2t_{LR}\Im\rho_{32},$$

$$\dot{\rho}_{12} = -\frac{i}{2}B_{ac}(\rho_2 - \rho_1) + it_{LR}\rho_{13} + i(\Delta_L - \omega)\rho_{12},$$

$$\dot{\rho}_{13} = -\frac{i}{2}B_{ac}\rho_{23} + it_{LR}\rho_{12} - i\omega_+\rho_{13},$$

$$\dot{\rho}_{23} = -\frac{i}{2}B_{ac}\rho_{13} - it_{LR}(\rho_3 - \rho_2) - i\omega_-\rho_{23}, \tag{A9}$$

where $\omega_\pm = \varepsilon_L - \varepsilon_R - \frac{\Delta_R \pm \Delta_L}{2} + V - U_R$. If the gate voltages are tuned in a way that $\omega_- = 0$, so the left electron can tunnel to the doubly occupied singlet state in the right dot (having both electrons opposite spin polarization), one finds that the frequency of the oscillations depends on both the tunneling coupling and the field intensity: $\Omega \propto \sqrt{B_{ac}^2 + 4t_{LR}^2}$.

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