Coulomb Promotion of Spin-Dependent Tunnelling

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We study transport of spin-polarized electrons through a magnetic single-electron transistor (SET) in the presence of an external magnetic field. Assuming the SET to have a nanometer size central island with a single electron level we find that the interplay on the island between coherent spin-flip dynamics and Coulomb interactions can make the Coulomb correlations promote rather than suppress the current through the device. We find the criteria for this new phenomenon — Coulomb promotion of spin-dependent tunnelling — to occur.

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Strong Coulomb correlations have important consequences for electronic transport on the nanometer length scale. Coulomb blockade (CB) of single electron tunnelling is, e.g., the fundamental physical phenomenon behind single electron transistor (SET) devices. The electron spin comes into play if the source and drain electrodes in the SET structure are made of magnetic material, allowing for the electrons that carry the current to be spin polarized. Materials with nearly 100 % spin polarization are now under intensive study and experiments have established that transport of spin-polarized electrons is sensitive to the relative orientation of the magnetization of the source and drain leads. This opens up for a spin-valve effect, where an external magnetic field can be used to control the current. Switching the magnetization in one lead with respect to the other is one way of achieving such a control. Another approach — to be pursued here — is to flip the spin of electrons carrying current from one magnetic lead to another via a central non-magnetic island (“Coulomb dot”) in a SET structure, keeping the lead polarizations fixed.

A giant magneto-conductance effect was suggested recently for magnetic “shuttle” devices, where spin-polarized electrons localized on a movable quantum dot are mechanically transported through a region, where the electronic spin dynamics is totally controlled by an external magnetic field. This suggestion brings into focus the very important question of the role of the spin dynamics in resonant electron tunnelling, where electrons can be trapped in a resonant state — e.g. on a Coulomb dot — for quite a long time. Since the resonant electron level of the dot may be doubly occupied by electrons with different spin, Coulomb blockade of single electron tunnelling should have an important influence on the spin-dependent resonant charge transfer. Understanding how these two effects combine is of general fundamental interest in the context of magnetic nanostructures.

In this Letter we will consider electronic transport through the simple magnetic SET device shown in Fig. 1.

Here a Coulomb dot, subject to an external magnetic field, is located between two spin-polarized leads. The external magnetic field is oriented perpendicular to the polarization in the leads and is taken to be small enough not to affect the magnetization of the leads. Assuming the dot is of nanometer size we consider only one electron energy level on the dot. This level may, however, accommodate two electrons of different spin orientation and the strength of the Coulomb interaction between them significantly affects the charge transfer through the device. The magnetic field in its turn, by inducing coherent spin-flip dynamics on the dot, actually promotes electronic transport if the magnetization in the leads point in opposite directions. This effect is most conspicuous when the leads are fully spin-polarized. In this case an electron can tunnel from one lead to the other only if its spin flips while the electron resides on the dot (without magnetic field the current is completely blocked by a “spin-blockade”). We have found, that Coulomb correlations on the dot, if they are strong enough to prevent a double occupancy of the resonant level, significantly stimulates such spin-flip processes and hence promote spin-
dependent electronic tunnelling.

To understand this phenomenon qualitatively let us consider a simplified set-up: a nonmagnetic quantum dot having a single electronic level with energy $\epsilon$ is linked only to the left, fully spin-polarized (up, say) metallic lead with electrochemical potential $\mu_L$. Let $\mu_L - \epsilon$ be much larger than the width $\Gamma_L$ of the dot level. Under such conditions the spin-up state on the dot will be fully occupied, while the spin-down state will be completely empty. If now a magnetic field $B \ll \Gamma_L/\mu$ (the Bohr magneton) oriented perpendicular to the lead magnetization is switched on at time $t = 0$, spin-flip processes that populate the spin-down state on the dot will be induced. The characteristic time $\tau_{sf}$ for populating the spin-down state is an important quantity. It turns out that $\tau_{sf}$ strongly depends on whether or not Coulomb interactions prevent a second electron from tunnelling onto the dot during the spin-flip process.

To see this, let us first consider the Coulomb blockade regime (CB-regime), where the energy difference $\mu_L - \epsilon$ is smaller than the Coulomb interaction energy $U/2$ between two dot electrons. The tunnelling of a second electron onto the dot is then blocked and the population of the spin-down state is simply controlled by the coherent spin dynamics of the one electron already there. The probability amplitude $A_{sf}$ for a spin-flip transition increases linearly with time, $A_{sf} = t/\tau_h$ (here $\tau_h \equiv \hbar/\mu h$), and the probability $\rho_{\downarrow}$ to find the electron in the spin-down state is

$$\rho_{\downarrow} (t) = |A_{sf}|^2 = (t/\tau_h)^2. \quad (1)$$

The spin-flip time $\tau_{sf}$ may be estimated from the condition $\rho_{\downarrow}(\tau_{sf}) = 1$. Hence in the CB-regime $\tau_{sf} \equiv \tau_c \simeq \tau_h$.

If on the other hand $\mu_L - \epsilon > U$, we are in the “free” regime where the Coulomb blockade is lifted and a second (spin-up) electron can tunnel onto the dot if there is a finite probability for its spin-up state to be unoccupied. This process couples the electronic state on the dot to a large number of states in the lead and breaks the coherence of any ongoing evolution of the on-dot spin state after a time $\Delta t \simeq \hbar/\Gamma_L$. The probability for a spin flip to occur during this time is $P_{sf}(\Delta t) \equiv |A_{sf}(t = \Delta t)|^2 = (\Delta t/\tau_h)^2$. For longer times $t$ the probability for a spin-flip to occur in $t/\Delta t$ coherent time intervals add incoherently. Therefore, if the Coulomb blockade is lifted, the probability to find the dot electron in the spin-down state can be written as

$$\rho_{\downarrow}(t) \simeq (t/\Delta t)P_{sf}(\Delta t) = (t/\Delta t)/\tau_h^2. \quad (2)$$

and the spin-flip time $\tau_f \simeq \tau_c (\hbar/\Delta t)$. It follows that $\tau_c \ll \tau_f$ in a weak magnetic field ($h \ll \Gamma_L/\mu$). Hence, if the tunnelling of a second electron is blocked, the probability for the spin of the electron already on the dot to flip is strongly enhanced.

Next we extend our qualitative discussion to the tunnelling of electrons through the entire SET device. For this purpose we switch on the coupling between the spin-down dot state and states in the right lead held at chemical potential $\mu_R$. If $\mu_R - \epsilon \simeq -\Gamma_R$, where $\Gamma_R/\hbar$ is the tunnelling rate between dot and right lead, the spin-down dot electron can tunnel to an empty state in the right lead. The resulting current through the SET is given by the product of the probability to find the dot electron in the spin-down state and the tunnelling rate $\Gamma_R/\hbar$. If the spin flip rate is much smaller than $\Gamma_R/\hbar$, the population of the spin-down state can be estimated as $\rho_{\downarrow}(t = \hbar/\Gamma_R)$. Furthermore, since electron exchange with the right lead also restricts the coherent spin-flip time one has to put $\Delta t \min\{\hbar/\Gamma_L, \hbar/\Gamma_R\}$ in Eq. (2). Therefore, in a strongly asymmetric situation, when the ratio $\Gamma_R/\Gamma_L$ is very different from one, the current through the SET is given by the expression

$$I = \frac{e}{\hbar} \frac{(\mu h)^2}{\Gamma_R} \bigg\{ \begin{array}{ll} 1 & \text{CB regime (c)} \\ \min\{1, \Gamma_R/\Gamma_L\} & \text{free regime (f)} \end{array} \bigg\} \quad (3)$$

One concludes that, when $\Gamma_R/\Gamma_L \ll 1$, the current in the CB-regime is larger by a factor $\Gamma_L/\Gamma_R$ than in the “free” regime, where the CB is lifted. We will refer to this as “Coulomb promotion” of spin-dependent tunnelling. From Eq. (3) it also follows that the current-voltage curve is strongly asymmetric in the CB-regime; the current changes by a factor $\Gamma_R/\Gamma_L$ if the sign of the bias voltage is reversed.

To provide a quantitative description of the Coulomb promotion phenomenon discussed above we consider a SET structure with magnetic leads polarized along the same z-direction, and a central island (dot) with a single electron energy level subject to an external magnetic field $\vec{h}$. The Hamiltonian $\hat{H}$ for our system is

$$\hat{H} = \hat{H}_l + \hat{H}_d + \hat{H}_r,$$

$$\hat{H}_l = \sum_{\alpha, \sigma, \kappa} \varepsilon_{\alpha, \sigma, \kappa} \hat{a}^\dagger_{\alpha, \sigma, \kappa} \hat{a}_{\alpha, \sigma, \kappa},$$

$$\hat{H}_d = \sum_{\sigma} \omega \hat{a}_\sigma^\dagger \hat{a}_\sigma - \frac{U}{2} \hat{a}_\uparrow^\dagger \hat{a}_\uparrow \hat{a}_\downarrow \hat{a}_\downarrow - \mu \sum_{i, \sigma, \sigma'} h_i \hat{a}_i^\dagger \sigma \hat{a}_i^\sigma \sigma',$$

$$\hat{H}_T = \sum_{\alpha, \sigma, \kappa} \kappa \sum_{\alpha, \sigma, \kappa} \omega a_\alpha^\dagger a_\sigma + \text{H.c.}, \quad (4)$$

and has several terms. The first term describes non-interacting electrons in the leads. Here $a_\alpha^\dagger_{\alpha, \sigma, \kappa}$ $(\hat{a}_{\alpha, \sigma, \kappa})$ is the creation (annihilation) operator for electrons in lead $\kappa$ with energy $\varepsilon_{\alpha, \sigma, \kappa}$ and spin projection $\sigma \in \{\uparrow, \downarrow\}$. The electron density of states $\rho_{\sigma}^\kappa$ in each lead is assumed to be independent of energy but strongly dependent on spin direction. The electrons in each lead are held at a constant electrochemical potential $\mu_{\sigma, \kappa} = \varepsilon_F \mp eV/2$, where $e$ is the charge of the electron, $V > 0$ is the bias voltage, and $\varepsilon_F$ is the Fermi energy of the ferromagnetic metal. The second term describes electronic states in the dot, their coupling to the external magnetic field $\vec{h} = (h_x, 0, h_z)$ and intra-dot electron correlations characterized by the
Coulomb energy $U$; the operator $a_\sigma^\dagger$ ($a_\sigma$) creates (destroys) an electron with spin $\sigma$ and $\Gamma^{\sigma\sigma'}_\kappa$ are Pauli matrices ($i = x, y, z$). The last term represents spin-conserving tunnelling of electrons between dot and leads.

We focus on the Coulomb promotion phenomenon discussed qualitatively above and assume the Coulomb energy $U$ to be much larger than both the thermal energy $kT$ and the width and Zeeman splitting of the dot level. We furthermore take the energy level on the dot to coincide with the Fermi energy of the leads, $\epsilon = E_F$, and consider the current for bias voltages $|V| > (|\mu|, \Gamma^\sigma_\kappa, kT)$ and $|V - U| > (|\mu|, \Gamma^\sigma_\kappa, kT)$ (here $\Gamma^\sigma_\kappa = 2\pi n^\sigma_\kappa T^2$ is the spin-dependent level width associated with tunnelling to the lead $\kappa$). If the external magnetic field is oriented along the polarization axis, charge transfer between the leads conserve spin projection and may be described within the "orthodox" CB theory [1]. A magnetic field oriented perpendicular to the polarization axis, on the other hand, generates coherent spin dynamics and makes the spin degree of freedom relevant for the electron transport problem.

The coupled processes of charge transfer and coherent spin dynamics is governed by a quantum Master equation for the corresponding reduced density operator $\hat{\rho}(t)$. It can be derived from the Liouville-von Neumann equation for the total system by projecting out the degrees of freedom associated with the leads $\{1\}, \{0\}, \{1\}$ [2]. The reduced density operator $\hat{\rho}(t)$ obtained in this way acts on the Fock space of the quantum dot which is spanned by the four basis vectors $|0\rangle$, $|\uparrow\rangle \equiv a_\uparrow^\dagger |0\rangle$, $|\downarrow\rangle \equiv a_\downarrow^\dagger |0\rangle$, and $|2\rangle \equiv a_\uparrow^\dagger a_\downarrow^\dagger |0\rangle$. In this basis the operator $\hat{\rho}(t)$ can be written as a $4 \times 4$ matrix. The diagonal elements $\rho_0 = \langle 0|\hat{\rho}(t)|0\rangle$ and $\rho_2 = \langle 2|\hat{\rho}(t)|2\rangle$ represent the probabilities for the dot to be unoccupied and doubly occupied, respectively. The singly occupied dot is described by the $2 \times 2$ matrix block $\hat{\rho}_1 \equiv \langle \sigma|\hat{\rho}|\sigma\rangle$

For bias voltages satisfying the given conditions, the time evolution of the probabilities $\rho_0$, $\rho_2$ and of the density matrix $\hat{\rho}_1$ is determined by the system of equations

$$\dot{\rho}_0 = -\text{Tr}(\hat{\Gamma}_L)\rho_0 + \text{Tr}(\hat{\Gamma}_R\hat{\rho}_1)$$
$$\dot{\rho}_2 = -\text{Tr}(\hat{\Gamma}_R + \theta(U - V)\hat{\Gamma}_L)\rho_2 + \theta(V - U)\text{Tr}(\hat{\Gamma}_L\hat{\rho}_1)$$
$$\dot{\hat{\rho}}_1 = i\mu[\hat{h}_\uparrow \hat{\tau}_1, \hat{\rho}_1] - \frac{1}{2}(\hat{\Gamma}_R, \hat{\rho}_1) - \frac{1}{2}\theta(V - U)(\hat{\Gamma}_L, \hat{\rho}_1)
+ \hat{\Gamma}_L\rho_0 + \theta(U - V)\hat{\Gamma}_L \rho_2,$$  \hspace{1cm} (5)

where $\hat{\Gamma}_\kappa(\Gamma^\uparrow_\kappa + \Gamma^\downarrow_\kappa)\hat{I}/2 + (\Gamma^\uparrow_\kappa - \Gamma^\downarrow_\kappa)\hat{\tau}_z/2$, $\hat{\tau}_1$ are the Pauli matrices, and we have set $e = \hbar = 1$.

The stationary solution of Eq. (5), and hence the stationary current, significantly depends on the relation between the bias voltage $V$ and the CB energy $U$. In particular, the third equation tells us that the CB significantly decreases the relaxation of the non diagonal elements of $\hat{\rho}_1$. As these describe the coherent spin flip dynamics, the spin-flip process turns out to be faster in the CB-regime than in the "free" regime without Coulomb blockade.

Within our approximations the current $I$ through the SET can be calculated from the formula

$$I = \text{Tr}(\hat{\Gamma}(\rho_1 + \rho_2 \hat{I})).$$  \hspace{1cm} (6)

It is convenient to think of the current as the sum of a background current $I^{(0)} = I(h = 0)$ and a magnetic-field promoted (MFP) current $J(h) = I - I^{(0)}$. Substituting the stationary solution of Eq. (5) into Eq. (6), one finds

$$I_l = I_l^{(0)} + J_l(h) = I_l^{(0)} + J_l \frac{h^2_z}{h^2_1 + h^2_2},$$  \hspace{1cm} (7)

where the label $l = c, f$ indicates whether we are in the CB-regime ($V < U$) or in the "free" regime where the blockade has been lifted ($V > U$). The quantities $I_l^{(0)}$, $J_l$, and $h_l$ do not depend on the transverse magnetic field and are given by

$$I^{(0)}_l = \frac{\Gamma_L \Gamma_R}{\Gamma_L + \Gamma_R} - J_c, \quad J_c = \frac{\Gamma_L (\Gamma^\uparrow_L \Gamma^\downarrow_R - \Gamma^\uparrow_R \Gamma^\downarrow_L)}{(\Gamma_L + \Gamma_R)(\Gamma^\uparrow_L + \Gamma^\downarrow_L)},$$
$$J^f_l = \frac{\Gamma_L \Gamma_R}{\Gamma} - J_f, \quad J_f = \frac{(\Gamma^\uparrow_L \Gamma^\downarrow_R - \Gamma^\uparrow_R \Gamma^\downarrow_L)}{\Gamma^\uparrow_L + \Gamma^\downarrow_L},$$
$$h^2_\kappa = \mu^2 (\Gamma^\uparrow_\kappa + \Gamma^\downarrow_\kappa)^2 \left[ \frac{1}{4} + \frac{(\mu h_z)^2}{(\Gamma^\uparrow_\kappa - \mu h_z)^2} \right].$$  \hspace{1cm} (8)

Here $\Gamma_\kappa = \sum_\sigma \Gamma^\sigma_\kappa$, $\Gamma^\sigma_\kappa = \sum_\kappa \Gamma^\sigma_\kappa$, and $\Gamma = \sum_{\sigma, \kappa} \Gamma^\sigma_\kappa$.

**FIG. 2:** Normalized current $I$ through a magnetic SET structure as a function of transverse magnetic field $h$ and bias voltage $V$. The polarizations of source and drain are antiparallel and described by $\alpha = 0.98$, while the asymmetry parameter $\beta = 0.9$. For a fixed magnetic field the current changes by a step at the CB threshold, where $V = U$. The background current at $h = 0$ and the large-field current ($h \rightarrow \infty$) increase as expected when the CB is lifted. In contrast, Coulomb correlations promote spin-dependent tunnelling for intermediate fields; the current drops when the CB is lifted, which is a signature of the Coulomb promotion phenomenon described in the text.

Our results Eqs. (7) and (8) for the current are valid for any values of $\hbar$ and $\Gamma^\sigma_\kappa$ that meet the conditions formulated above. In this Letter we are particularly interested
in situations where the Coulomb promotion phenomenon can be observed. Therefore, we assume in the analysis to follow that the leads are made from the same magnetic material and are polarized in opposite directions. This implies that \( \Gamma^\parallel = \Gamma^\perp/\Gamma^R \equiv (1 - \beta)/(1 + \beta) \), where \( \beta \in [0, 1] \) is a polarization parameter. The limiting values \( \beta = 1 \) and \( \beta = 0 \) denote fully polarized and unpolarized leads, respectively. Furthermore, we take the magnetic field to be directed perpendicular to the magnetization in the leads.

From Eq. (5) it is then clear that \( J_c/h^2 \gg J_f/h^2 \) and \( h_f > h_c \). Invoking in addition Eq. (7) we conclude that the magnetic-field promoted (MFP) current is larger in the CB-regime than in the “free” regime if \( h > h_c \). In relatively small magnetic fields, therefore, the MFP-current drops when the bias voltage lifts the Coulomb blockade. This drop, shown in Fig. 2, is a signature of the Coulomb promotion phenomenon.

The relative value of the drop \( \Delta J(h)(J_c(h) - J_f(h))/I_{||} \) (\( I_{||} \equiv I^{(0)}_{||} \)) in the MFP-current at \( V = U \) depends on the magnetic field as well as on the degree of spin polarization. It also depends on the asymmetry of the SET structure, which we characterize by the parameter \( \alpha \equiv (\Gamma_L - \Gamma_R)/\Gamma \). The value \( \Delta J \equiv \max \Delta J(h) \) as a function of \( \alpha \) and \( \beta \) is shown in Fig. 3. We note that the Coulomb promotion phenomenon is most prominent for strongly asymmetric SET structures with highly spin polarized leads, i.e., when \( \alpha \approx 1 \) and \( \beta \approx 1 \). If the leads are fully polarized, \( \beta = 1 \), the background current vanishes. As a result, the MFP-current gives the total current and the Coulomb promotion manifests itself as a negative differential conductance at the bias voltage \( V \approx U \). It is interesting to note that if \( \beta = 1 \), \( \alpha \approx \pm 1 \) and \( h/h_c \ll 1 \) (the case which was qualitatively discussed above) then Eqs. (4) and (5) reproduce Eq. (3) to leading order in small parameters. However, if the leads are not completely spin polarized the Coulomb blockade of the background current competes with the Coulomb of the MFP-current. As a result the negative differential conductance appears only in a small region where the parameter \( \beta > 0.9 \) (see the insert in Fig. 3).

In conclusion we have studied resonant tunnelling of spin-polarized electrons through a magnetic SET device with a central island subject to an external magnetic field directed perpendicular to the magnetization in the leads. The combined effects of spin-dependent tunnelling between a source and a drain with antiparallel magnetizations and of Coulomb correlations were considered. We find that a Coulomb blockade preventing the single electron level on the central ”Coulomb dot” to be doubly occupied may significantly stimulate the transport of electrons through the device. This effect gives rise to a new phenomenon – Coulomb promotion of spin-dependent tunnelling.

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FIG. 3: Drop of the magnetic-field promoted current \( \Delta J \), caused by the lifting of the CB at \( V = U \), as a function of the polarization and asymmetry parameters \( \beta \) and \( \alpha \) (see text). The domain in the \( \alpha, \beta \)-plane, where a negative differential conductance (NDC) may be observed (dark) is shown on the inset. The current drop associated with a NDC is of the order of \( I_{||} \) and peaks at \( \beta = 1, \alpha \approx +1 \).

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