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Current-induced magnetic switching of an arbitrary oriented single-molecule magnet in the cotunneling regime

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ABSTRACT

Dynamics of current-induced magnetic switching of a single-molecule magnet in the case of Coulomb blockade is investigated theoretically. The molecule is weakly coupled to two ferromagnetic metallic electrodes with collinear magnetic moments, and the molecule's easy axis is assumed to form an arbitrary angle with these moments. The central focus of the paper is placed on discussing the influence of magnetic configuration of the system on the switching mechanism. It is shown that the crucial role in the switching process is played by the angle between the SMM's easy axis and electrodes' magnetic moments.

1. Introduction

Single-molecule magnets (SMMs) have been drawing attention for more than 15 years [1,2]. Due to their unique magnetic properties at low temperatures, resulting from the considerable magnetic anisotropy and large spin number, SMMs have been suggested as promising candidates for applications in novel spintronic devices [3–5]. It has been shown recently that charge transport through a single magnetic molecule is experimentally feasible [6–8]. One of key issues related to the concept of integrating SMMs into electronic circuits is the possibility of manipulating the SMM's spin with a spin-polarized current [9–13]. The main objective of this paper is to address the problem of current-induced magnetic switching (CIMS) of a SMM in the Coulomb blockade regime, where only transport owing to higher-order processes is allowed. As CIMS of a SMM in the sequential transport regime already has been studied and was shown to be quite effective, switching of a SMM in the Coulomb blockade regime requires more detailed analysis.

2. Theoretical description: model and method

We consider a system consisting of a SMM weakly coupled to metallic ferromagnetic electrodes (also called here leads), as shown in Fig. 1(a). The leads are characterized by a band of noninteracting electrons with the dispersion relation $\varepsilon^{q}_{\boldsymbol{k}\sigma}$, where $\boldsymbol{k}$ denotes a wave vector, σ is a spin index and q refers to either left ($q=L$) or right ($q=R$) electrode. Only the case of collinear configurations of the leads' magnetic moments is taken into account. In the sequential tunneling regime [10,11], electronic transport occurs due to tunneling of electrons between the leads and the lowest unoccupied molecular orbital (LUMO) level of the SMM, described by the energy ε. The CIMS can then result from the exchange coupling of the LUMO level with the molecule's spin. In the opposite limit, which is within the scope of this paper, the sequential tunneling processes are suppressed, whereas cotunneling ones start playing a leading role.

Hamiltonian describing the molecule has the form

$$\mathscr{H}_{SMM} = -\left(D+\sum_{\sigma} D_1 c^{\dagger}_{\sigma}c_{\sigma} + D_2 c^{\dagger}_{\uparrow}c_{\uparrow}c^{\dagger}_{\downarrow}c_{\downarrow}\right)S_z^2 + \sum_{\sigma}\varepsilon c^{\dagger}_{\sigma}c_{\sigma} + U c^{\dagger}_{\uparrow}c_{\uparrow}c^{\dagger}_{\downarrow}c_{\downarrow} - \frac{1}{2}\sum_{\sigma\sigma'} J\boldsymbol{\sigma}_{\sigma\sigma'}\cdot\boldsymbol{S}c^{\dagger}_{\sigma}c_{\sigma'}. \quad (1)$$

The first term accounts for the uniaxial magnetic anisotropy of the molecule. Here D is the anisotropy constant of an uncharged molecule, while D_1 and D_2 take into account the effects of the SMM's reduction. Moreover, the operator $c^{\dagger}_{\sigma}$ (c_{σ}) creates (annihilates) an electron in the LUMO level, whereas $\boldsymbol{S}$ is the molecule's spin operator. The next two terms describe the LUMO level, where U is the Coulomb energy of two electrons of opposite spins in the LUMO level. Finally, the last term represents the exchange interaction between the SMM's core and the LUMO level, with $\boldsymbol{\sigma}$ denoting the Pauli spin operator for electrons in the LUMO level and J being the relevant exchange parameter.

Generally, we assume that the SMM's easy axis can be tilted from the orientation collinear with magnetic moments of the electrodes, forming an arbitrary angle φ as shown in Fig. 1(a).

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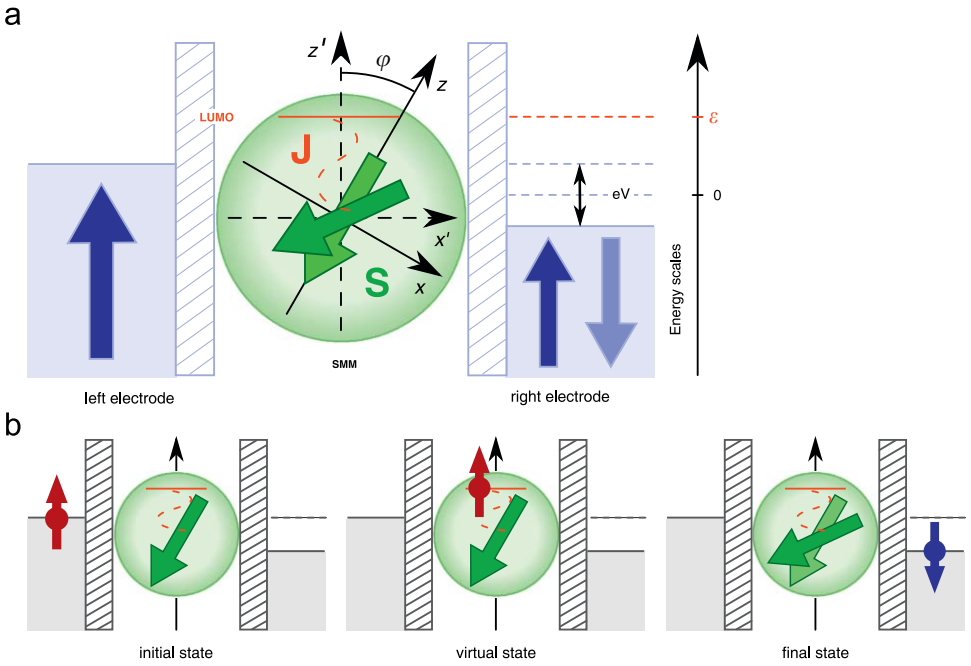

Fig. 1. (Color online) (a) Schematic representation of the system under investigation for collinear (parallel or antiparallel) configuration of the electrodes' magnetic moments. Axes y and y' are perpendicular to planes xz and $x'z'$, respectively. (b) A sketch illustrating one of possible cotunneling processes leading to increase of the z component of the molecule's spin.

In such a case, tunneling of electrons between electrodes and the molecule is described by $\mathscr{H}_T = \sum_{q\boldsymbol{k}\sigma}[T_q a^{q\dagger}_{\boldsymbol{k}\sigma}(\cos(\varphi/2)c_\sigma - \eta_q \sin(\varphi/2)c_{\bar{\sigma}}) + \text{H.c.}]$, where $\eta_{L(R)} = \pm 1$ and $\bar{\sigma} = -\sigma$. Furthermore, T_q denotes the tunneling matrix parameter between the molecule and the qth lead, whereas the operator $a^{q\dagger}_{\boldsymbol{k}\sigma}$ creates an electron of a wave vector $\boldsymbol{k}$ and spin σ in the qth electrode. Due to tunneling processes, the LUMO level acquires a finite spin-dependent width $\Gamma_\sigma = \sum_q \Gamma^q_\sigma$, where $\Gamma^q_\sigma = 2\pi|T_q|^2 D^q_\sigma$ with D^q_σ being the spin-dependent density of states (DOS) at the Fermi level in the lead q. Distinguishing spin-majority $(+)$ and spin-minority $(-)$ electrons, the parameters $\Gamma^q_\pm = \Gamma_q(1 \pm P_q)$ are used to describe the coupling strength between the LUMO level and leads. Here, $\Gamma_q = (\Gamma^q_+ + \Gamma^q_-)/2$, and P_q is the spin polarization of the qth lead, $P_q = (D^q_+ - D^q_-)/(D^q_+ + D^q_-)$. For simplicity the coupling is assumed to be symmetric, $\Gamma_L = \Gamma_R = \Gamma/2$.

It turns out that the Hamiltonian of the molecule, Eq. (1), can be analytically diagonalized, so that each state $|S_t; n, m\rangle$ of the resulting energy spectrum can be represented by the eigenvalue m of the z component of the molecule's total spin, $S^z_t \equiv S_z + 1/2(c^\dagger_\uparrow c_\uparrow - c^\dagger_\downarrow c_\downarrow)$, the corresponding total spin number S_t, and the occupation number n of the LUMO level (for details see Refs.[10,12]). In the case of Coulomb blockade, only the states $|S; 0, m\rangle$, i.e. states with unoccupied LUMO level, can be realized. For natational clarity we assume, therefore, $|S; 0, m\rangle \equiv |m\rangle$. Consequently, the average value of the z component of the SMM's spin, being the quantity to be used for analyzing the switching process, is given by $\langle S_z\rangle = \sum_m m P_{|m\rangle}$, where $P_{|m\rangle}$ denotes the probability of finding the molecule in the state $|m\rangle$. In order to find the probabilities in question, a set of relevant master equation is solved

$$\frac{dP_{|m\rangle}}{dt} = \sum_{qq'}\sum_{m'}[\gamma^{|m'\rangle|m\rangle}_{qq'} P_{|m'\rangle} - \gamma^{|m\rangle|m'\rangle}_{qq'} P_{|m\rangle}]. \tag{2}$$

The molecule is assumed to be initially saturated in the state $|-10\rangle$, and then at time $t = 0$ a constant bias voltage V is applied.

The transition rates $\gamma^{|m\rangle|m'\rangle}_{qq'}$ for inelastic cotunneling processes can be obtained within the standard higher-order perturbation approach

$$\gamma^{|m\rangle|m'\rangle}_{qq'} = \sum_{\substack{\boldsymbol{k}\sigma \in q \\ \boldsymbol{k}'\sigma' \in q'}} W^{q\boldsymbol{k}\sigma,m}_{q'\boldsymbol{k}'\sigma',m'} f(\varepsilon^q_{\boldsymbol{k}\sigma})[1 - f(\varepsilon^{q'}_{\boldsymbol{k}'\sigma'})]. \tag{3}$$

Here, $f(\varepsilon)$ is the Fermi–Dirac distribution function, and the rate at which the system transfers from the initial state $|i\rangle$ to the final one $|f\rangle$ is given by the following expression [13]:

$$W^i_f = \frac{2\pi}{\hbar}\left|\sum_\nu \frac{\langle f|\mathscr{H}_T|\nu\rangle\langle\nu|\mathscr{H}_T|i\rangle}{E_i - E_\nu}\right|^2 \delta(E_f - E_i). \tag{4}$$

In Eq. (3), the initial and final states are defined as $|i\rangle \equiv |\boldsymbol{k}\sigma\rangle_q|m\rangle$ and $|f\rangle \equiv |\boldsymbol{k}'\sigma'\rangle_{q'}|m'\rangle$, where the first ket corresponds to the state of electrodes. From a physical point of view, the above equation represents the process during which one electron is moved from the lead q to the lead q' via a virtual state $|\nu\rangle \equiv |0\rangle|S \pm 1/2; 1, m''\rangle$. The electron virtually occupying the LUMO level may flip its spin orientation, altering via exchange interaction the state of the molecule's spin, Fig. 1(b). Furthermore, E_i, E_f and E_ν denote the energies of the initial, final, and virtual states.

The final expression for the transition rates $\gamma^{|m\rangle|m'\rangle}_{qq'}$ takes the form

$$\gamma^{|m\rangle|m'\rangle}_{qq'} = \frac{\hbar}{2\pi}\sum_{\sigma\sigma'}\Gamma^q_\sigma\Gamma^{q'}_{\sigma'}\int d\varepsilon\left|\sum_{S^\nu_t, m_\nu}\frac{\mathscr{A}(m,m',\nu)}{\varepsilon + \varepsilon_m - \varepsilon_\nu + \mu_q}\right|^2 \times f(\varepsilon)[1 - f(\varepsilon + \varepsilon_m - \varepsilon_{m'} + \mu_q - \mu_{q'})], \tag{5}$$

where μ_q is the electrochemical potential of the qth lead, and $\varepsilon_{m(\nu)}$ is the energy of the SMM with an unoccupied (a singly occupied) LUMO level (see Ref. [10]). Additionally, the coefficient $\mathscr{A}(m, m', \nu)$