Shuttling of Spin Polarized Electrons in Molecular Transistors

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Shuttling of electrons in single-molecule transistors with magnetic leads in the presence of an external magnetic field is considered theoretically. For a current of partially spin-polarized electrons a shuttle instability is predicted to occur for a finite interval of external magnetic field strengths. The lower critical magnetic field is determined by the degree of spin polarization and it vanishes as the spin polarization approaches 100%. The feasibility of detecting magnetic shuttling in a C60-based molecular transistor with magnetic (Ni) electrodes is discussed [A. N. Pasupathy et al., Science 306, 86 (2004)].

Keywords: single-electron shuttling, molecular transistors, spintronics

1. INTRODUCTION

In recent years the effect of the spin of electrons on the transport properties of nanostructures have been studied intensively, both theoretically and experimentally. In the context of spin-based electronics (spintronics) the possibility to control electrical currents by a weak external magnetic field using the Zeeman and/or the spin-orbit interaction is one of the main goals.

Magnetic materials and especially half-metals are natural sources of spin-polarized electrons for spintronics. Transport of spin-polarized electrons in nanostructures (quantum dots, suspended nanowires, etc.) in external magnetic field results in new phenomena where spin, charge and mechanical degrees of freedom are strongly inter-related. In this new field of investigations (spin-mechanics, see Ref. 1) the presence of a mechanically “soft” subsystem results both in a strong enhancement of spintronic effects and in magnetic control of the mechanical subsystem in the classical as well as in the quantum transport regimes.

Vibrational effects are known to be important for the transport properties of molecular transistors (see, e.g., the reviews in Refs. 2 and 3). In single-molecule transistors a strong electron-vibron coupling was observed in a C60-based transistor with nonmagnetic (gold) leads 4. The measured current-voltage characteristics in this experiment revealed low-energy periodic step-like features. They were interpreted as a signature of vibron-assisted electron tunneling via the fulleren molecule. Experimental I − V curves were theoretically explained 5, 6 in the frames of a simple model of a single-level quantum dot strongly coupled to a single vibrational mode and weakly coupled to the source and drain electrodes.

Later on C60-based molecular transistors with magnetic (Ni) leads were fabricated 7. In samples where the tunneling coupling to the ferromagnetic electrodes were relatively strong (∼ tens of meV), Kondo-assisted tunneling via the C60 molecule was observed. These measurements also proved the presence of a strong inhomogeneous magnetic field produced by the ferromagnetic electrodes in the nano-gap between them. In samples with weak tunneling couplings the usual Coulomb blockade picture for a single-electron transistor was observed.

In the present paper we formulate the conditions for the appearance of a vibrational instability of a fullerene molecule suspended in the gap between two magnetic leads with opposite magnetization. Electron shuttling of spin-polarized electrons produced by magnetic (exchange) forces was predicted in Ref. 8 for the case of 100% polarization of the leads. In this limit (realized for half-metals) the electric current is blocked (spin blockade) in the absence of spin-flips induced by, e.g., an external magnetic field. It was shown that in the absence of dissipation in the mechanical subsystem such a magnetic field triggers a shuttle instability even for vanishingly small fields 9. In the presence of dissipation a threshold magnetic field is determined by the rate of dissipation and it is small for a weak dissipation.

One of our aims here is to develop a theory of magnetic shuttling for conditions corresponding to the experimental set-up of Ref. 2, where the electrons in the ferromagnetic leads were partially polarized (∼ 30%). The absence of a spin blockade in this case qualitatively changes the criterion for electron shuttling. We will show that even in the absence of mechanical dissipation, a shuttling

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regime of electron transport occurs in a finite interval of external magnetic field strengths, \( H_{\text{min}} < H < H_{\text{max}} \), where \( H_{\text{min}} \) is determined by the degree of spin polarization \( \eta \).

In particular, for a high degree of spin polarization \( (\eta \rightarrow 1) \) and if \( \Gamma \gg \hbar \omega \) (where \( \Gamma/\hbar \) is the tunneling rate of majority spin electrons and \( \omega/2\pi \) is the mechanical vibration frequency of the fullerene) the threshold magnetic field for reaching the shuttling regime of electron transport reads: \( H_{\text{min}} \sim \sqrt{1-\eta} \Gamma \). In the limit of "hard" vibrations, \( \Gamma \ll \hbar \omega \), the threshold field is determined by the vibration energy, \( H_{\text{min}} \sim \sqrt{1-\eta} \hbar \omega \).

The calculations outlined below aim at determining the rate of change \( r(H) \) (to be defined later) of the center-of-mass coordinate of the single-molecule shuttle, the sign of which then allows us to formulate the conditions required to observe shuttling of spin-polarized electrons in \( C_{60} \)-based molecular transistors.

2. HAMILTONIAN AND EQUATIONS OF MOTION

The Hamiltonian of a magnetically driven single electron shuttle (see Refs. [6]–[11]) consists of four terms,

\[
\hat{H} = \sum_{j=\text{S,D}} (\hat{H}_j + \hat{H}_{t,j}) + \hat{H}_d + \hat{H}_v, \tag{1}
\]

where \( \hat{H}_j \) is the standard Hamiltonian of noninteracting electrons in the source \((j = S)\) and drain \((j = D)\) electrodes, \( \hat{H}_{t,j} \) is a tunneling Hamiltonian with coordinate dependent tunneling amplitudes \( t_j(\hat{x}) \) and \( \hat{H}_d \) is the Hamiltonian of a single level \((\varepsilon_0)\) quantum dot (QD) magnetically coupled to leads of spin-polarized electrodes by coordinate dependent exchange interactions \( J_j(\hat{x}) \). It has been shown \([3]\) that the shuttling regime of single-electron transport can be realized in the presence of an external magnetic field \( H_{\text{ext}} \) for oppositely magnetized source and drain electrodes. If the external magnetic field is directed perpendicular to the antiparallel polarization vectors of the leads, the QD Hamiltonian reads

\[
\hat{H}_d = \left[ \varepsilon_0 - \frac{J(\hat{x})}{2} \right] a_\uparrow^\dagger a_\uparrow + \left[ \varepsilon_0 + \frac{J(\hat{x})}{2} \right] a_\downarrow^\dagger a_\downarrow - \frac{\mu_H}{2} \left( a_\uparrow^\dagger a_\downarrow + a_\downarrow^\dagger a_\uparrow \right) + U a_\uparrow^\dagger a_\uparrow a_\downarrow^\dagger a_\downarrow. \tag{2}
\]

Here \( a_\sigma^\dagger \) (\( a_\sigma \)) is the creation (annihilation) operator for an electron with spin projection \( \sigma = \uparrow, \downarrow \) on the dot, \( J(\hat{x}) = J_S(\hat{x}) - J_D(\hat{x}) \), \( \mu \) is the Bohr magneton \((g \approx 2)\) and \( U \) is the Coulomb repulsion energy. QD vibrations are described by the harmonic oscillator Hamiltonian

\[
\hat{H}_v = \frac{\hat{p}^2}{2m} + \frac{m\omega^2\hat{x}^2}{2}, \tag{3}
\]

where \( \hat{x} \) is the displacement operator, \( \hat{p} \) is the canonical conjugated momentum \( (\hat{x}, \hat{p} = i\hbar) \), \( m \) is the mass and \( \omega \) is the (angular) vibration frequency of the QD.

The aim of the present paper is to find the conditions under which magnetically driven shuttling in experiments with fullerene-based single-molecule transistors \([4, 7]\) can be realized. In this case the ferromagnetic leads are characterized by a certain degree \( 0 < \eta < 1 \) of spin polarization \((\sim 30\% \) in the experiment of Ref. [6]) and the characteristic vibron energy \( \hbar \omega \) \((\sim 10 \text{ meV}, \text{Ref. [8])} \) is larger or of the order of the energy scale \( \Gamma \) that characterizes the tunneling coupling to the leads, \( \Gamma \leq \hbar \omega \).

It follows from the above considerations that we are not in the adiabatic regime of mechanical motion (which is said to be antiadiabatic when \( \Gamma \ll \hbar \omega \)) and the physical picture of "magnetic shuttling" developed in Ref. [8] (see also Ref. [11]) for the adiabatic regime does not hold here. As in Ref. [12] we will solve the problem using equations of motion for the reduced density operator. In the limit \( eV \gg \Gamma, \hbar \omega, \mu H, k_B T \) \((T \equiv \text{the temperature}, V \equiv \text{the bias voltage})\) the density operator can be factorized into the product of a QD density operator and an equilibrium density matrix for the leads. In the Coulomb blockades regime, \( eV, T \ll U \), the matrix elements of the QD density operator \( \langle \rho_0 = 0 | \rho_\sigma(0) | \rho_\sigma = \langle \sigma | \rho_\sigma | \sigma \rangle, \rho_{\uparrow\downarrow} = \langle \uparrow \downarrow | \rho_{\uparrow\downarrow} | \downarrow \uparrow \rangle \rangle \) in the Hilbert space of a singly occupied dot level (they are still operators in the Hilbert space of a harmonic oscillator) are determined by the following set of equations [13],

\[
\frac{\partial \rho_0}{\partial t} = -i [H_0, \rho_0] - \frac{1}{2} \left\{ \Gamma_0^S(\hat{x}) + \Gamma_0^D(\hat{x}), \rho_0 \right\} + \sqrt{\Gamma_0^D(\hat{x}) \rho_\uparrow} \sqrt{\Gamma_0^D(\hat{x}) \rho_\uparrow}, \tag{4}
\]

\[
\frac{\partial \rho_\uparrow}{\partial t} = -i [H_0, \rho_\uparrow] + \frac{i}{2} \left( J(\hat{x}), \rho_\uparrow \right) + \frac{i\hbar}{2} \left( \rho_{\uparrow\downarrow} - \rho_{\downarrow\uparrow} \right) + \sqrt{\Gamma_0^D(\hat{x}) \rho_\uparrow} \sqrt{\Gamma_0^D(\hat{x}) \rho_\uparrow}, \tag{5}
\]

\[
\frac{\partial \rho_\downarrow}{\partial t} = -i [H_0, \rho_\downarrow] - \frac{i}{2} \left( J(\hat{x}), \rho_\downarrow \right) - \frac{i\hbar}{2} \left( \rho_{\uparrow\downarrow} - \rho_{\downarrow\uparrow} \right) + \sqrt{\Gamma_0^D(\hat{x}) \rho_\downarrow} \sqrt{\Gamma_0^D(\hat{x}) \rho_\downarrow}, \tag{6}
\]

\[
\frac{\partial \rho_{\uparrow\downarrow}}{\partial t} = -i [H_0, \rho_{\uparrow\downarrow}] + \frac{i}{2} \left( J(\hat{x}), \rho_{\uparrow\downarrow} \right) + \frac{i\hbar}{2} \left( \rho_{\uparrow\downarrow} - \rho_{\downarrow\uparrow} \right) - \rho_{\uparrow\downarrow} \sqrt{\Gamma_0^D(\hat{x}) \rho_\uparrow} \sqrt{\Gamma_0^D(\hat{x}) \rho_\uparrow}. \tag{7}
\]

In order to solve Eqs. [13] it is convenient to introduce dimensionless variables for time \((\hbar \omega \rightarrow t)\), dot displacement \((\hat{x}/x_0 \rightarrow \hat{x})\), where \( x_0 = \sqrt{\hbar/m \omega} \) is the zero-point oscillation amplitude), momentum \((p_{\uparrow\downarrow}/\hbar \rightarrow \hat{p})\) and for various characteristic energies \((\hbar \omega \rightarrow 1, g \mu_H/\hbar \omega \rightarrow \hbar, \Gamma_0^S(\hat{x})/\hbar \omega \rightarrow \Gamma_0^S(\hat{x}), \Gamma_0^D(\hat{x}) \rightarrow 2\pi \nu \sqrt{t_{j,\sigma}(\hat{x})^2} \) is the level width, \( \nu \) is the density of states).
for which $\text{Im} \Omega < 0$, corresponding to the shuttle regime of electron transport, satisfy the inequality

$$h^2 + A_1 h^2 + A_2 < 0,$$

where

$$A_1 = -3 \left( \Gamma_1^2 + 1 \right) - \frac{\Gamma_1^2 - \Gamma_2^2}{8 \Gamma_1^2} \left( 5 \Gamma_1^2 + 4 \right),$$

$$A_2 = \frac{\left( \Gamma_2^2 - \Gamma_3^2 \right) \left( \Gamma_1^2 + 4 \right)}{8 \Gamma_1^2} \left( \Gamma_1^2 + 1 \right) + \frac{\Gamma_2^2 - \Gamma_3^2}{4}.$$

Note that in the absence of an external magnetic field the nanoelectromechanical coupling results in additional damping of the mechanical subsystem (since the coefficient $A_2 > 0$).

It is evident from Eq. (13), which is biquadratic in the magnetic field $(h)$, that a shuttle instability occurs in a finite interval of magnetic fields, $h_{\text{min}} < h < h_{\text{max}}$. Now we introduce the degree of spin polarization as

$$\eta = \frac{|N_{1+} - N_{1-}|}{N_{1+} + N_{1-}} \sim \frac{\Gamma - \gamma}{\Gamma + \gamma}$$

where $N_\sigma$ is the number of particles with spin projection $\sigma$. The last relation in Eq. (16) comes from the definition of tunneling rates and our assumption that tunneling amplitudes do not depend on spin projections. We analyze Eq. (13) in two limiting cases:

(i) $\Gamma \gg 1, 1 - \eta \ll 1$, and
(ii) $\Gamma \ll 1, 1 - \eta \ll 1$.

Note that in the limit of weak polarization, $\eta \to 0$, magnetic forces are small, $J \to 0$, and electron shuttling is supported by Coulomb forces. This case was considered in Ref. [12], where it was shown that in the limit of weak polarization a magnetic field ceases to influence single electron shuttling.

In the adiabatic limit, (i), one finds (reverting to using parameters with dimensions) that the shuttle instability region is defined by the double inequality

$$\frac{\Gamma}{2} \sqrt{\frac{1 - \eta}{3}} < g \mu H < \sqrt{3} \Gamma.$$ 

In the antiadiabatic limit, (ii), the critical magnetic fields are determined by the vibron energy $\hbar \omega$ rather than the tunneling rate $\Gamma$, so that

$$\hbar \omega \sqrt{\frac{1 - \eta}{3}} < g \mu H < \sqrt{3} \hbar \omega.$$ 

For the general case that part of the $\Gamma, H$ parameter space which corresponds to a magnetic shuttle instability is shown in Fig. 1 for different degrees of spin polarization. For a given (normalized) tunneling coupling $\Gamma/\hbar \omega$ the range of magnetic fields for which an instability occurs is shifted towards higher fields as the spin polarization decreases. The upper critical field for high spin polarizations, $\eta \to 1$, depends linearly on $\eta$,

$$g \mu H_{\text{max}} \sim \sqrt{3 (\Gamma^2 + (\hbar \omega)^2)} |1 + K (1 - \eta)|,$$
where $K \sim 1$ is a positive constant.

The dependence of the lower critical magnetic field on $\eta$ is weaker. In the antiadiabatic regime one finds that

$$g\mu H_{\text{min}} \sim \hbar \omega \sqrt{1 - \eta}, \quad \Gamma \ll \hbar \omega,$$

and hence $H_{\text{min}}$ rapidly saturates to a constant value of order $\hbar \omega / g\mu$ with decreasing spin polarization (see Fig. 2, short-dashed curve). In the adiabatic regime, $\Gamma \gg \hbar \omega$, the lower critical field decreases linearly with increasing spin polarization, except in the close vicinity of complete spin polarization, where $H_{\text{min}} \propto \sqrt{1 - \eta}$ (compare the solid curve in Fig. 2).

The appearance of an upper and a lower critical magnetic field has a simple physical explanation. When $\mu H$ is the largest energy scale in our problem, $\mu H \gg \Gamma, \hbar \omega$, the fast precession of the electron spin of the dot in a perpendicular external magnetic field nullifies the average spin and the magnetic shuttle instability disappears. To estimate the upper field one may compare the characteristic spin precession frequency, $\mu H_{\text{max}} / \hbar$, with the electron tunneling rate, $\Gamma / \hbar$, or the frequency of vibrations $\omega$. That is $\mu H_{\text{max}} \sim \max (\Gamma, \hbar \omega)$. The lower critical field can be readily estimated for a high degree of spin polarization, $1 - \eta \ll 1$. In this case we have to compare the average time between spin flips, $\tau_f$, induced by a constant magnetic field $H$ in the presence of an electron tunneling coupling $\Gamma$ with the characteristic life-time of minority spin electrons on the dot, $\sim \hbar / \gamma$. The spin-flip rate $\nu_f$ in weak magnetic fields $H$ can be estimated by perturbation theory with the result that $\hbar \nu_f \sim (\mu H)^2 / \max (\Gamma, \hbar \omega)$. Therefore the lower magnetic field is strongly sensitive to spin polarization,

$$\mu H_{\text{min}} \sim \sqrt{\Gamma \gamma} \max (\Gamma, \hbar \omega) \sim \sqrt{1 - \eta} \max (\Gamma, \hbar \omega), \quad (21)$$

and disappears for 100% spin-polarized electrons ($\eta = 1$).

Next we estimate the maximum rate of (exponential) increase, $r_m = -\text{Im} \{\Omega (H_{\text{opt}})\}$, of the QD oscillation amplitude in the shuttle regime. In the adiabatic limit, $\Gamma \gg \hbar \omega$, one finds that $g\mu H_{\text{opt}} \simeq 0.4 \Gamma$ and that

$$r_m \simeq C \frac{\omega J}{\Gamma} \left( \frac{x_0}{\Gamma} \right)^2,$$

where $C \sim 0.1$ is a small numerical factor. In the case $\Gamma \ll \hbar \omega$, which we are interested in here, the maximum rate is realized when $g\mu H_{\text{opt}} \simeq \hbar \omega$, corresponding to

$$r_m \simeq \frac{\Gamma J}{\hbar \omega} \left( \frac{x_0}{\Gamma} \right)^2,$$

where we omit a numerical factor of the order of one.

In the presence of dissipation in the mechanical sub-system, which can be described by adding a phenomenological friction term $\gamma_d \dot{x} \dot{x} (\tau)$ to the equation of motion (9) ($\gamma_d = \omega / Q$, where $Q$ is the quality factor), the shuttling regime appears when $r_m > \omega / Q$. Therefore electron shuttling in a C60-based molecular transistor with magnetic electrodes could be realized if the quality factor $Q$ of the mechanical resonator obeys the inequality

$$Q > Q_{\text{opt}} = \frac{(\hbar \omega)^2}{J \Gamma} \left( \frac{1}{x_0} \right)^2. \quad (24)$$

For the experimental setup in Ref. 4, where fullerene vibrations were observed, the factor $(l / x_0)^2 \simeq 10^3$ and
\[ \Gamma \ll \hbar \omega \sim 5 \text{ meV} \] (one can estimate \( \Gamma \sim 0.1 - 0.5 \text{ meV} \) from the maximal current measured in Ref. [4]). In the \( \text{C}_{60}\)-based transistor with magnetic (Ni) leads \( J \sim \Gamma \sim 10 \text{ meV} \) (see Ref. [2]). From Eq. (24) one can estimate that the required quality factor is \( Q \geq 10^3 - 10^4 \). However the optimal external magnetic field in this case, \( H_{\text{opt}} \approx 50 \text{ T} \), is too high. Instead, we therefore estimate \( Q \) for magnetic fields in the vicinity of the lower critical magnetic field \( H \geq H_{\text{min}} \) where magnetic fields for a very high degree of electron spin polarization (\( \sim 99\% \)) could be of the order of a few tesla. In this case \( \hbar \omega \gg \Gamma \), \( 1 - \eta \ll 1 \)

\[ r(\eta) \simeq \omega \frac{J \Gamma (1 - \eta)}{\Gamma^2 + 4(1 - \eta)(\hbar \omega)^2} \left( \frac{l}{x_0} \right)^2. \tag{25} \]

Assuming that \( \Gamma \simeq \sqrt{1 - \eta} \hbar \omega \) we find \( Q \sim Q_{\text{opt}}/(1 - \eta) \).

4. CONCLUSIONS

In summary we have considered the feasibility of observing magnetically driven single-electron shuttling under realistic conditions corresponding to an already experimentally realized \( \text{C}_{60}\)-based single-molecule transistor with magnetic leads. The main requirement for magnetic shuttling is the presence of an external magnetic field that induces electron spin flips. We have shown that the optimal magnetic field, defined as the field that maximizes the rate of increase of the shuttling amplitude, is determined by the vibration frequency \( \omega \). For fullerene-based single-electron transistors this frequency could be in the THz region \([4]\) with corresponding magnetic fields in the region of several tenths of teslas. For magnetic electrodes with a very high degree of spin polarization one needs less strong (by an order of magnitude) magnetic fields. However, the quality factor of the corresponding mechanical resonator has to be exceptionally high, \( Q \geq 10^5 \).

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[13] Notice that in Eq. (8) of Ref. [12] (analogous to Eq. (7) of the present paper) there are misprints. The corrections are as follows - minus sign in the \( h \)-term and the terms \( \Gamma^\uparrow_0(x) \) and \( \Gamma^\downarrow_0(x) \) have to be omitted.