Spin resonance without spin splitting

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We predict that an excitationless Coulomb-blockaded quantum dot develops a nonequilibrium spin precession resonance when embedded into a spin valve. At resonance the spin-valve effect is mitigated. The resonance can be detected by stationary $dI/dV_b$ spectroscopy and by oscillations in the time-averaged current using a gate-pulsing scheme. The generic noncollinearity of the ferromagnets and junction asymmetry allows for an all-electric determination of spin injection, anisotropy of spin relaxation, and exchange field. We investigate the impact of a nearby superconductor on the resonance position.

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Gaining fast, coherent control over a few spins or even a single spin is at the heart of current experimental efforts in both spintronics and solid-state quantum computing. Single-molecule magnets in gateable nanojunctions, or adatoms and molecules manipulated by STM, provide a bottom-up approach to achieve this goal. Promising top-down routes combine conventional spin valves with nanoscale quantum dot (QD) devices. Here, due to the strong Coulomb repulsion of electrons confined on the nanometer scale, single electron spins can be sufficiently isolated from their environment to address them individually, e.g., through electromagnetic pulses. This requires that the frequency of the applied pulses matches the splitting of the QD spin levels. Such splittings can also be detected in transport experiments. For instance, single-electron tunneling and inelastic cotunneling resonances in a $dI/dV_b$ stability diagram resolve orbital and spin splittings. The tunnel coupling, besides enabling spectroscopy, also results in renormalization effects. Here we focus on a prominent example, the interaction-induced exchange field, which arises from spin-dependent fluctuations of QD electrons into the attached ferromagnets, probing their entire band structure. It has been observed for strong tunnel coupling $\Gamma$ as an induced level splitting in inelastic tunneling spectroscopy. Similar level renormalizations appear in systems with orbitally or vibrationally (quasi)degenerate states, also in combination with the spin exchange field.

In this Letter we predict a pronounced spin resonance that does not involve a matching to a spin splitting but relies on a coherent precession of a single spin. It gives rise to an anomalous, strongly gate-voltage dependent feature in the stationary $dI/dV_b$ stability diagram of a noncollinear QD spin-valve, extending all across the Coulomb blockade regime. It arises for nonequilibrium conditions but disappears upon reversing the bias voltage. Strikingly, it can appear at voltages much larger than any of the naively expected energy scales. Though caused by the exchange-field spin splitting, the sharp resonance appears for moderate tunnel coupling when this splitting can not be resolved. All these features distinguish this resonance from other effects in the Coulomb blockade regime including those due to inelastic cotunneling which require resolvable excitations. In previous works, measurable consequences of such a precession (or similar coherence effects) have been predicted for the stationary conductance, the noise spectrum, and also for hybrid setups with a superconductor. However, most of these works use only the electric control of the magnitude of the exchange field. On the contrary, the above-mentioned feature relies on the electrical tuning of the exchange field direction for fixed, nearly antiparallel magnetizations of the ferromagnets. For this generic experimental configuration beyond simplifications frequently made in theory – the exchange field can point perpendicular to the spin polarization of the electrons injected into the QD, engendering a spin precession that lifts the spin-valve effect. This might be a serious problem for future low-power spin-valve devices operated in the Coulomb blockade regime. The resonance mechanism is effective only in the Coulomb blockade regime since there the leading-order spin-decoherence rate is exponentially suppressed with the distance of the level position to the Fermi levels, and therefore dominated by the much smaller cotunneling contribution $\propto \Gamma^2/U$. By contrast, the spin precession period is still dominated by the leading-order exchange field $\propto \Gamma$, as noted earlier. Thus, the Coulomb interaction does not only induce the precession, but it can also make the precession underdamped. Direct evidence of this underdamped precession, however, requires a time-dependent detection, addressed by the final result of this Letter: We propose a pulsing detection scheme that probes multiple spin revolutions by a time-averaged...
current measurement. It provides electrical control of the rotation axis over an angle $\pi$ as well as the rotation angle. Hence, a simple QD spin valve already has built-in capabilities for single-spin operations.

Model. The simplest model of an excitationless, interacting QD is the single-level Anderson model [cf. Fig. 1(a)], $H = \sum_{\sigma} \epsilon n_{\sigma} + U n_{\uparrow} n_{\downarrow}$. Here $n_{\sigma} = d_{\sigma}^\dagger d_{\sigma}$ is the electron number operator for spin $\sigma = \uparrow, \downarrow$ and $U$ is the interaction energy of the electrons. The level is spin degenerate and its position $\epsilon = -V_g$ is electrically controllable by the gate voltage $V_g$ ($e = h = k_B = 1$).

The QD is tunnel-coupled to two ferromagnetic leads $H_T = \sum_{rks} t_{rs} c_{rks}^\dagger c_{skr}$, incorporating the tunneling amplitude $t$, which sets the tunneling rates $\Gamma_r = \pi |t|^2 (\nu_{r\uparrow} + \nu_{r\downarrow})$. We also study an extension of the above model by adding a superconducting terminal at electrochemical potential $\mu_{\text{sup}} = 0$, tunnel-coupled with rate $\Gamma_{\text{sup}}$, as sketched in Fig. 1(b). In the limit of infinite superconducting gap, $\Delta \to \infty$, the effect of the superconductor can be incorporated by adding a pairing term $-\Gamma_{\text{sup}}(d_{\uparrow}^\dagger d_{\downarrow} + d_{\downarrow}^\dagger d_{\uparrow})/2$ to the QD Hamiltonian.

Nonequilibrium spin dynamics. We first focus on the setup sketched in Fig. 1(a). The transport signatures of the QD spin valve are governed by the nonequilibrium dynamics on the QD, described by its reduced density matrix $\rho$. The reduced density matrix is characterized by the occupation probabilities $p_n$ for each of the charge states $n = 0, 1, 2$, and the average spin vector $\vec{S} = \text{tr} (\hat{S} \rho)$ with $\hat{S} = \sum_{\sigma \sigma'} \frac{1}{2} \sigma \sigma' \sigma_{\sigma'}^\dagger \sigma_{\sigma'}$ and $\sigma$ denoting the vector of Pauli matrices. For $U \to \infty$ the kinetic equations for these dynamical variables read in the Markovian limit

$$
\begin{align*}
\dot{p}_1 &= 2 \Gamma_0 p_0 - \Gamma_1 p_1 + \vec{A}_S \cdot \vec{S}, \\
\dot{\vec{S}} &= \vec{A}_0 p_0 + \vec{A}_1 p_1 - \vec{R}_S \cdot \vec{S} - \vec{B} \times \vec{S},
\end{align*}
$$

with $p_0 = -\dot{p}_1$ due to probability conservation: $p_0 + p_1 = 1$ and $p_2 = 0$. The rates in the above equation, as well as the current flowing through the QD, are computed including all leading $O(1)$ and next-to-leading $O(T^2)$ terms. In the Suppl. Material we give analytic expressions and comment on how to solve Eq. (1). The latter extends the master equation for the occupation probabilities $p_n$ by an intense coupling to the coherence of the (quasi)degenerate spin states (contained in the spin vector $\vec{S}$). This leads to a significant modification of the charge dynamics, detectable in transport.

Spin-precession resonance. A main result of this Letter is presented in Fig. 1(c). It shows the stationary conductance obtained by numerically solving the generalization of Eq. (1) to finite $U$ for the stationary state. Close to the antiparallel magnetic configuration, we find a strikingly sharp wiggle in $dI/dV_b$, i.e., a peak in the current plotted vs. $V_b$, which extends through all the Coulomb-blockade regime. Notably, the resonance starts at the Coulomb-diamond edges, then bends towards the particle-hole symmetry point at $V_b = 0$, $\varepsilon = -U/2$, which opens a strikingly sharp window in the conductance. This leads to a significant modification of the charge dynamics, detectable in transport.
where its magnitude vanishes. It then continues point-symmetrically with respect to this point. We therefore focus our discussion on the $V_g < U/2$ part of Fig. 1(c). We chose the labels “source” and “drain” such that the resonance appears in this case at $V_g > 0$. To understand the origin of this resonance, we first note that the current through the QD is largely suppressed for nearly antiparallel polarizations by the spin-valve effect: Source electrons with majority spin along $\hat{n}_s$ can tunnel into the QD with a large rate $\approx \Gamma_s(1+n_s)$, while they leave to the drain with a very small rate $\sim \Gamma_d(1-n_s)$ as their spin is of the drain minority type ($\hat{n}_d \approx -\hat{n}_s$). However, if the polarizations of the electrodes are merely slightly non-collinear, the spin-precession resonance appears in Fig. 1(c). The reason is that the drain contribution to the exchange field, $\vec{B} = B_s\hat{n}_s + B_d\hat{n}_d$, adds a component $B_{d,\perp} = B_d\sin\alpha$ that is perpendicular to the source polarization $\hat{n}_s$, i.e., $\vec{B} = (B_s + B_{d,\|})\hat{n}_s + B_{d,\perp}\hat{n}_s,\perp$, with $B_{d,\|} = B_d\cos\alpha$. The seemingly innocuous component $B_{d,\perp}$ causes a precession of the spin along $\hat{n}_s$ of an electron injected from the source towards $\hat{n}_d$. Consequently, the electron can easily leave the QD to the drain. This prevents a spin accumulation from building up antiparallelly to the drain – the spin-valve effect is mitigated. Yet, this rotation is effective only if the opening angle of the spin precession is large [cf. Fig. 2(c)(ii)]. Hence, the resonance appears when the total exchange field component parallel to the source polarization $\hat{n}_s$ vanishes:

$$\vec{B} \cdot \hat{n}_s = B_s + B_{d,\|} = 0.$$  

The resonance position can be predicted from the $O(\Gamma)$ approximation for the exchange field $\vec{B}$

$$B_r = \sum_{k=0,2} n_k \Gamma_k \Re \int_{-W}^{+W} d\omega \frac{\epsilon}{\pi \epsilon + kU/2} \left(f_s(\omega) - f_d(\omega)\right) \left(1 + \frac{e^{\epsilon - \mu_c}}{T + 1}\right),$$

where $f_s(\omega) = 1/(e^{\epsilon + \mu_c}/T + 1)$. It comprises a contribution from particle ($k = 0$) and hole ($k = 2$) processes. Inserting Eq. (3) into the resonance condition (2) and solving for the resonant bias $V_g^{\ast}$ as a function of $V_g$ yields the white dashed curve in Fig. 1(c). (The reason is that the drain contribution to the precession appears when the total exchange field component parallel to $\hat{n}_s$ vanishes: $\vec{B} \cdot \hat{n}_s = B_s + B_{d,\|} = 0$.)

Remarkably, for a given gate voltage $V_g$, the condition (2) is fulfilled only for one bias direction – a clear feature to rule out other effects in experimental data. This strong rectification of the current can be attributed to the electrical tunability of the exchange field direction. In Fig. 2(a), we plot $B_s$, $B_{d,\|}$ and their sum (4) as function of the bias $V_g$. For electrode $r$ the magnitude $B_r$ is maximal when the electrochemical potential matches the addition energies, i.e., $\mu_r = \epsilon$, or $\mu_r = \epsilon + U$. Moreover, each contribution $B_r$ vanishes at $\mu_r = \epsilon + U/2$ [as marked in Fig. 2(b) by (i), (iii)]. In the vicinity of the cancellation points the full exchange field $\vec{B}$ is thus given by the contribution from only one electrode, that is, it points along $\hat{n}_s$ [see Fig. 2(c)(i)] or $\hat{n}_d$ [see Fig. 2(c)(iii)], respectively. Here, the spin precesses with a small opening angle and the spin transport stays blocked. However, when tuning the bias between these two cancellation points, the exchange field rotates and the sum $B_s + B_{d,\|}$ vanishes for a specific bias $V_g^{\ast}$. This results in a spin precession with a large opening angle $\sim \pi/2$ [see Fig. 2(c)(ii)], effectively lifting the spin-valve effect producing the spin-precession resonance in Fig. 1(c). Notably, the resonant bias $V_g^{\ast}$ takes a nonzero value if the spin injection rates are asymmetric, $a := (\Gamma_s n_s)/[\Gamma_d n_d \cos\alpha] \neq 1$, since the cancellation points lie symmetrically at $V_g = \pm 2(\epsilon + U/2)$, but the slopes of $B_s$ and $B_{d,\|}$ are different, as sketched in Fig. 2(b). If the magnitude of the exchange field is much larger than the spin relaxation rate, the resonant current $I_0$ is restored to the value obtained when ignoring spin accumulation, c.f. also Fig. 2(b). Thus, Eq. (2) pinpoints where the spin precession completely ruins the operation of a QD spin valve already for small non-collinearity angles.

Characterization of (hybrid) QD spin-valves. As Fig. 1(c) clearly shows, the bias scale $V_g^{\ast}$ does not match any obvious energy scale of the problem attesting to its non-spectral origin. Depending on the gate voltage, it may exceed $\Gamma$, $T$, and even approach a sizable fraction of $U$. This relates to the distinct nature of the resonance mechanism as compared to other types of resonances. The effect may be utilized to characterize QD spin valves in an alternative way: As we show in the Suppl. Material, one can (1) extract the ratio $a$ from an experimentally measured stability diagram using the slope of the resonance near the particle-hole symmetry point. Such an in-situ characterization is preferable since transport characteristics expected from bulk properties of the leads can be considerably altered due to the nanoscale contacting of the QD. Furthermore, if the asymmetries of the tunnel-
(a) Schematics of the pulsing scheme. (b) Stationary current as function of $V_g$, obtained by solving Eq. (1) exactly ($l_{st}$, green), and by neglecting the spin accumulation, i.e., forcing $\vec{S} = 0$ ($l_0$, dashed black), as well as taking a Lorentzian approximation near resonance ($l_{st}$, red). (c)–(e) Average current $\bar{I}$ (green curves) obtained as a function of $\tau$ for three different $V_g$ indicated in (b) and fixed $\tau^0 = 2 \cdot 10^3 \cdot T / \Gamma = 0.46 T$ and $\tau^0 = 30T$. The times $\tau^0$ and $\tau$ are given in units of the precession period at resonance, $\tau_p \approx 4.7 \cdot 10^3 / T$. The current is offset by $l_{st}$, the current that would flow if the QD were in the stationary state at each instant of time. Also plotted is the spin component along the drain polarization $\vec{S} \cdot \vec{n}_d$ (blue curves) computed from Eq. (1) for initial condition $\vec{S} = \vec{n}_s / 2$ and $p_1 = 1 - p_0 = 1$. Throughout we used $n_s = n_d = 0.09$ (see remark in caption of Fig. (1), $\alpha = 0.005\pi$, $V_b = 50T$, $D = 500T$, $\Gamma_s = 0.15T$, $\Gamma_d = 0.17T$. To make use of analytical results, we take $U \to \infty$, requiring a tiny angle $\alpha$ here. For finite $U$, this restriction is unnecessary.

FIG. 3: (a) Schematics of the pulsing scheme. (b) Stationary current as function of $V_g$, obtained by solving Eq. (1) exactly ($l_{st}$, green), and by neglecting the spin accumulation, i.e., forcing $\vec{S} = 0$ ($l_0$, dashed black), as well as taking a Lorentzian approximation near resonance ($l_{st}$, red). (c)–(e) Average current $\bar{I}$ (green curves) obtained as a function of $\tau$ for three different $V_g$ indicated in (b) and fixed $\tau^0 = 2 \cdot 10^3 \cdot T / \Gamma = 0.46 T$ and $\tau^0 = 30T$. The times $\tau^0$ and $\tau$ are given in units of the precession period at resonance, $\tau_p \approx 4.7 \cdot 10^3 / T$. The current is offset by $l_{st}$, the current that would flow if the QD were in the stationary state at each instant of time. Also plotted is the spin component along the drain polarization $\vec{S} \cdot \vec{n}_d$ (blue curves) computed from Eq. (1) for initial condition $\vec{S} = \vec{n}_s / 2$ and $p_1 = 1 - p_0 = 1$. Throughout we used $n_s = n_d = 0.09$ (see remark in caption of Fig. (1), $\alpha = 0.005\pi$, $V_b = 50T$, $D = 500T$, $\Gamma_s = 0.15T$, $\Gamma_d = 0.17T$. To make use of analytical results, we take $U \to \infty$, requiring a tiny angle $\alpha$ here. For finite $U$, this restriction is unnecessary.

Pulsing detection scheme: Probing underdamped spin precession. We estimate that the transport-induced spin decoherence time $\sim U / T^2$ can be made comparable or longer than experimentally measured spin dephasing times due to other mechanisms. Hence, multiple revolutions of an individual electron spin while dwelling in the QD are feasible. Probing this underdamped spin precession requires time-resolved measurements. Even though there is no discernable spin splitting, an experimentally well-developed pulsing scheme can be applied here. As sketched in Fig. 3(a), one repeatedly applies a rectangular voltage pulse to the gate electrode, switching from $V_b$ to $V_g$ for a time duration $\tau$, and then back to $V_b$ for a time duration $\tau^0$. By measuring the time-averaged current over many pulses, $\bar{I} = \int_0^\tau dt / I(t)$, we can extract the magnitude of the exchange field $|\vec{B}|$ at $(V_b, V_g)$. Figs. 3(c)–(e) show that the time-averaged current obtained in this way oscillates as a function of $\tau$ with a period given by $2\pi / |\vec{B}|$, which coincides with the period of the spin oscillations plotted alongside the current. The current oscillations can be understood as follows. The scheme has been set up such that at $V_b$ only a small current flows through the QD; initially the transport to the drain is blocked due to the spin-valve effect [Fig. 3(a)(i)]. If the gate voltage $V_g$ after switching is near the resonance, the exchange field $\vec{B}$ is quickly rotated, and the injected spin can subsequently precess [Fig. 3(a)(ii)], but not leave due to stronger Coulomb blockade at $V_g$. When tuning back to $V_b$, it can thus leave to the drain with increased probability [Fig. 3(a)(iii)]. The resulting enhancement of the current first depends on the rotation angle of the spin, controlled by the duration $\tau$ [cf. Fig. 3(c)–(e)]. Second, the enhancement depends on the opening angle of the precession, controlled by $V_g$, as the series of Figs. 3(c)–(e) show. Most pronounced oscillations occur when $V_g$ is tuned to the resonance, determined by Eq. (2), where the exchange field is perpendicular to the injected spin.

Conclusion. We have identified a spin resonance that is quite distinct from usual types of resonances: It does not appear when scalar energies of the local system and reservoir match, but rather requires a vector condition, Eq. (2), to be satisfied. The nontrivial voltage dependence of the resonance derives from the drastic changes in the direction of the exchange field vector $\vec{B}$, rather than its magnitude, allowing a real-time detection by a gate-pulsing scheme. Although the spin resonance is not tied to an energetic resonance with a spin splitting, it can still be exploited to extract the ratio of the spin injection rates, the exchange field magnitude – even when modified by Andreev reflection processes – and the anisotropy of the spin relaxation rates. The resonance signals a substantial loss of magnetoresistance already for tiny noncollinearities, issuing a warning against simplifications regarding this important parameter. Finally, this type of resonance is not tied to the spintronic problem studied here; it could appear in a broad class of systems whenever states from a discrete spectrum are (quasi)degenerate, such as in double QDs or vibrating molecular devices.

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Typical dephasing times of spins measured in GaAs range from $\tau_d \sim 10^{-30}$ ns. In our case, the cotunneling current through the QD leads to additional dephasing, with time constant $\sim U/\Gamma^2 \approx 30$ ns for typical values of $\Gamma \sim 10 \mu$eV, and $U \sim 5$ meV for semiconductor QDs. The exchange field may be on the order of $B > B_{d,\perp} \approx |\log(1/2)| \Gamma g n_d \sin \alpha/\pi \sim 1 \mu$eV for $n_d \sim 0.5$ and $\alpha \sim 0.2\pi$. This translates into a maximal period of $2\pi/B_{d,\perp} \approx 6$ ns at the resonance and even smaller periods away from it.

Although in spin-splitting transport measurements\cite{Fujisawa2003, Donarini2006} the precession is underdamped as well, a real-time detection becomes more challenging: There, due to the required strong coupling and correspondingly large exchange fields, precession periods are only $\sim 100$ ps. Here, due to the moderate coupling, the period can be much longer.