Dynamical Aspects of Magnetic Switching in a Single Molecule-Based Spin Valve

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The dynamics of the current-induced magnetic switching process is theoretically studied in a spin-valve device containing a single magnetic molecule of spin $S = 1$. The analysis is performed by using the real-time diagrammatic technique in the sequential electron tunneling regime. In particular, we show that the magnetic moment of a molecule can be reversed also in the presence of intrinsic spin relaxation processes. Moreover, we discuss how the process of magnetic switching depends on a transport bias voltage as well as on some key parameters of the device.

1. Introduction

Thanks to their magnetic bistability [1, 2], single molecular magnets (SMMs) have been extensively studied as suitable candidates for applications in devices used for storing and processing information [3]. One of the key issues in this context is how to control the magnetic state of a SMM, or in other words, how to effectively write a bit of information on a SMM. For instance, the current-induced magnetic switching has been suggested as a prospective mechanism [4–6]. Importantly, the coherent manipulation of a SMM spin can be only achieved at time scales limited by the spin relaxation processes [7]. The main goal of this paper is, thus, to analyze how such spin-relaxation processes affect the dynamics of the SMM spin reversal. Furthermore, we study how the switching time of a molecule depends on its intrinsic parameters.

2. Model and method

The considered spin-valve device consists of a magnetic tunnel junction with a carbon nanotube (CNT) bridging its electrodes, and a SMM deposited on the CNT [8]. In such a setup, which has recently been employed experimentally [9], the CNT acts essentially as a quantum dot (QD), so that whenever the dot is occupied by a single electron, its spin $\hat{s}$ couples *via* exchange interaction J to the effective ground spin $\hat{S}$ of the molecule.

Formally, the system is described by the total Hamiltonian $\hat{\mathcal{H}} = \hat{\mathcal{H}}_{\mathrm{s-valve}} + \hat{\mathcal{H}}_{\mathrm{mol}} + \hat{\mathcal{H}}_{\mathrm{QD-mol}}$. The first term, $\hat{\mathcal{H}}_{\mathrm{s-valve}} = \hat{\mathcal{H}}_{\mathrm{el}} + \hat{\mathcal{H}}_{\mathrm{QD}} + \hat{\mathcal{H}}_{\mathrm{tun}}$, represents a bare spin valve (i.e., without a molecule), whose ferromagnetic metallic electrodes are approximated as reservoirs of noninteracting electrons, $\hat{\mathcal{H}}_{\mathrm{el}} = \sum_{qk\sigma} \varepsilon_{qk\sigma} \hat{c}^\dagger_{qk\sigma} \hat{c}_{qk\sigma}$. The operator $\hat{c}^\dagger_{qk\sigma}$ accounts here for creation of a spin-σ electron with momentum k and the energy dispersion $\varepsilon_{qk\sigma}$

in the qth electrode [$q = \mathrm{L(eft)}, \mathrm{R(ight)}$]. Note that to facilitate the angular momentum transfer between the molecule and spin-polarized electrons tunneling through the QD, which underlies the current-induced magnetic switching mechanism [4], the antiparallel magnetic configuration of spin moments in electrodes is considered.

Furthermore, the QD is modeled as a single-level Anderson impurity, $\hat{\mathcal{H}}_{\mathrm{QD}} = \varepsilon \sum_\sigma \hat{n}_\sigma + U \hat{n}_\uparrow \hat{n}_\downarrow$, with $\hat{n}_\sigma = \hat{d}^\dagger_\sigma \hat{d}_\sigma$ denoting the particle number operator for an electron with spin σ and energy ε occupying the QD, and U being the charging energy of the dot. Electron tunneling processes between electrodes and the QD are captured by $\hat{\mathcal{H}}_{\mathrm{tun}} = \sum_{qk\sigma} (t_q \hat{c}^\dagger_{qk\sigma} \hat{d}_\sigma + \mathrm{H.c.})$, where t_q describes the tunnel matrix elements between the qth electrode and the QD. Tunneling of electrons through the QD leads to spin-dependent broadening of its levels given by $\Gamma^\sigma_q = 2\pi \rho^\sigma_q |t_q|^2$, with ρ^σ_q denoting the spin-dependent density of states in the qth electrode. One can further parameterize Γ^σ_q in terms of the coefficients p_q characterizing spin polarization of electrodes, $p_q = (\Gamma^\uparrow_q - \Gamma^\downarrow_q)/(\Gamma^\uparrow_q + \Gamma^\downarrow_q)$, so that $\Gamma^\sigma_q = (\Gamma_q/2)(1 \pm p_q)$ with $\Gamma_q = \Gamma^\uparrow_q + \Gamma^\downarrow_q$ describing the total broadening due to the tunnel coupling to the qth electrode. Here, we assume that the system under consideration is fully symmetric, i.e., $p_q \equiv p$ and $\Gamma_q \equiv \Gamma$.

Finally, the magnetic molecule is approximated as an effective ground-state giant spin $\hat{S}$ subject to uniaxial magnetic anisotropy along the z-axis, and quantified by the parameter D, with the Hamiltonian of the form [2, 4]

$$\hat{\mathcal{H}}_{\mathrm{mol}} = -D\hat{S}^2_z. \tag{1}$$

On the other hand, the exchange coupling between the molecular spin $\hat{S}$ and the spin $\hat{s}$ of the QD occupied by a single electron can be written as

$$\hat{\mathcal{H}}_{\mathrm{QD-mol}} = -J\hat{S} \cdot \hat{s}, \tag{2}$$

which means that the ferromagnetic (FM) [antiferromagnetic (AFM)] J-coupling is given by $J > 0$ [$J < 0$].

To address the dynamics of molecular spin reversal stimulated by the interaction with electrons tunneling

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through the QD, in the following we calculate the time-dependence of the average value $\langle S_z^{t} \rangle$. This quantity corresponds to the zth component of the spin $\hat{\boldsymbol{S}}^{t} = \hat{\boldsymbol{S}} + \hat{\boldsymbol{s}}$ — i.e., the total spin of the subsystem including the QD and the molecule, defined by $\hat{\mathcal{H}}' = \hat{\mathcal{H}}_{QD} + \hat{\mathcal{H}}_{mol} + \hat{\mathcal{H}}_{QD-mol}$. We obtain it from $\langle S_z^{t} \rangle = \sum_\chi \langle \chi | \hat{S}_z^{t} | \chi \rangle \mathcal{P}_\chi(t)$, where $\mathcal{P}_\chi(t)$ describes the probability of the subsystem to be at time t in the state $|\chi\rangle$, with $\hat{\mathcal{H}}' |\chi\rangle = \varepsilon_\chi |\chi\rangle$. Consequently, the key task is to find the time evolution of probabilities $\mathcal{P}_\chi(t)$, which we achieve by solving iteratively in time the master equation

$$\boldsymbol{\mathcal{P}}(t + dt) = \boldsymbol{\mathcal{P}}(t) + \boldsymbol{W}\boldsymbol{\mathcal{P}}(t)\,dt, \tag{3}$$

for a given initial condition $\boldsymbol{\mathcal{P}}(t = 0) = \boldsymbol{\mathcal{P}}_0$. The vector $\boldsymbol{\mathcal{P}}(t)$ consists of components being probabilities $\mathcal{P}_\chi(t)$, whereas the matrix $\boldsymbol{W} = \boldsymbol{W}^0 + \boldsymbol{W}^r$ in general contains transition rates on the off-diagonal positions, with the diagonal terms $W_{\chi\chi} = -\sum_{\chi'(\neq\chi)} W_{\chi'\chi}$ representing the probability outflow from a state $|\chi\rangle$. We assume that $\boldsymbol{W}$ remains constant in time.

Specifically, the matrix $\boldsymbol{W}^0$ describes transitions due to sequential tunneling of electrons between electrodes and the QD, and it is derived using the real-time diagrammatic technique [10, 11], with its elements given by $W_{\chi\chi'}^0 = W_{\chi\chi'}^{0L} + W_{\chi\chi'}^{0R}$, where

$$W_{\chi\chi'}^{0q} = \sum_\sigma \frac{\Gamma_q^\sigma}{\hbar} \Big\{ f_q(\varepsilon_\chi - \varepsilon_{\chi'}) \big| \langle \chi | \hat{d}_\sigma^\dagger | \chi' \rangle \big|^2$$
$$+ [1 - f_q(\varepsilon_{\chi'} - \varepsilon_\chi)] \big| \langle \chi | \hat{d}_\sigma | \chi' \rangle \big|^2 \Big\}. \tag{4}$$

Above, $f_q(\varepsilon) = \big\{ \exp[(\varepsilon - \mu_q)/(k_B T)] + 1 \big\}^{-1}$ is the Fermi-Dirac distribution for the qth electrode, with μ_q and T denoting the electrochemical potential and temperature, respectively. Furthermore, we assume that both the QD and molecular spin can be in principle subject to various sources of relaxation [2]. These relaxation processes are phenomenologically included *via* the effective relaxation time τ and captured by the matrix $\boldsymbol{W}^r$, whose elements have the form

$$W_{\chi\chi'}^r = \frac{\eta_{\chi\chi'}}{\tau} \cdot \frac{\exp\big[(\varepsilon_{\chi'} - \varepsilon_\chi)/(2k_B T)\big]}{2\cosh\big[(\varepsilon_{\chi'} - \varepsilon_\chi)/(2k_B T)\big]}. \tag{5}$$

Here, $\eta_{\chi\chi'} \equiv \delta_{N(\chi),N(\chi')} \big[\delta_{S_z^t(\chi)-1,S_z^t(\chi')} + \delta_{S_z^t(\chi)+1,S_z^t(\chi')}\big]$ represents the selection rules for relaxation processes, stating basically that such processes conserve the charge of the dot, $N(\chi) = \sum_\sigma \langle \chi | \hat{n}_\sigma | \chi \rangle$, and they cannot change the zth component of molecular spin, $S_z^t(\chi) = \langle \chi | \hat{S}_z^t | \chi \rangle$, by more that one quantum of angular momentum.

3. Numerical results and discussion

To illustrate how the dynamics of the current-induced magnetic switching of a SMM depends on spin-relaxation processes, we consider a model molecule of spin $S = 1$. First of all, we note that here in fact we discuss the dynamics of the total spin $\langle S_z^t \rangle$, that is, the spin of the system consisting of the SMM spin J-exchanged coupled to the QD spin. Since this J-coupling can be of either FM or AFM type, in Fig. 1 we consider both these

cases for a device in the antiparallel magnetic configuration. We assume the initial condition: $\mathcal{P}_\chi(t = 0) = 1$
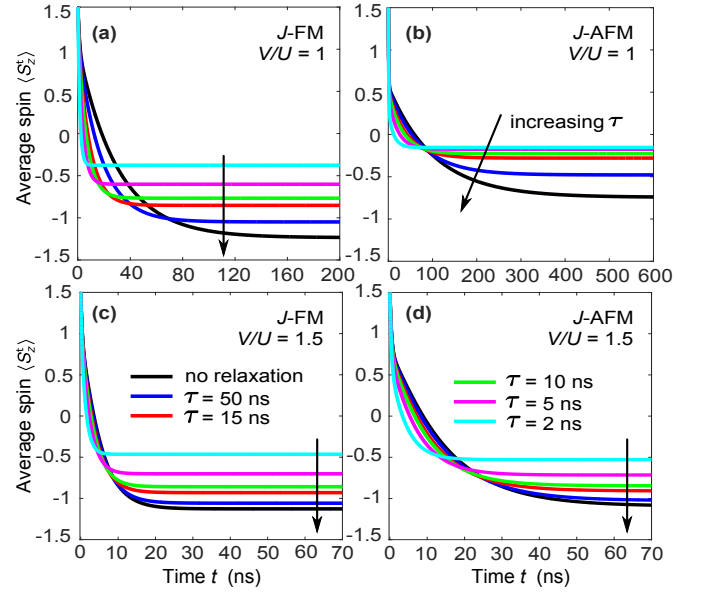

Fig. 1. Time evolution of the average value of the total spin $\langle S_z^t \rangle$ for specified values of the relaxation time τ. Left [right] panels correspond to the ferromagnetic (FM) [antiferromagnetic (AFM)] J-coupling for two values of bias voltage: (a)-(b) $V/U = 1$ and (c)-(d) $V/U = 1.5$. Arrows indicate the direction of increasing τ. Other parameters: $U = 1$, $\varepsilon/U = -0.5$, $D/U = 0.2$, $|J|/U = 0.1$, $S = 1$, $k_B T/U = 0.05$, $\Gamma/U = 0.01$ and $p = 0.5$.

with $N(\chi) = 1$ and $S_z^t(\chi) = 3/2$, while $\mathcal{P}_{\chi'}(t = 0) = 0$ for all states $|\chi'\rangle \neq |\chi\rangle$. Moreover, since the spin reversal becomes possible only if the bias voltage V is of the order of the threshold voltage V_{thr} [4], $V \gtrsim V_{thr}$, the two voltage regimes, $V < V_{thr}$ in (a)-(b) and $V > V_{thr}$ in (c)-(d), are shown. In general, V_{thr} is determined by energy gap between the two states responsible for activating sequential tunneling through the system, and it strongly depends on the value of energy ε, the type and strength of the J-coupling and magnetic anisotropy D. For the two types of exchange interaction and for $\varepsilon = -U/2$, the threshold voltage is explicitly given by: $V_{thr}^{FM} = U + JS$ and $V_{thr}^{AFM} = U - J/2 - D(2S - 1) + \Delta(D, J, S)$, with $\Delta(D, J, S) = \big[D(D - J)(2S - 1)^2 + (J/2)^2(2S + 1)^2\big]^{1/2}$.

In the absence of spin-relaxation processes, the FM J-coupling and for $V < V_{thr}^{FM}$, see Fig. 1(a), the only possible mechanism of relaxation is due to thermally stimulated transitions which involve transfer of spin-polarized electrons across the junction. These processes, in turn, lead to stabilization of the spin with $\langle S_z^t \rangle$ taking large negative value. Importantly, since such thermally stimulated charge transfer processes are exponentially suppressed, they manifest themselves as a slow change of $\langle S_z^t \rangle$. However, as soon as the spin-relaxation processes start to play a role, that is, for shorter relaxation times τ, stabilization of the spin occurs much faster, but then the achieved value of $\big|\langle S_z^t \rangle\big|$ gets reduced. On the

other hand, for the AFM J-coupling and $V < V_{\text{thr}}^{\text{AFM}}$, see Fig. 1(b), the process of reaching the stationary state is slower than for the FM case. This state is again characterized by negative $\langle S_z^{\text{t}} \rangle$, however, with magnitude slightly lower compared to the FM J-coupling case. The difference in time scales results from different threshold voltages depending on the type of exchange interaction. Generally, $V_{\text{thr}}^{\text{FM}} < V_{\text{thr}}^{\text{AFM}}$, such that more time needs to elapse for the system to reach the stationary state through thermally-activated processes.

In the opposite regime of $V > \{V_{\text{thr}}^{\text{FM}}, V_{\text{thr}}^{\text{AFM}}\}$, shown in Figs. 1c,d, where sequential tunneling of electrons through the QD is permitted, the spin of the system switches its orientation more rapidly. As one can see, the spin-relaxation processes affect the final state of the system, hindering the full spin switching for small τ. Noticeably, for assumed parameters of the model, the final state of the system is characterized roughly by the same values of $\langle S_z^{\text{t}} \rangle$ for both types of the J-coupling, though it takes longer for the system to reach this final state in the AFM case.

effectively decoupled from the QD spin, so that the time evolution of the system is almost entirely determined by the dynamical behavior of the latter spin and the spin-relaxation processes.

Furthermore, recall that the magnetic anisotropy parameter D influences the threshold voltage V_{thr}. Consequently, in order to induce the magnetic switching in a molecule exhibiting strong uniaxial magnetic anisotropy (larger D), one has to apply larger bias voltage V. This, in turn, means that for a given value of V the stabilization process of the spin of the system should occur slower for molecules with larger D.

In conclusion, we have studied the dynamics of the current-induced magnetic switching process in a spin-valve system with an embedded SMM of spin $S = 1$. Firstly, we have shown that the time scale at which the spin reversal takes place is strongly affected by the spin-relaxation processes. Secondly, we have also discussed how the dynamics of the magnetic switching depends on a magnitude of key parameters of the system, such as the uniaxial magnetic anisotropy constant and the exchange coupling of the molecular spin to the electronic spins tunneling through the device.
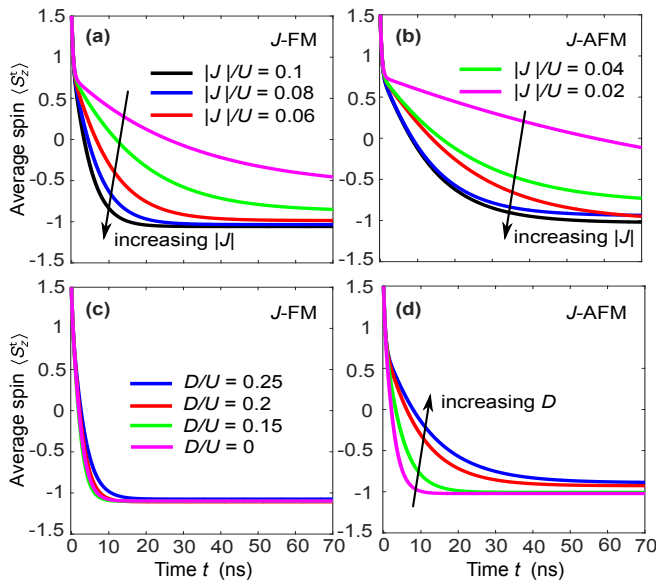
Fig. 2. Dependence of the magnetic switching on internal parameters of the system: (a)-(b) the exchange coupling J between the molecular and QD spins, (c)-(d) the uniaxial magnetic anisotropy parameter D for bias voltage $V/U = 1.5$, $\tau = 50$ ns and other parameters as in Fig. 1. The left/right panels represent the FM/AFM J-coupling, whereas arrows indicate the direction of increase for a given parameter.

The effect of magnetic switching is also affected by the relation between the key model parameters describing the molecule and its coupling to a QD. Figure 2 presents the time evolution of $\langle S_z^{\text{t}} \rangle$ for different values of the exchange coupling parameter J, (a)-(b), and the uniaxial magnetic anisotropy parameter D, (c)-(d), calculated for $V > V_{\text{thr}}$. It can be seen that the spin reversal becomes possible only if the molecule is sufficiently strongly coupled to the dot, that is, for large $|J|$. Otherwise, the molecular spin is

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