Tunneling anisotropic magneto-resistance in single-molecule magnet junctions

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We theoretically investigate quantum transport through single-molecule magnet (SMM) junctions with ferromagnetic and normal-metal leads in the sequential regime. The current obtained by means of the rate-equation gives rise to the tunneling anisotropic magnetoresistance (TAMR), which varies with the angle between the magnetization direction of ferromagnetic lead and the easy axis of SMM. The angular dependence of TAMR can serve as a probe to determine experimentally the easy axis of SMM. Moreover, it is demonstrated that both the magnitude and sign of TAMR are tunable by the bias voltage, suggesting a promising TAMR based spintronic molecule-device.

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In the past two decades, magnetoresistance in magnetic tunnel-junctions has received much attention because of its strong dependence on the relative magnetization-orientations of two ferromagnetic (FM) electrodes, in terms of which the tunneling magnetoresistance (TMR) devices are developed for various applications in magnetic sensors and information storage technology [1, 2]. More recently the tunneling anisotropic magnetoresistance (TAMR) [3, 4] beyond the conventional TMR has been observed in the presence of spin-orbit interactions, which depends on the relative directions between magnetization-orientation of the FM lead and the crystal axis. Particularly, the TAMR even exists in tunnel junctions with a single magnetic electrode such as Fe/GaAs/Au [4], while the conventional TMR effect does not appear in this configuration. Therefore, the TAMR effect may lead to new spintronic devices with only one magnetic lead [4, 5].

Single-molecule magnets (SMMs) what we consider possess high spins \( S > 1/2 \) and uniaxial magnetic anisotropy with an easy axis, which have potential applications in molecular spintronics [6]. In recent experimental [7, 8] and theoretical [9, 10] studies on electronic transport through magnetic molecules many fascinating properties have been found, such as complex tunneling spectra [9], negative differential conductance [7, 10, 11], Kondo effect [12], Berry phase blockade [13], and colossal spin fluctuations [14]. In particular, the spin-polarized transport through a SMM shows that the magnetization of SMM can be controlled by spin polarized current [10, 13, 17], spin-bias [18] and thermal spin-transfer torque [19]. In addition, a pure spin-current generated by thermoelectric effects [20], polarization reversal of spin-current [21], spin diode behavior [11] and spin filter effect [22, 23] are also found. The conventional TMR effect in spin-dependent transport through a SMM in the sequential, cotunneling and Kondo regimes is also investigated by Misiorny et al. [24], where the magnetizations of FM leads are collinear with the magnetic easy axis of SMM. On the other hand, they also investigate theoretically the magnetic switching of a SMM coupled to two collinear FM leads, in which the magnetic easy axis of SMM forms an angle with the FM electrodes [22].

In this paper, we study anisotropic effects of the tunnel junction (see Fig. 1), which consists of a SMM sandwiched between one FM electrode and one normal metallic (NM) electrode, where the FM electrode is not collinear with the easy axis of SMM different from the TMR case. This setup could be realized by the scanning tunneling microscope with a spin-polarized tip and a SMM on a nonmagnetic metallic substrate [11, 27, 28]. We are going to show that the transport current depends on the magnetization direction of FM electrode, resulting in the TAMR effect.

The system is described by a Hamiltonian [10, 18], \( H = H_{\text{leads}} + H_{\text{SMM}} + H_T \), in which the first term \( H_{\text{leads}} = \sum_{\alpha=L,R} \sum_{k,\tau=\pm} \varepsilon_{\alpha k} c_{\alpha k}^{\dagger} c_{\alpha k\tau} \) is for non-interacting electrons in the magnetic leads with \( c_{\alpha k\tau}^{\dagger} (c_{\alpha k\tau}) \) being the creation (annihilation) operator of an electron in the lead-\( \alpha \) (with \( \alpha = L, R \) denoting the left and right leads respectively) of energy \( \varepsilon_{\alpha k} \) and spin index \( \tau = \pm \). In addition, the spin polarization of the FM lead-\( \alpha \) is defined as \( P_\alpha = (\rho_{\alpha+} - \rho_{\alpha-})/(\rho_{\alpha+} + \rho_{\alpha-}) \), with \( \rho_{\alpha,\pm} \) denoting the state density of the majority (minority) electrons. The model Hamiltonian of SMM is

\[
H_{\text{SMM}} = \sum_{\chi=\pm} \varepsilon d_\chi^{\dagger} d_\chi + Ud_+^{\dagger} d_+ d_-^{\dagger} d_- - Js \cdot S - K(S^z)^2,
\]  

(1)
where $d^i_\alpha(d_\alpha)$ is the relevant electron creation (annihilation) operator in the lowest unoccupied molecular orbital (LUMO) level and $s = \sum_{\chi} d^\dagger_\chi (\sigma \chi/\sqrt{2}) d_{\chi}$ is the corresponding electron spin operator ($\sigma$ is the vector of Pauli matrices, $\chi = \pm$ denotes the spin index). $\varepsilon$ is the single-electron energy of the LUMO level, which is tunable by the gate voltage. $U$ represents the Coulomb energy of two electrons with opposite spins in the LUMO level. $J$ is the exchange coupling parameter between the spin-$S$ of SMM and electron spin-$s$, which can be of either FM ($J > 0$) or antiferromagnetic (AFM) ($J < 0$) type, and the parameter $K > 0$ describes the easy-axis anisotropy of SMM. Moreover the total spin is defined as $S_t \equiv S + s$, and the many-body states of SMM and electron can be written as $|n_S, S_t; m\rangle$, with $n$ denoting the charge state of the molecule, $S_t$ the total spin quantum-number, and $m$ the eigenvalues of $S_t^z$, while the corresponding eigenenergy is $\varepsilon_{n_S, S_t, m}$.

The tunneling processes between the molecule and leads can be described by the Hamiltonian $H_T = \sum_{\alpha k x} t_{\alpha k} c_{\alpha k \dagger} (\cos \frac{\varphi_0}{2} d_k + \chi \sin \frac{\varphi_0}{2} d_{\bar{k}}) \delta_{\chi \bar{\chi}} + H.c.$, where $t_{\alpha}$ denotes the tunnel matrix element of the molecule and the lead-$\alpha$, and $\phi_0$ (with the angle definition $\phi_L = \phi$ and $\phi_R = 0$) is the angle between the magnetization direction of lead-$\alpha$ and the easy axis of the SMM (as $z$-axis). The spin-dependent tunnel coupling-strength is described by $\Gamma_{\alpha \pm} = \Gamma_\alpha (1 \pm P_\alpha)/2$ for spin-majority (upper sign) and spin-minority (lower sign) electrons in the lead-$\alpha$, with $\Gamma_\alpha = \Gamma_{\alpha +} + \Gamma_{\alpha -}$.

For weak coupling between the SMM and leads, i.e., $\Gamma_\alpha \ll k_B T$ and $|J|$, the molecule has enough time to relax to the eigenstates of $H_{SM}^M$ between two consecutive electron tunneling processes. Therefore, we adopt the rate equation approach to study the stationary transport in the sequential regime. Denoting $P_i(t)$ as the occupation probability of the SMM in the molecular state $|i\rangle$ at time $t$, we have

$$\frac{dP_i}{dt} = \sum_{i'} W_{i,i'} P_{i'} - W_{i',i} P_i. \quad (2)$$

Applying the Fermi’s golden rule, the sequential transition rate $W_{i,i'}$ from the state $|i\rangle$ to $|i'\rangle$ with respect to the lead-$\alpha$ is given by

$$W_{i,i'} = \sum_\chi \frac{\Gamma_\alpha (1 + \chi P_\alpha)}{2\hbar} [\langle i| b_{\chi} |i'\rangle]^2 f(\varepsilon_{i'} - \varepsilon_i - \mu_\alpha) + |\langle i'| b_{\chi} |i\rangle|^2 f(\varepsilon_{i'} - \varepsilon_i + \mu_\alpha)], \quad (3)$$

where $b_{\chi} = \cos \frac{\varphi_0}{2} d_\chi + \chi \sin \frac{\varphi_0}{2} d_{\bar{\chi}}$, $\mu_\alpha = (-1)^{S_\alpha} eV/2$, $\varepsilon_i$ being the energy of state $|i\rangle$ and $f(x)$ is the Fermi distribution function. After some algebra, the transition rate Eq. (3) can be rearranged as

$$W_{i,i'} = \sum_\chi \frac{\Gamma_\alpha (1 + \chi P_\alpha \cos \phi_\alpha)}{2\hbar} [\langle i| d_{\chi} |i'\rangle]^2 f(\varepsilon_{i'} - \varepsilon_i - \mu_\alpha) + |\langle i'| d_{\chi} |i\rangle|^2 f(\varepsilon_{i'} - \varepsilon_i + \mu_\alpha)]. \quad (4)$$

Furthermore, by introducing the effective tunnel strength $\Gamma_{\alpha \pm} = \Gamma_\alpha (1 \pm P_\alpha \cos \phi_\alpha)/2$ with the effective spin polarization $P_\alpha \cos \phi_\alpha$ [29], the magnetization of FM lead may be considered as collinear with the easy axis of the SMM.

The stationary probabilities obtained from the condition, $\frac{dP_i}{dt} = 0$, with the transition rate together result in the current through lead-$\alpha$

$$I_\alpha = (-1)^{S_\alpha} e \sum_i (n_i - n_{i'}) W_{i,i'} P_i,$$

and the TAMR ratio in the FM/SMM/NM tunnel junctions is defined as

$$R_{TAMR}(\phi) = \frac{I(0) - I(\phi)}{I(\phi)}, \quad (5)$$

where $I(\phi)$ is the current with the angle $\phi$ between the magnetization direction of FM lead and the easy axis of the SMM.

In Fig. 2(a), we show the bias-voltage dependence of the current for different angles $\phi$ with the FM exchange coupling ($J > 0$), where only the angle range $\phi \in [0, \pi/2]$ is considered, since for $\phi \in [\pi/2, \pi]$ one just reverses the polarization direction of FM electrode. The current spectrum is asymmetric under the bias voltage reversal due to the asymmetric coupling of the SMM and the electron-spin in the FM lead. The current magnitude especially in the bias voltage region, $V = 1 - 5$ mV is more sensitive (with large changes) to the angle variation and increases with it. The current plateau decreases with the angle increase around $V = -2$ mV, while the situation is just opposite around the bias voltage value $V = 2$ mV. It is a well known difficulty that the easy axis of SMM relative to the laboratory frame can not be well determined in SMM transistor experiments [26, 27], and thus the angular-dependent current with respect to the FM lead makes it possible to detect experimentally the magnetic easy axis of SMM [23, 24].

The TAMR ratio as a function of the angle $\phi$ for different bias voltages is plotted in Fig. 2(b), which exhibits a periodic behavior with increasing the angle $\phi$. The TAMR is positive only for the bias voltage $V = -2$ mV (solid line), where the current decreases monotonously with the increase of the angle $\phi$ [see Fig. 2(a)], and is negative otherwise. The TAMR amplitudes with $V = 2$ mV (dashed line) and $V = 4$ mV (dotted line) are larger evidently. Furthermore, when the bias voltage is high enough all transport channels open up and the TAMR
becomes symmetric with respect to the positive and negative bias voltages $V = \pm 6 \text{ mV}$.

Figure 2 displays transport characteristics for the AFM ($J < 0$) exchange interaction, in which the SMM state $|1, 3/2; m\rangle$ has a lower energy than the state $|1, 5/2; m\rangle$. Different from the FM case, the current plateau around $V = \pm 2 \text{ mV}$ decreases (increases) with the increasing angle $\phi$ [Fig. 2(a)], and the TAMR for $V = \pm 2 \text{ mV}$ is positive (negative) [dash (solid) line in Fig. 2(b)]. The TAMR amplitude has the largest value at the bias voltage $V = -2 \text{ mV}$ and exhibits the same behavior as the FM case for $V = \pm 6 \text{ mV}$.

In order to understand the characteristic current-spectrum and the TAMR effect, the main probability distribution of the steady molecular states as a function of the bias voltage $V$ is plotted in Fig. 3, where the FM electrode is collinear with the easy axis of SMM (i.e., $\phi = 0$). We can find that the steady transport for positive bias voltages is determined by the states with negative eigenvalues of spin operator $S_z^T$, while the situation is just opposite for negative bias voltages. This is because of the spin-flip process between the transport electron-spin and the SMM [10,11,14,17].

In the FM case, the main occupied states of the SMM are $|0, 2; -2\rangle$ and $|1, 5/2; -5/2\rangle$ at the bias voltage $V = 2 \text{ mV}$ [see Fig. 3(a)], and thus the transport is dominated by the transition $|0, 2; -2\rangle \leftrightarrow |1, 5/2; -5/2\rangle$ of a spin-down electron. The effective tunnel strength $\Gamma_L^- = \Gamma_L (1 - P_L \cos \phi)/2$ increases with the angle $\phi$ variation from 0 to $\pi/2$, and therefore the current increases, while the TAMR is negative [dash line in Fig. 2(b)]. On the other hand, when $V = -2 \text{ mV}$, the transport is dominated by the transition $|0, 2; 2\rangle \leftrightarrow |1, 5/2; 5/2\rangle$ of a spin-up electron. Since the effective tunnel strength $\Gamma_L^+ = \Gamma_L (1 + P_L \cos \phi)/2$ decreases with the increasing angle $\phi$, the TAMR is positive [solid line in Fig. 2(b)]. At the bias voltage $V = 4 \text{ mV}$, the additional main transitions $|0, 2; -2\rangle \leftrightarrow |1, 3/2; -3/2\rangle$ (spin-up) and $|1, 3/2; -3/2\rangle \leftrightarrow |2, 2; -2\rangle$ (spin-down) take part in the transport processes, and the TAMR exhibits a similar behavior as the case of $V = 2 \text{ mV}$. While at $V = -4 \text{ mV}$ the TAMR amplitude is smaller due to the competition between different transitions of opposite spins.

The state occupations of the AFM exchange interaction are plotted in Fig. 4(b), from which we see that the transport is dominated by the transition $|0, 2; -2\rangle \leftrightarrow |1, 3/2; -3/2\rangle \leftrightarrow |0, 2; 2\rangle \leftrightarrow |1, 3/2; 3/2\rangle$ of a spin-up (spin-down) electron at bias voltage $V = \pm 2 \text{ mV}$, and thus the TAMR is positive (negative) [dash (solid) line in Fig. 3(b)]. The competition between different transitions of opposite spins suppresses the TAMR amplitudes as shown by dotted and dash-and-dot lines respectively for $V = \pm 4 \text{ mV}$ in Fig. 3(b).

In summary, the TAMR is obtained explicitly from the angular-dependent transport in FM/SMM/NM magnetic tunnel junctions by means of the rate-equation ap-
molecular eigenstates as a function of the bias voltage $V$ for FM (a) and AFM (b) exchange couplings in the collinear configuration ($\phi = 0$).

It is demonstrated that the angle between the FM lead and the easy axis of SMM is crucial to induce the TAMR through the exchange coupling between the SMM of uniaxial magnetic anisotropy and transport electron-spin. Both the magnitude and sign of TAMR are efficiently controllable by the bias voltage, suggesting that this SMM tunnel-junction can be a promising spintronic device. On the other hand, this TAMR effect may serve as a probe to detect the uniaxial magnetic anisotropy of the SMM.

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