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Spin reversal processes in a single molecular magnet between two ferromagnetic leads

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The effect of spin polarized transport between ferromagnetic metallic electrodes on the relaxation process of a single molecular magnet (SMM) is considered theoretically. The relaxation times are calculated in the second order approximation (Fermi golden rule). The main objective of the work is to analyze the possible mechanisms responsible for the reversal of a SMM's spin. We investigate the regime in which the spin reversal is driven by an external magnetic field. In such a case, the magnetic switching of a SMM is essentially induced by the quantum tunneling of magnetization. The total charge flowing between the electrodes during the reversal process is calculated, and the analysis shows that such a system under consideration can serve as an electronic pump.

Key words: *single molecular magnet; quantum tunneling of magnetization; transport through magnetic structures*

1. Introduction

In recent years, with the advent of new experimental techniques allowing to attach electrodes to individual molecules, transport properties of single molecules have attracted much attention [1–4]. Among many distinctive classes of molecules, the ones with permanent magnetic moment (single molecular magnets, SMMs) are of particular interest. Their uniqueness follows from their special behaviour in the presence of magnetic field [5, 6]. Due to their large spins and high anisotropy barriers, these molecules exhibit magnetic hysteresis loops with characteristic steps caused by the quantum tunneling of magnetization (QTM). Both their properties and their nanoscale size put SMMs forward as a potential candidate for future applications in information storage and information processing, as well as in spintronics devices.

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The main objective of this work is to investigate the influence of exchange interaction between tunneling electrons and a SMM on the relaxation processes which accompany QTM driven by an external magnetic field. Coupling of the SMM to external leads (via tunneling electrons) is the main source of spin relaxation in the case under consideration. Moreover, the relaxation processes are associated with a transfer of charge from one lead to the other (at zero bias voltage). Consequently, the system may be considered an electronic pump, in which the charge flow between electrodes is driven by an external magnetic field due to the effect of QTM.

2. Model

The system under consideration consists of a SMM sandwiched between two ferromagnetic leads: left (L) and right (R). Magnetic moments of the leads are collinear and either parallel or antiparallel to an external magnetic field along the z axis. The leads are characterized by parabolic conduction bands with the energy dispersion $\varepsilon_{k\sigma}^{p}$, for $p = L, R$. Interaction between the electrons in the leads and the SMM is described by the Appelbaum Hamiltonian [7, 8]:

$$H_T^{\mathrm{int}} = \sum_{kk'\alpha\beta} \left\{ \frac{J}{\sqrt{N_L N_R}}\, \boldsymbol{\sigma}_{\alpha\beta} \cdot \mathbf{S} \left(a_{k\alpha}^{L+} a_{k'\beta}^{R} + a_{k\alpha}^{R+} a_{k'\beta}^{L} \right) \right\} \tag{1}$$

where $\boldsymbol{\sigma} = (\sigma_x, \sigma_y, \sigma_z)$ is the Pauli spin operator, $\mathbf{S}$ stands for the SMM spin operator, $a_{k\sigma}^{p+}$ ($a_{k\sigma}^{p}$) is the creation (annihilation) operator of an electron in the lead p ($p = L, R$), and J is the exchange interaction constant (assumed independent of energy and polarizations of both leads). Furthermore, J is normalized in such a way that it is independent of the size of the electrodes, and N_L, (N_R) denotes the number of elementary cells in the left (right) lead. Using the Hamiltonian (1), we restrict our considerations only to the case of all electrons scattered off the SMM to the opposite electrode (electrons interact with the SMM during the tunneling processes).

The effective Hamiltonian for the ground state spin multiplet of an independent SMM can be written in the form [5, 6],

$$H_{SMM} = -DS_z^2 + E\left(S_x^2 + S_y^2\right) + g\mu_B \left(H_z S_z + H_x S_x\right) \tag{2}$$

where D and E are the second-order anisotropy constants, and the last term stands for the Zeeman energy. The transverse terms (the terms proportional to E and H_x) account for the occurrence of QTM at resonant fields during the magnetic field sweeping. In the absence of transverse terms and assuming that $H_Z = -H$, the energy of the state corresponding to $S_z = m$ is

$$E_m = -Dm^2 - g\mu_B Hm \tag{3}$$

3. Theory and method

The Fermi golden rule was applied to derive characteristic transition times between the neighbouring molecular states. The times for transitions from $S_z = m$ to $S_z = m + 1$, i.e., to the state with larger S_z component, are given by the following formulae:

$$\frac{1}{\tau_m^{>}} = \frac{1}{\tau_m^{LR>}} + \frac{1}{\tau_m^{RL>}} \tag{4}$$

$$\begin{cases} \dfrac{1}{\tau_m^{LR>}} = \dfrac{2\pi}{\hbar} |J|^2 \, v_{ec}^L D_\uparrow^L \, v_{ec}^R D_\downarrow^R \, [A_+(m)]^2 \, \zeta\left[D(2m+1) + g\mu_B H \right] \\ \dfrac{1}{\tau_m^{RL>}} = \dfrac{2\pi}{\hbar} |J|^2 \, v_{ec}^L D_\downarrow^L \, v_{ec}^R D_\uparrow^R \, [A_+(m)]^2 \, \zeta\left[D(2m+1) + g\mu_B H \right] \end{cases} \tag{5}$$

where the indices LR and RL distinguish between electrons tunneling from left to right and from right to left, respectively. Apart from this, D_σ^p is the density of states at the Fermi level in the lead p, v_{ec}^p is its primitive cell volume, $A_\pm(m) = \sqrt{S(S+1) - m(m \pm 1)}$, and $\zeta(\varepsilon) = \varepsilon/[1 - \exp(-\varepsilon\beta)]$ with $\beta = (k_B T)^{-1}$. Analogously, for the transitions from $S_z = m$ to $S_z = m - 1$ we find

$$\frac{1}{\tau_m^{<}} = \frac{1}{\tau_m^{LR<}} + \frac{1}{\tau_m^{RL<}} \tag{6}$$

$$\begin{cases} \dfrac{1}{\tau_m^{LR<}} = \dfrac{2\pi}{\hbar} |J|^2 \, v_{ec}^L D_\uparrow^L \, v_{ec}^R D_\downarrow^R \, [A_-(m)]^2 \, \zeta\left[-D(2m-1) - g\mu_B H \right] \\ \dfrac{1}{\tau_m^{RL<}} = \dfrac{2\pi}{\hbar} |J|^2 \, v_{ec}^L D_\downarrow^L \, v_{ec}^R D_\uparrow^R \, [A_-(m)]^2 \, \zeta\left[-D(2m-1) - g\mu_B H \right] \end{cases} \tag{7}$$

Now, we calculate the mean value of the SMM's spin as a function of magnetic field H taking into account the effect of QTM in the molecule:

$$\langle S_z \rangle = -S\left(1 - \sum_{n=0}^{M} P_{S-n}(H) \right) + \sum_{n=0}^{M} (S-n) P_{S-n}(H) \tag{8}$$

with the system assumed to occupy initially the state $S_z = -S$. Furthermore, the state $S_z = S - M$ is the state with the lowest S_z component, to which the system can tunnel before the complete depletion of the state $S_z = -S$.

The probabilities of a given spin state of the SMM were found by solving numerically the set of master equations, separately for all field ranges between the successive resonant fields. For Mth resonant field the equations are:

$$\begin{cases} c\dot{P}_S = \dfrac{1}{\tau^{>}_{S-1}} P_{S-1} \\ c\dot{P}_m = \dfrac{1}{\tau^{>}_{m-1}} P_{m-1} - \dfrac{1}{\tau^{>}_{m}} P_m, \qquad S-M+1 \le m \le S-1 \\ c\dot{P}_{S-M} = -\dfrac{1}{\tau^{>}_{S-M}} P_{S-M} \end{cases} \tag{9}$$

where $\dot{\circ} \equiv d\circ/dH$ and $c \equiv dH/dt$ is the field sweeping rate. In Eqs. (9) we neglected the relaxation to the lower states, for transition times $\tau^{<}_m \to 0$ when $m \ge 1$ for the relevant magnetic fields. Additionally, relaxation times $\tau^{>}_m$ are considerably smaller than the time scale set by the rate at which the field is varied. Consequently, we assumed that the SMM relaxes to the state $S_z = S$ before each subsequent tunneling act. Therefore the boundary conditions for resonant field $H^{(M)}$ are:

$$\begin{cases} P_S\left(H = H^{(M)}\right) = \sum_{j=0}^{M-1} \tilde{P}_{S-j} \\ P_m\left(H = H^{(M)}\right) = 0, \qquad S-M+1 \le m \le S-1 \\ P_{S-M}\left(H = H^{(M)}\right) = \tilde{P}_{S-M} \end{cases} \tag{10}$$

where $\tilde{P}_{S-j}$ is the probability for SMM to tunnel from the state $S_z = -S$ to the state $S_z = S - j$. The latter can be obtained analytically with the use of the two-level Landau–Zener model [1, 9, 10],

$$\tilde{P}_{S-j} = \left(1 - F_j\right) \prod_{n=0}^{j-1} F_n \qquad \text{and} \qquad F_n = \exp\left[-\frac{\pi \Delta_n^2}{2\left(2S-n\right)\hbar g \mu_B c} \right] \tag{11}$$

Each relaxation process is associated with a single electron charge transfer from one electrode to the other. The total average number of electrons flowing between the electrodes due to relaxation processes during the reversal of the SMM's spin driven by an external magnetic field owing to QTM is

$$Q_{LR} = \frac{1}{c} \sum_{j=1}^{M} \int_{H^{(j)}}^{H^{(j+1)}} dH \;\; P_{S-j}\left(H\right) \left[\frac{1}{\tau^{LR>}_{S-j}\left(H\right)} - \frac{1}{\tau^{RL>}_{S-j}\left(H\right)} \right] \tag{12}$$

where M is determined from the condition $\tilde{P}_{S-(M+1)} \approx 0$, which essentially means that QTM is no longer observed in the system. Since relaxation processes due to the interaction of electrons tunneling between the leads with the SMM take place extremely

fast, we assumed that transition times $\tau_{S-j}^{LR>}$ and $\tau_{S-j}^{RL>}$ do not depend on the field. Consequently, we were able to solve analytically Eqs. (9) and (12):

$$Q_{LR} = \Gamma \sum_{j=1}^{M} j \, \tilde{P}_{S-j} \tag{13}$$

where Γ is the coefficient whose value depends on whether we consider the parallel or anitiparallel configuration of magnetic moments of the electrodes. It can be expressed in terms of polarizations of the electrodes, P^p for $p = L, R$, defined as

$$P^p = \frac{D_+^p - D_-^p}{D_+^p + D_-^p}$$

where + and – denote majority and minority electrons in the electrodes, respectively:

$$\Gamma_{\text{parallel}} = \frac{P^L - P^R}{1 - P^L P^R} \qquad \text{and } \Gamma_{\text{antiparallel}} = \frac{P^L + P^R}{1 + P^L P^R} \tag{14}$$

4. Numerical results and discussion

Consider now the numerical results obtained with the formulae derived above. The average spin of a SMM (see Fig. 1) and the number of electrons pumped between the electrodes during the reversal of the SMM's spin driven by an external magnetic field (Fig. 2), have been calculated for Fe_8, using the parameters from Refs. [5] and [10]. Since the blocking temperature for Fe_8 is 0.36 K, the numerical calculations have been done for $T = 0.01$ K, when no thermal excitations are allowed. For the electrodes we assumed: $v_{ec}^L = v_{ec}^R = 10^{-28}$ m^3, and free electron density $n \approx 10^{29}$ m^3. Apart from this, we assumed $J = 1$ meV.

It is apparent that the average charge, Q_{LR}, transferred between the electrodes during the reversal process of SMM's spin depends on M, i.e. the number of states to which the system can tunnel from its initial state $S_z = -S$ (Eq. (13)). Moreover, Q_{LR} depends on the field sweeping rate c through the probabilities $\tilde{P}_{S-j}$ (Eq. (11)). It follows from the analysis of their behaviour as functions of c that we can restrict considerations to the range $0.1 \le c \le 1$ T/s, in which up to 5 levels are engaged in the reversal process.

The mean value of the SMM's spin is shown in Fig. 1 as a function of magnetic field. It exhibits characteristic steps which occur owing to the QTM for certain values of the field. The height of steps depends on the field sweeping rate through the Landau–Zener probabilities (11). The higher the rate, the more levels take part in the reversal process.

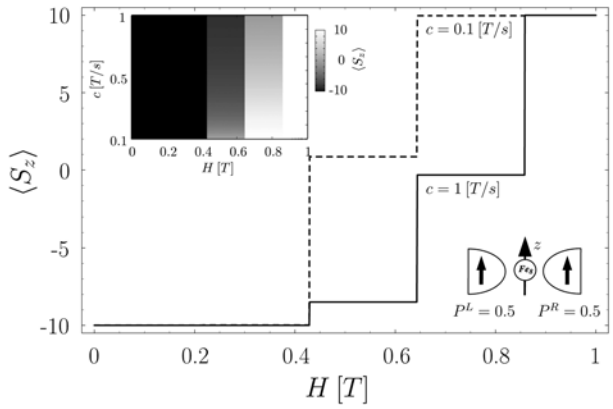

Fig. 1. The mean value of the SMM's spin as a function of magnetic field for $P^L = P^R = 0.5$ in the parallel configuration. The inset presents the continuous change of the field sweeping rate in the range $0.1 \le c \le 1$ T/s

The average charge pumped between the leads depends on the field sweeping rate, magnetic configuration of the electrodes, and the spin polarization of the leads. In Figure 2, we show the average charge pumped between the electrodes in the parallel configuration. One may notice that the sign of the average charge pumped between the leads (direction of average electron flow) is determined by the electrode polarizations. The direction of electron flow can be described by an auxiliary parameter $\alpha = P^R/P^L$. Let us assume that P^L is kept constant, while P^R is varied. In the case shown in Fig. 2, the electrons flow from left to right for $0 \le \alpha < 1$, whereas for $1 < \alpha \le 1/P^L$ they flow in the opposite direction. For $\alpha = 1$ there is no resultant flow of charge.

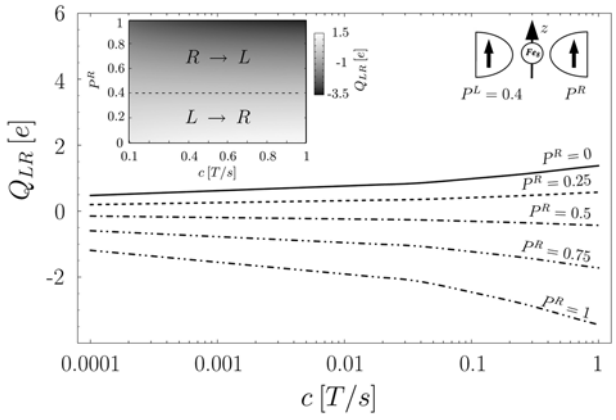

Fig. 2. The average charge pumped between the electrodes in the spin reversal process as a function of the field sweeping rate for various polarizations of the right lead. The leads are kept in the parallel configuration

In conclusion, we have shown that the exchange interaction of electrons tunneling between two electrodes and a SMM leads to charge pumping during the spin reversal process due to QTM driven by an external magnetic field. The effect can be accounted for by taking into account spin relaxation processes involving electron transitions between different electrodes.

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