Control of Spin Blockade by ac Magnetic Fields in Triple Quantum Dots

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We analyze coherent spin phenomena in triple quantum dots in triangular configuration under crossed dc and ac magnetic fields. In particular, we discuss the interplay between Aharonov-Bohm current oscillations, coherent electron trapping and spin blockade under two-electron spin resonance configurations. We demonstrate an unexpected antiresonant behaviour in the current, allowing for both removal and restoration of maximally entangled spin blockaded states by tuning the ac field frequency. Our theoretical predictions indicate how to manipulate spin qubits in a triangular quantum dot array.

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Electronic transport through mesoscopic systems can become correlated not only by charge interaction but also by the spin degree of freedom. A dramatic combination of both can be found in systems where strong Coulomb interaction limits the population to a small number of electrons (Coulomb blockade) and where Pauli exclusion principle avoids certain internal transitions — spin blockade (SB). This was first observed as a rectification effect in the current through a double quantum dot (DQD)1. Recent experiments have taken advantage of SB to achieve qubit operations in a double dot by electric gate control2 or by electron spin resonance (ESR)3. It consists in inducing transitions between the electron’s spin-up and spin-down states, which are splitted by the Zeeman energy coming from a dc magnetic field, $B_{dc}$. Different mechanisms have been considered: crossed dc and ac magnetic fields ($B_{ac}$), where the ac frequency is resonant with the Zeeman splitting4,5, effective $B_{ac}$ induced by ac electric fields in the presence of spin-orbit interaction6, slanting Zeeman fields7 or hyperfine interaction8.

Lately, a next step towards quantum dot arrays has been reached: tunnel spectroscopy measurements with triple quantum dots (TQD), both in series9 and in triangular configurations10 have been achieved. Theoretical works in these systems11–12 analyze their eigenstates and stability diagram, as well as the effect of a magnetic field penetrating the structure. TQDs with strongly correlated electrons have also been investigated in the Kondo regime13 and have been proposed as spin entanglers14. Additionally, these systems show a more peculiar property which is intrinsic to three-level systems, namely coherent population trapping, which is a well-known effect in quantum optics and which was observed in three-level atoms excited by two resonant laser fields15. There, the electronic wave function evolves towards an eigenstate superposition, a so-called dark state, which is decoupled from the laser fields and therefore it manifests as an antiresonance in the emission spectrum. An analogy in transport has been made when coherent superpositions avoid transport by interference between tunneling events. These dark states can be achieved by driving three-level double dots with bichromatic ac electric fields16 or by the interference of tunneling processes in TQDs17–18. It was shown19 how coherent trapping can be lifted in closed-loop TQDs by means of the Aharonov-Bohm (A-B) effect20.

Here, we will discuss the electron spin dynamics and transport for the case where a triangular TQD contains up to two extra electrons, as shown in Fig. 1. In contrast to the single electron case, spin correlations can influence transport due to SB21. We will show that at certain $B_{ac}$ frequencies and sample configurations, the magnetic field brings the electronic wave function into a superposition of parallel spins states, unexpectedly bringing the system back to SB.

Model — We consider a system consisting of three dots
which are coupled through tunnel barriers, and dots 1 and 3 are also connected to source and drain contacts respectively. The Hamiltonian of the system is \( \hat{H}(t) = \hat{H}_{\text{TQD}} + \hat{H}_t + \hat{H}_{\text{leads}} + \hat{H}_B(t) \). \( \hat{H}_{\text{TQD}} = \sum_{i>\sigma} c_{i\sigma}^\dagger c_{i\sigma} + \sum_{i} U_i n_i n_i + \sum_{i,i\neq j} V_{ij} n_i n_j \) describes the uncoupled TQD. \( \hat{H}_t = -\sum_{ij} (\tau_{ij} c_{i\sigma}^\dagger c_{j\sigma} + \text{h.c.}) \) describes the coherent tunneling between the dots, \( \hat{H}_B = \sum_{i \in \text{L,R}, \sigma} (\gamma_i d_{i\sigma}^\dagger c_{i\sigma} + \text{h.c.}) \) describes the coupling of the dots to the leads, and \( \hat{H}_{\text{leads}} = \sum_{i \sigma} \epsilon_i d_{i\sigma}^\dagger d_{i\sigma} \) the leads themselves. \( \epsilon_i \) is the energy of an electron located in dot \( i \), \( U_i \) is the intradot and \( V_{ij} = V \) the inter-dot Coulomb repulsion. If not stated otherwise, we set \( |\tau_{ij}| = \tau \).

The Hamiltonian for the magnetic field, \( \hat{H}_B(t) \), has two components: a time-independent dc component along the \( z \)-axis that breaks the spin-degeneracy by a Zeeman-splitting \( \Delta_i = g_i B z_i \), and a circularly polarized ac component in the \( xy \)-plane that rotates the \( z \)-component of the electron spin when its frequency fulfills the resonance condition \( \hbar \omega_i = \Delta_i \). It reads \( \hat{H}_B(t) = \sum_{i=1}^3 \Delta_i \hat{S}_{z_i} + B_{\text{ac}}(\cos(\omega t) \hat{S}_{z_i} + \sin(\omega t) \hat{S}_{y_i}) \), being \( \hat{S}_i = \frac{1}{2} \sum_{\sigma \sigma'} c_{i\sigma}^\dagger c_{i\sigma'} \sigma \sigma' \) the spin operator of the \( i \)-th dot. As shown in, the ac magnetic field has no effect on SB unless the Zeeman splitting is inhomogeneous in the sample. Here we will consider the simplest configuration that allows to analyze the relevant mechanisms: \( \Delta_1 = \Delta_2 \neq \Delta_3 \). The experimental feasibility justifies the choice of the present configuration.

The dynamics of the system is given by the time evolution of the reduced density matrix whose equations of motion read as, within the Born-Markov-approximation:

\[
\dot{\rho}(t) = -i \langle \{ \hat{H}_{\text{TQD}} + \hat{H}_t + \hat{H}_B(t), \rho \} | n \rangle \\
+ \sum_{k \neq n} (\Gamma_{nk} \rho_{kk} - \Gamma_{kn} \rho_{nn}) \delta_{ln} - \Lambda_{nn} \rho_{nn} (1 - \delta_{ln})
\]

The commutator accounts for the coherent dynamics in the TQD, \( \Gamma_{nn} \) are the transition rates from state \( |n \rangle \) to state \( |l \rangle \) induced by the coupling to the leads and decoherence appears by \( \Lambda_{nn} = \frac{1}{2} \sum_k (\Gamma_{kl} + \Gamma_{lk}) \).

We consider a configuration where the dot coupled to the drain is permanently occupied by one electron (see Fig. 1), and only up to two electrons can be in the system. Double occupancy is only allowed in the drain dot. This is the case when the chemical potentials in the leads satisfy \( \epsilon_3 + V < \mu_R < \epsilon_3 + U_3 \) and \( \mu_L < \epsilon_1 + 2V \). For resonant tunneling, \( \epsilon_1 = \epsilon_2 \) and \( \epsilon_1 + 2V = \epsilon_3 + U_3 \). Out of the full TQD basis with up to two electrons, there are then eleven one- and two-electron states that dominate the dynamics: \( |0,0,\sigma \rangle, |\chi_{\sigma\sigma} \rangle = |\sigma,0,\sigma \rangle, |\chi_{2\sigma\sigma} \rangle = |0,\sigma,\sigma \rangle \) and \( |S_3 \rangle = |0,0,\uparrow \rangle \), with \( \sigma, \sigma' = \{ \uparrow, \downarrow \} \). Transport is biased from left to right and only state \( |S_3 \rangle \) contributes to tunneling through to the drain, acting as a bottleneck for the current: \( I(t) = \sum_n \Gamma_{nS_3} \rho_{S_3S_3}(t) \). Though being confined, the electron in dot 3 is essential to induce spin correlated transport. A \( B_{\text{ac}} \) perpendicular to the plane of the triangular dot structure (Fig. 1) encloses a magnetic flux \( \Phi \) such that electron tunneling acquires an additional phase \( \phi = 2\pi \Phi/\Phi_0 \), with \( \Phi_0 = h/e \) being the flux quantum. We accumulate the phase between dot 1 and 2, \( \tau_{12} = \tau e^{-i\phi} \).

Undriven case \((B_{\text{ac}} = 0)\): — It is well known\(^2\) for a TQD with up to one extra electron, that due to interference, the current oscillates with \( \Phi \) (A-B oscillations) and periodically drops to zero with a periodicity of \( \Phi_0/2 \). For the understanding of the two-electron spin dynamics, it is crucial to look at the eigenstates of this system, which change depending on the flux \( \Phi \). For \( \Phi/\Phi_0 = 0 \):

\[
|\psi^-_{\sigma\sigma} \rangle = \frac{1}{\sqrt{2}}(|\chi_{2\sigma\sigma} \rangle - |\chi_{\sigma\sigma} \rangle) \quad \sigma, \sigma' = \{ \uparrow, \downarrow \} \\
|\psi^+_{\sigma\sigma} \rangle = \frac{1}{\sqrt{2}}(|\chi_{2\sigma\sigma} \rangle + |\chi_{\sigma\sigma} \rangle) \quad \sigma = \{ \uparrow, \downarrow \}
\]

are eigenstates of the closed system. States (2) avoid tunneling to \( |S_3 \rangle \): \( \langle \psi^-_{\sigma\sigma}, \hat{H}_B | S_3 \rangle = 0 \), which is why they are also called dark states, see Fig. 1. Occupation of \( |S_3 \rangle \) thus decays by the coupling to the drain (Fig. 2b) and current is blocked. The states in (2) remind of the dark states found in the single electron case\(^2\). A significant difference is that for two electrons the spin degree of freedom plays a role: Pauli exclusion principle introduces spin correlation such that dark states \( |\psi^\pm_{\sigma\sigma} \rangle \) with \( \sigma = \sigma' \) are avoided. The electrons are rather being trapped in combinations of dark states \( |\psi^\pm_{\sigma\sigma} \rangle \) with \( \sigma \neq \sigma' \) and spin-blockaded states \( |\psi^\pm_{\sigma\sigma'} \rangle \). Thus, SB competes with coherent population trapping in the blocking of the current, and the relative occupation of \( |\psi^\pm_{\sigma\sigma} \rangle \) (\( \sigma \neq \sigma' \)), and \( |\psi^\pm_{\sigma\sigma'} \rangle \) depends on the initial condition. If however \( \Phi = \Phi_0/4 \), the A-B phase removes the dark state and only eigenstates with parallel spins are decoupled from \( |S_3 \rangle \):

\[
|\psi^\pm_{\sigma\sigma} \rangle = \frac{1}{\sqrt{2}}(|\chi_{2\sigma\sigma} \rangle \pm i|\chi_{\sigma\sigma} \rangle) \quad \sigma = \{ \uparrow, \downarrow \}
\]

Coherent trapping is hence lifted, however, transport is still cancelled by SB (Figs. 1, 2b). One can appreciate that without \( B_{\text{ac}} \), the system is always blocked for transport — the stationary current is insensitive to A-B effect due to SB.

Driven case \((B_{\text{ac}} \neq 0)\): — In order to remove SB, we apply a time-dependent \( B_{\text{ac}} \). Fig. 2 shows the \( I-\Phi \) characteristics of the TQD excited by \( B_{\text{ac}} \), where for every value of \( \Phi \), the magnetic field frequency fulfills the resonance condition \( \hbar \omega = \Delta_1 - 2 \). For \( \Phi/\Phi_0 = n/2 \), dark states are avoided by A-B effect, and \( B_{\text{ac}} \) enables transitions of the form \( |\chi_{\sigma\sigma} \rangle \rightarrow |\chi_{\sigma'\sigma} \rangle \rightarrow |S_3 \rangle \) that produce a finite current.

It can be shown that \( B_{\text{ac}} \) does not affect the destructive tunneling interference of the superpositions (2). Then, if \( \Phi/\Phi_0 = n/2 \) the system evolves towards a state which is only composed of dark states performing spin rotations: \( |\psi^-_{\sigma\sigma} \rangle \leftrightarrow |\psi^+_{\sigma\sigma} \rangle \), as shown schematically in Fig. 1. Since the dark states are decoupled from transport, the oscillations can only be affected by decoherence due to spin scattering processes, which are not considered here. Hence, a \( B_{\text{ac}} \) induces current through the system only
The quenching of the current can be understood analytically by transforming the Hamiltonian into the rotating frame. Applying the unitary operator $\hat{U}(t) = \exp\{-i\omega t \sum_{i=1}^3 \hat{S}_{i+}\}$, the magnetic field term reads: 
\[
\hat{H}_B = \sum_{i=1}^3 [(\Delta_i - \hbar \omega)\hat{S}_{i+} + B_{ac}\hat{S}_{i-}].
\]
One can easily verify that, at $\hbar \omega_0 = (\Delta + \Delta_3)/2$, the coherent superpositions
\[
|\Psi^\pm\rangle = \frac{1}{\sqrt{2}} \left( |\xi_{++}^\pm\rangle - |\xi_{+-}^\pm\rangle \right)
\]
are eigenstates of the Hamiltonian $\hat{H}' = \hat{H}_r + \hat{H}_B'$. Since the electrons in $\mathbf{3}$ have parallel spins, current is quenched due to SB. Note that the electron spins in $\mathbf{3}$ are maximally entangled. We want to emphasize that $\mathbf{SB}$ can be switched on and off by tuning the frequency of $B_{ac}$, which is usually introduced to lift it, or by changing the flux $\Phi$ at a fixed frequency $\omega$, see Fig. $\mathbf{3}$. 

In TQDs, a necessary condition for $\mathbf{3}$ to be eigenstates of $\hat{H}'$ and thus for the current blocking to occur, is the equal coupling of dots 1 and 2 to $B_{ac}$, i.e. $\Delta_1 = \Delta_2$ ($\neq \Delta_3$). If $\Delta_1 \neq \Delta_2$ though, this symmetry is broken and $\hat{H}_B'$ couples all parallel to antiparallel spin states and thus to the transport state $|S_3\rangle$. However, numerical results show that even in the asymmetric case, a pronounced antiresonance due to SB still appears in the current. By means of a perturbative analysis for $\Delta_1 - \Delta_2 \ll \tau$ it can be shown that the antiresonance occurs at a frequency $\omega_1 \approx \frac{\hbar}{2} (\Delta_1^2 + \Delta_3/2 + \Delta_3)$, see Fig. $\mathbf{4}$. The electrons drop into

when assisted by the A-B lifting of dark states — i.e. for $\Phi/\Phi_0 \neq n/2$ (Fig. $\mathbf{2}$).

Remarkably, not imposing the resonance condition $\hbar \omega = \Delta_i$, one can find a novel kind of SB induced by $B_{ac}$, quenching the current even in the presence of A-B effect. This is the main result of our work. As can be seen in Fig. $\mathbf{3}$, the current shows a resonant behaviour as the frequency of $B_{ac}$ approaches the ESR condition (i.e. $\hbar \omega \sim \Delta_1, \Delta_3$). Surprisingly though, an antiresonance appears for $\hbar \omega_0 = (\Delta_1 + \Delta_3)/2$, i.e. when the two electrons are equally far from the ESR condition. Note that the two peaks around the antiresonance are not Lorentzian-like and cannot be identified as two different resonance peaks centered at the conditions $\hbar \omega = \Delta_1 = \Delta_2$ and $\hbar \omega = \Delta_3$, but as a collective effect due to the simultaneous rotation of the two electron spins (2ESR), cf. Fig. $\mathbf{1}$.

We want to stress that the appearance of the antiresonance does not depend on the field intensity $B_{ac}$ or tunnel couplings $\tau_{ij}$ (see Fig. $\mathbf{3}$,a,b): it occurs for different $\tau_{ij}$ as well as for linear TQD configurations (setting $\tau_{13} = 0$) and DQDs in series (setting $\tau_{12} = \tau_{23} = 0$), see Fig. $\mathbf{3}$. The width of the antiresonance scales with the Rabi frequency of the coherent processes involved, spin rotation ($\propto B_{ac}$) and interdot tunneling ($\propto \tau_{ij}$) (Figs. $\mathbf{3b}$ and $\mathbf{4b}$, respectively); it also depends on the tunneling rate through the contact barriers, which induce decoherence, see Fig. $\mathbf{3}$.
tuning its frequencies to $\hbar \omega_1$ increases as $\Delta$ reduces. A small leakage current in states decouples from an eigenstate in (5) thereby blocking the current. 

![FIG. 4: $I-\omega$ in a TQD for $\Delta_1 \neq \Delta_2$. For $\tau \gg \Delta_1 - \Delta_2$, $I$ shows an antiresonance with a pronounced minimum at $\hbar \omega_1 = \frac{1}{2}(\Delta_1 + \Delta_2)$. Dashed-dotted line: $\Delta_2 - \Delta_1 = 0.0013$; dashed line: $\Delta_1 - \Delta_2 = 0.0013$; solid line: $\Delta_1 = \Delta_2$. Parameters: $\tau = \Gamma = 0.01$.](image)

![FIG. 5: $\rho_{\sigma}$ in a TQD excited by a bichromatic $B_{ac}$, where $\omega_1 = \Delta_{1,2}$ and $\omega_2 = \Delta_3$. In the stationary limit, parallel spin states decouple from $B_{ac}$ and form coherent superpositions as in (11) thereby blocking the current.](image)

an eigenstate $|\Psi^*\rangle$ which is similar to (11) but includes a small contribution of antiparallel spin states which produces a small leakage current. This leakage current increases as $\Delta_1 - \Delta_2$ becomes of the order of $\tau$.

**Bichromatic $B_{ac}$** — Finally, we will show that for $\Delta_1 = \Delta_2$ SB can also be induced by a bichromatic $B_{ac}$, tuning its frequencies to $\hbar \omega_1 = \Delta_{1,2}$ and $\hbar \omega_2 = \Delta_3$, so every electron is kept in resonance regardless of its location. Assuming that the inhomogeneity in the Zeeman splittings is high enough so one can neglect the off-resonance terms, the Hamiltonian $\hat{H}_B(t)$ can be written as

$$\hat{H}_{B,2} = \sum_{i=1}^{3} \Delta_i \hat{S}_{zi} + B_{ac} [\cos(\Delta_i t) \hat{S}_{zi} + \sin(\Delta_i t) \hat{S}_{yi}]$$

Again, by means of the unitary transformation $U(t) = \exp\{-i \sum \Delta_i \hat{S}_{zi} t\}$, the states (11) turn out to still be eigenstates of the transformed $\hat{H}_{B,2}$, even if the two spins are now rotating in resonance. The bichromatic $B_{ac}$ washes out all the states with antiparallel spins, driving the system into SB in spite of the Zeeman inhomogeneity, see Fig. 4.

**Conclusions** — In summary, we have shown theoretically that TQD systems in triangular configuration under dc and ac magnetic fields exhibit rich dynamics due to the interplay of different coherent phenomena induced by the magnetic fields. For two extra electrons in the system the interplay of Pauli exclusion principle and coherent trapping is discussed in terms of the magnetic flux piercing the TQD. We have shown that, in contrast to the one electron case, due to SB, electrons remain trapped even for $\Phi/\Phi_0 \neq n/2$. We demonstrate that a generic property of monochromatic and bichromatic magnetic fields is to induce SB at certain frequencies in both DQDs and TQDs. Furthermore, the coherent superposition induced by the $B_{ac}$ constitutes a novel SB state and is decoupled from the field. Its experimental realization will allow one to infer properties of the system such as Zeeman inhomogeneities, and to manipulate spin qubits in DQDs and TQDs. It opens new perspectives for manipulating spin transport properties, thereby providing possibilities for designing spintronic devices.

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