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Spin current generator in a single molecular magnet with spin bias

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ABSTRACT

In this paper we study the spin- and charge-transport behaviors of a single molecular magnet (SMM) coupled to external reservoirs with spin-dependent chemical potentials. By using the Hubbard operator Green's function method, we show that in the limit of a pinned macrospin the SMM can serve as an effective spin current filter or generator. Two configurations of spin bias are considered. For SMM coupled with symmetric dipolar spin bias, the system can generate spin-polarized current and pure spin current by adjusting the relative magnitude of spin bias and Coulomb interaction. It originates from the competition of two spin-opposite transport channels. Also due to this competition, one can observe a negative differential spin conductance. For SMM coupled with asymmetric dipolar spin bias, the system can only generate spin-polarized currents. The SMM as a spin current generator shows features different from the usual quantum dot system and these features can be useful in molecular spintronics.

1. Introduction

To generate and manipulate a highly spin-polarized current is an essential task in both spintronics and quantum information [1,2]. The quantum dot (QD) tunnel junctions have been proved to be good candidates for obtaining a spin-polarized current. By coupled with external magnetic electrodes, spin bias, spin-orbit coupling, external magnetic field, or polarized light, many fascinating theoretical and experimental results have been reported [3–7]. For example, it was pointed out [4] theoretically that the interplay of Coulomb correlations on the dot and spin polarization of the leads significantly enhances spin precession and leads to negative differential conductance.

On the other hand, with the development of materials science, the single molecular magnet (SMM) has been shown to be another suitable candidate for future molecule-based spintronic devices [8–13]. For instance, it was reported that spin polarized current can induce Magnetic switching of SMM [8] and at low temperatures the interplay of the Kondo effect and spin-polarized transport can leads to suppression of the Kondo effect [12], which also leads to a nontrivial behavior of tunnel magnetoresistance [13]. Interesting enough, owing to the uniaxial anisotropy induced energy barrier, at low temperatures, the SMM can be trapped in one of the two lowest magnetic states $|\pm S\rangle$and only electrons with spin parallel to the local large spin's magnetization can flow through the SMM and generate 100% spin-polarized current [14–16]. Due to this spin selection property, SMM seems to be a very appropriate candidate for designing spin injector or spin-related heat-electricity conversion devices.

Motivated by these achievements, and to explore new materials to generate and manipulate spin-related currents, in this paper we consider a molecule-based spin current generator, which consists of a SMM connected with an external either symmetric or asymmetric dipolar spin bias [17]. The spin bias can inject spin-polarized currents into SMM. In the mean time, following Refs. [15,16], we simplify the SMM as a two-channel model possessing spin-opposite configuration. The interplay of spin bias and SMM's spin-opposite configuration induces interesting current behaviors. With SMM coupled to symmetric dipolar spin bias, we find that by adjusting the relative magnitude of spin bias and Coulomb interaction, the system can generate 100% spin-polarized current or pure spin current, which originates from a competition between two spin-opposite transport channels. Also due to this competition, we find a negative differential spin conductance at the transition point. Here the spin polarization of the current $p = (I_\uparrow - I_\downarrow)/(I_\uparrow + I_\downarrow)$ can reach 100% or infinite. The 100% polarized spin means that there is only one spin component current that can flow through the system ($I_\uparrow = 0$ or $I_\downarrow = 0$). Whereas infinite spin polarization implicates the achievement of a pure spin current ($I_\uparrow + I_\downarrow = 0$, while $I_\uparrow - I_\downarrow \neq 0$). With SMM coupled to asymmetric dipolar spin bias, one can observe only spin-polarized current. It is a feature to distinguish SMM connected with symmetric dipolar spin bias from asymmetric dipolar spin bias.

The paper is organized as follows. In Section 2 we describe the

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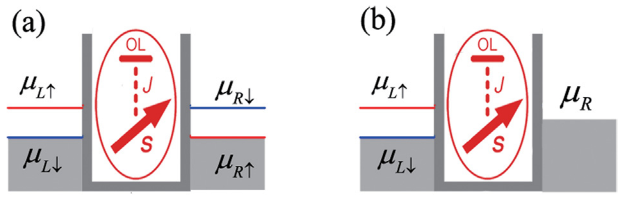

Fig. 1. Schematic depiction of the model under consideration. It consists of a SMM coupled to external either (a) symmetric dipolar spin bias or (b) asymmetric dipolar spin bias. The SMM's orbital level (OL) is exchanged coupled to a local large spin (see Hamiltonian in the text).

model of SMM placed between two leads with symmetric or asymmetric dipolar spin bias. We also describe there the method used in this paper: the Hubbard operator Green's function method. Basic formulas in this Hubbard operator representation are presented. In Section 3 we present the numerical study of spin- and charge-transport behaviors of SMM coupled with two configurations of spin bias. Finally, Section 4 is devoted to a brief conclusion.

2. Model and method

Our system is shown in Fig. 1. The whole system is composed of a SMM connected with spin bias and described by the Hamiltonian as follows [18–21]:

$$H = \sum_{\sigma} \varepsilon_0 d_\sigma^\dagger d_\sigma + U \hat{n}_\uparrow \hat{n}_\downarrow - J\,\mathbf{s}\cdot\mathbf{S} - K(S^z)^2 + \sum_{k,\alpha,\sigma} \varepsilon_{k\alpha} c_{k\alpha\sigma}^\dagger c_{k\alpha\sigma} + \sum_{k,\alpha,\sigma} (t_{k\alpha} c_{k\alpha\sigma}^\dagger d_\sigma + H.\,c.\,), \tag{1}$$

where the SMM is modeled as a molecular orbital level (OL) exchange coupled to a local large spin $\mathbf{S}$, and the total spin operator is $\mathbf{S}_{tot} = \mathbf{s} + \mathbf{S}$. ε_0 is the energy of OL, $d_\sigma^\dagger(d_\sigma)$is the electron creation (annihilation) operator in the OL, with $\sigma = \uparrow, \downarrow$representing the up and down electron spins, U denotes the Coulomb interaction strength, J is the exchange coupling strength between the electron spin $\mathbf{s}$ on the OL and the local spin $\mathbf{S}$, and K is an uniaxial anisotropy parameter. This anisotropy term induces an energy barrier which is crucial for our following discussion. $c_{k\alpha\sigma}^\dagger(c_{k\alpha\sigma})$is the electronic creation (annihilation) operator with momentum k, spin σ, and energy $\varepsilon_{k\alpha s}$ in the electrodes $\alpha = L, R$. The OL is tunnel coupled with electrodes and $t_{k\alpha}$ describes the tunneling amplitude between SMM and leads.

To calculate the current flowing through the system, we use Keldysh non-equilibrium Green's function formalism, a widely used method in the discussion of electronic transport [22–25]. The spin-polarized current is expressed as

$$I_\sigma = -\frac{2e}{h}\int \frac{\Gamma_L\Gamma_R}{\Gamma_L+\Gamma_R}[f_{L\sigma}(\omega) - f_{R\sigma}(\omega)]\,Im G_\sigma^r(\omega)d\omega. \tag{2}$$

For simplicity, in this paper we consider the case of symmetric coupling $\Gamma_L = \Gamma_R = \Gamma = 2\pi\,|t|^2\rho$, where $\rho$the constant density of states for the square wide band approximation. In Eq. (2) $f_{\alpha\sigma}(\omega) = \{\exp[(\omega - \mu_{\alpha\sigma})/k_B T] + 1\}^{-1}$ stands for the Fermi distribution function, with $\alpha = L, R$. Noticing that spin bias means difference between the spin-up and spin-down chemical potentials and it can be considered in the chemical potential $\mu_{\alpha\sigma}$. $G_\sigma^r(\omega) = \langle\langle d_\sigma | d_\sigma^\dagger\rangle\rangle^r$ is the on-site retarded Green's function. Accordingly, the charge current is defined as $I_c = I_\uparrow + I_\downarrow$, and the spin current is defined as $I_s = I_\uparrow - I_\downarrow$ [15].

The next step is to calculate the Green's function $G_\sigma^r(\omega)$. We solve it by the equation of motion (EOM) technique. Before calculation we would like to point out that SMM is a special system involved with a local large spin with $S \geqslant 10$ usually, and this large spin is not expressed as second quantization notation (see the model Hamiltonian in Eq. (1)). Thus it fails the usual second quantization EOM technique. Here, following our previous work [26–32], we use the EOM technique combined with Hubbard operators [33]. The completeness basis of the OL is $\{|0\rangle, |\uparrow\rangle, |\downarrow\rangle, |2\rangle\}$and the electron operators in the OL are rewritten as $d_\sigma = X^{0\sigma} + \delta_\sigma X^{\bar{\sigma}2}$, with $\delta_\sigma = +1(-1)$ for $\sigma = \uparrow(\downarrow)$ and $\bar{\sigma} = -\sigma$; the electron spin operators are written as $s^z = (X^{\uparrow\uparrow} - X^{\downarrow\downarrow})/2$, $s^+ = X^{\uparrow\downarrow}$ and $s^- = X^{\downarrow\uparrow}$, where $X^{ij} = |i\rangle\langle j|$are defined in terms of electron basis states $|i\rangle(|j\rangle)(i, j = 0, \uparrow, \downarrow, 2)$ of the OL. The local large spin operators are expressed as $S^z = \Sigma_{m=-S}^{S} m Y^{mm}$, $S^+ = \Sigma_{m=-S}^{S} C_m^+ Y^{m+1,m}$, and $S^- = \Sigma_{m=-S}^{S} C_m^- Y^{m-1,m}$, with S representing the large spin's quantum number, where $Y^{m,n} = |Sm\rangle\langle Sn|$, and $C_m^\pm = \sqrt{(S \pm m + 1)(S \mp m)}$. Hence in this representation the total Hamiltonian of Eq. (1) is rewritten as

$$\begin{aligned} H = {} & \sum\nolimits_{k,\alpha,\sigma} \varepsilon_{k\alpha} c_{k\alpha\sigma}^\dagger c_{k\alpha\sigma} + \sum_{k,\alpha,\sigma}[t_{k\alpha} c_{k\alpha\sigma}^\dagger (X^{0\sigma} + \delta_\sigma X^{\bar{\sigma}2}) + H.\,c.\,] \\ & + \sum_{\sigma} \varepsilon_0 X^{\sigma\sigma} + (2\varepsilon_0 + U)X^{22} - \frac{J}{2}\sum_{m=-S}^{S} m(X^{\uparrow\uparrow} - X^{\downarrow\downarrow})Y^{mm} \\ & - \frac{J}{2}\sum_{m=-S}^{S} C_m^- X^{\uparrow\downarrow} Y^{m-1,m} - \frac{J}{2}\sum_{m=-S}^{S} C_m^+ X^{\downarrow\uparrow} Y^{m+1,m} - K\sum_{m=-S}^{S} m^2 Y^{mm}. \end{aligned} \tag{3}$$

In the same way, the retarded Green's Function is written as $G_\sigma^r(\omega) = \Sigma_{m=-S}^{S}\langle\langle X^{0\sigma}Y^{mm}|d_\sigma^\dagger\rangle\rangle^r + \Sigma_{m=-S}^{S}\delta_\sigma\langle\langle X^{\bar{\sigma}2}Y^{mm}|d_\sigma^\dagger\rangle\rangle^r$. Before calculating these Hubbard operator Green's Functions, following Refs. [15,16], we introduce a large spin approximation to simplify our problem. Owing to the anisotropy-induced energy barrier (KS^2) of SMM, at low temperatures, the local large spin can be trapped in one of the two lowest bistable magnetic states with $m \geqslant 0$ prepared originally. The electron occupation on the OL of solo SMM could be $n = 0$, 1, or 2 electrons. For $n = 1$, when the local large spin is trapped in $|S\rangle$state, due to exchange coupling, only spin-up electron can occupy the OL; when one more electron tunnels into the OL ($n = 2$), it can only be a spin-down electron. Hence, under this approximation the completeness basis of the OL simplifies to $\{|0\rangle, |\uparrow\rangle, |2\rangle\}$and the total Hamiltonian reads now

$$\begin{aligned} H = {} & \sum\nolimits_{k,\alpha,\sigma} \varepsilon_{k\alpha} c_{k\alpha\sigma}^\dagger c_{k\alpha\sigma} + \sum_{k,\alpha}[t_{k\alpha} c_{k\alpha\uparrow}^\dagger (X^{0\uparrow} + X^{\downarrow 2}) + H.\,c.\,] \\ & + \sum_{k,\alpha}[t_{k\alpha} c_{k\alpha\downarrow}^\dagger(-X^{\uparrow 2}) + H.\,c.\,] + \varepsilon_0 X^{\uparrow\uparrow} + (2\varepsilon_0 + U)X^{22} - \frac{JS}{2}X^{\uparrow\uparrow}Y^{SS} \\ & - KS^2 Y^{SS}. \end{aligned} \tag{4}$$

Accordingly, one needs to calculate the retarded Green's Functions $G_\uparrow^r(\omega) = \langle\langle X^{0\uparrow}Y^{SS}|d_\uparrow^\dagger\rangle\rangle^r$ and $G_\downarrow^r(\omega) = \langle\langle(-X^{\uparrow 2})Y^{SS}|d_\downarrow^\dagger\rangle\rangle^r$. We use the standard EOM of the Green's function $\omega\langle\langle A|B\rangle\rangle^r = \langle\{A, B\}\rangle + \langle\langle[A, H]|B\rangle\rangle^r$ to calculate the retarded Green's Function. $\langle\langle X^{0\uparrow}Y^{SS}|d_\uparrow^\dagger\rangle\rangle^r$ is calculated as

$$\begin{aligned} \left(\omega - \varepsilon_0 + \frac{JS}{2}\right)\langle\langle X^{0\uparrow}Y^{SS}|d_\uparrow^\dagger\rangle\rangle^r = {} & \langle X^{00}Y^{SS}\rangle + \langle X^{\uparrow\uparrow}Y^{SS}\rangle \\ & + \sum_{k\alpha} t_{k\alpha}^*\langle\langle(X^{00} + X^{\uparrow\uparrow})c_{k\alpha\uparrow}Y^{SS}|d_\uparrow^\dagger\rangle\rangle^r \\ & + \sum_{k\alpha} t_{k\alpha}\langle\langle c_{k\alpha_\downarrow}^\dagger X^{02}Y^{SS}|d_\uparrow^\dagger\rangle\rangle^r, \end{aligned} \tag{5}$$

and in the same way $\langle\langle(-X^{\uparrow 2})Y^{SS}|d_\downarrow^\dagger\rangle\rangle^r$ is obtained as

$$\begin{aligned} \left(\omega - \varepsilon_0 - U - \frac{JS}{2}\right)\langle\langle(-X^{\uparrow 2})Y^{SS}|d_\downarrow^\dagger\rangle\rangle^r = {} & \langle X^{\uparrow\uparrow}Y^{SS}\rangle + \langle X^{22}Y^{SS}\rangle \\ & + \sum_{k\alpha} t_{k\alpha}^*\langle\langle(X^{\uparrow\uparrow} + X^{22})c_{k\alpha\downarrow}Y^{SS}|d_\downarrow^\dagger\rangle\rangle^r \\ & + \sum_{k\alpha} t_{k\alpha}\langle\langle c_{k\alpha\uparrow}^\dagger X^{02}Y^{SS}|d_\downarrow^\dagger\rangle\rangle^r \\ & - \sum_{k\alpha} t_{k\alpha}^*\langle\langle X^{\uparrow\downarrow}c_{k\alpha\uparrow}Y^{SS}|d_\downarrow^\dagger\rangle\rangle^r. \end{aligned} \tag{6}$$

Here in Eqs. (5) and (6) the average values $\langle X^{\uparrow\uparrow}Y^{SS}\rangle$ and $\langle X^{22}Y^{SS}\rangle$can be obtained as follows [7,34,35]