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

A. Płomińska, M. Misiorny and I. Weymann

Faculty of Physics, Adam Mickiewicz University, 61-614 Poznań, Poland
Department of Microtechnology and Nanoscience MC2, Chalmers University of Technology, SE-412 96 Göteborg, Sweden

Vol. 133 (2018) ACTA PHYSICA POLONICA A

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 spin relaxation processes 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.

The considered spin-valve device consists of a magnetic (AFM) D, so that whenever the dot is occupied by a single electron, its spin \( \vec{s} \) couples via exchange interaction \( J \) to the effective ground spin \( \vec{S} \) of the molecule.

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

2. Model and method

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.

The dynamics of the current-induced magnetic switching process depends on a transport bias voltage as well as on some key parameters of the device. 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 \( \vec{s} \) couples via exchange interaction \( J \) to the effective ground spin \( \vec{S} \) of the molecule.

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

\[
\hat{\mathcal{H}}_{\text{mol}} = -D \hat{S}_z^2.
\]

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

\[
\hat{\mathcal{H}}_{\text{QD-mol}} = -JS \cdot \vec{s}.
\]

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...
through the QD, in the following we calculate the time-
dependence of the average value \( \langle S^z \rangle \). This quantity cor-
responds to the 2th component of the spin \( \hat{S}^z = \hat{S} + \hat{s} \)
— i.e., the total spin of the subsystem including the QD and
the molecule, defined by \( \hat{H} = \hat{H}_{\text{QD}} + \hat{H}_{\text{mol}} +
\hat{H}_{\text{QD-mol}} \). We obtain it from \( \langle S^z \rangle = \sum_\chi \langle \chi | S^z | \chi \rangle \mathcal{P}_\chi (t) \),
where \( \mathcal{P}_\chi (t) \) describes the probability of the subsystem
to be at time \( t \) in the state \( |\chi \rangle \), with \( \hat{H} |\chi \rangle = -\epsilon_\chi |\chi \rangle \).
Consequently, the key task is to find the time evolution of
probabilities \( \mathcal{P}_\chi (t) \), which we achieve by solving iter-
atively in time the master equation
\[
\mathcal{P}(t + dt) - \mathcal{P}(t) + W \mathcal{P}(t) dt,
\]
for a given initial condition \( \mathcal{P}(t = 0) = \mathcal{P}_0 \). The vec-
tor \( \mathcal{P}(t) \) consists of components being probabilities
\( \mathcal{P}_\chi (t) \), whereas the matrix \( W = W^0 + \hat{W} \) in general
contains transition rates on the off-diagonal positions,
with the diagonal terms \( W_{\chi\chi} = -\sum_\chi (\delta_{\chi\chi} W_{\chi\chi} \) repre-
senting the probability outflow from a state \( |\chi \rangle \). We as-
sume that \( W \) remains constant in time.

Specifically, the matrix \( W^0 \) describes transitions due to
sequential tunneling of electrons between electrodes
and the QD, and it is derived using the real-time dia-
grammatic technique [10, 11], with its elements given by
\( W^0_{\chi\chi} = W^0_{\chi\chi} + W^0_{\chi\chi} \), where
\[
W^0_{\chi\chi} = \sum_\sigma \left\{ \frac{\Gamma_{\chi\chi}^{\sigma}}{\hbar} \left\{ f_\sigma (\epsilon_\chi - \epsilon_\chi^{(\uparrow)}) - \frac{1}{2} \right\} |\chi^\sigma\rangle \langle \chi^\sigma | \right\} ^2,
\]
\[
+ \left\{ 1 - f_\sigma (\epsilon_\chi - \epsilon_\chi^{(\uparrow)}) \right\} |\chi^\sigma\rangle \langle \chi^\sigma | \right\} ^2. \tag{4}
\]
Above, \( f_\sigma (\epsilon) = \frac{\exp(\epsilon - \mu_\sigma) / (k_B T)}{1 + \exp(\epsilon - \mu_\sigma) / (k_B T)} \) is the Fer-
ni-Dirac distribution for the \( q \)th electrode, with \( \mu_\sigma \) and
\( T \) denoting the electrochemical potential and tempera-
ture, 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 \( \text{via} \) the effective relaxation
time \( \tau \) and captured by the matrix \( W^\tau \), whose elements have the form
\[
W^\tau_{\chi\chi} = -\frac{\eta_{\chi\chi}}{\tau} \exp \left( \frac{(\epsilon_\chi - \epsilon_\chi^{(\uparrow)}) / (2k_B T)}{2 \cosh \left( (\epsilon_\chi - \epsilon_\chi^{(\uparrow)}) / (2k_B T) \right)} \right. \tag{5}
\]
Here, \( \eta_{\chi\chi} = \delta_{N(\chi), \chi} |\chi^\sigma\rangle \left\{ \delta_{S^\chi | \chi^\sigma | 1, S^\chi^\sigma | \chi^\sigma |} + \delta_{S^\chi | \chi^\sigma | +1, S^\chi^\sigma | \chi^\sigma |} \right\} \) repre-
sents the selection rules for relaxation processes, stating
basically that such processes conserve the charge of
the dot, \( N(\chi) = \sum_\sigma |\chi^\sigma\rangle \langle \chi^\sigma | \rangle \), and they cannot
change the \( \chi \)th component of molecular spin, \( S^\chi_{\sigma} =
\langle \chi | S^\chi_{\sigma} | \chi \rangle \), by more that one quantum of angu-
lar 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 dy-
namics of the total spin \( \langle S^z \rangle \), that is, the spin of the
system consisting of the SMM spin \( J \)-exchanged cou-
pled 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 configu-
ration. We assume the initial condition: \( \mathcal{P}_\chi (t = 0) - 1

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 $\langle S_z \rangle$, 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 $\tau$. Noticeably, for assumed parameters of the model, the final state of the system is characterized roughly by the same values of $\langle S_z \rangle$ for both types of the $J$-coupling, though it takes longer for the system to reach this final state in the AFM case.

![Dependence of the magnetic switching on internal parameters](image)

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 $\langle S_z \rangle$ 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 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 $V_{\text{thr}}$. 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.

Acknowledgments

This work was supported by the National Science Center in Poland as the Project No. DEC-2013/10/E/ST3/00213. M.M. also acknowledges financial support from the Polish Ministry of Science and Education through a young scientist fellowship (0066/E-336/9/2014) and from the Knut and Alice Wallenberg Foundation.

References