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Kondo peak splitting and Kondo dip in single molecular magnet junctions

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ABSTRACT

Many factors containing bias, spin–orbit coupling, magnetic fields applied, and so on can strongly influence the Kondo effect, and one of the consequences is Kondo peak splitting (KPS). It is natural that KPS should also appear when another spin degree of freedom is involved. In this work we study the KPS effects of single molecular magnets (SMM) coupled with two metallic leads in low-temperature regime. It is found that the Kondo transport properties are strongly influenced by the exchange coupling and anisotropy of the magnetic core. By employing Green's function method in Hubbard operator representation, we give an analytical expression for local retarded Green's function of SMM and discussed its low-temperature transport properties. We find that the anisotropy term behaves as a magnetic field and the splitting behavior of exchange coupling is quite similar to the spin–orbit coupling. These splitting behaviors are explained by introducing inter-level or intra-level transitions, which account for the seven-peak splitting structure. Moreover, we find a Kondo dip at Fermi level under proper parameters. These Kondo peak splitting behaviors in SMM deepen our understanding to Kondo physics and should be observed in the future experiments.

1. Introduction

Kondo effect [1–3], first discovered experimentally in the 1930s and then explained theoretically in the 1960s, still serves as an extensively studied prototype in condensed matter physics nowadays. Below a characteristic temperature, in quantum dot experiments [4] it was observed that the strong coupling between leads and dot forms a spin singlet state. These singlet states give rise to a narrow peak in the quantum dot's density of states (DOS) and build up a macroscopically correlated state with properties that is denoted herein as “Kondo singlet”.

Some recent work focused on Kondo peak splitting, which deepen and enrich our understanding to Kondo effect. For instance, it was recently observed in experiments that when the valley degree of freedom of external leads is involved, it reveals a zero-Zeeman-field valley splitting and shows strong temperature dependence [5]. In the mean time, some recent theories and experiments focused on Kondo effect in single molecular magnets (SMMs) [6–12]. SMM, characterized by a large spin magnetic core and magnetic anisotropy, exhibits many interesting transport phenomena, such as negative differential conductance [13–15], current-induced switching [16–18], Berry phase blockade [19], and thermoelectric effects [20,21]. In Kondo regime, due to its large spin degree of freedom, SMM also shows many unique transport features, such as topologically tunable Kondo resonance caused by Berry-phase interference between two quantum tunneling paths of the molecule's spin [8] and tunable tunnel magneto-resistance by exchange coupling and magnetic anisotropy when considering spin-polarized leads [11].

There are many factors that can induce Kondo peak splitting, such as spin–orbit coupling [22], bias and magnetic fields applied [23]. It is expected that when the SMM's large spin degree of freedom is involved, it should induce energy level splitting of the singly occupied orbital level, then Kondo singlet can transport through one of these levels or through two of these levels due to its virtual spin-flip nature, and hence Kondo peak splitting should also appear. Motivated by this thought, we in this work study the Kondo peak splitting in single molecular magnets, mainly focus on the effects of the exchange coupling between singly occupied orbital level and magnetic core and the anisotropy of the magnetic core. We find that the anisotropy term behaves as a magnetic field

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and the splitting behavior of exchange coupling is quite similar to the spin–orbit coupling, resulting from the interplay of exchange coupling and anisotropy of the magnetic core. Indeed, it was found that spin–orbit coupling is responsible for the magnetic anisotropy of SMMs [24]. These splitting behaviors are explained by introducing inter-level or intra-level transitions, which account for the seven-peak splitting structure. To evaluate the dot's density of states and differential conductance we use a non-equilibrium Hubbard operator Green's function method [25]. This method is quantitatively less accurate but qualitatively correct in predicting the Kondo effect at low temperatures. To calculate the relevant Hubbard operator Green's functions with the equations of motion we apply the truncation procedure introduced in Ref. [23].

This paper is organized as follows. In Section 2 we describe the model of single molecular magnets and Hubbard operator Green's function method. Specific Hamiltonian, detailed derivation, basic formula for non-equilibrium Green's functions and the corresponding analytical results are presented there. In Section 3 we present the numerical results and explain the Kondo splitting structures induced by the exchange coupling and anisotropy of magnetic core for minimal non-trivial large spin $S=1$. Finally, Section 4 is devoted to a brief conclusion.

2. Model and Hubbard operator Green's function formalism

Single molecular magnet, as a typical model of molecular systems, can be described by the Hamiltonian proposed by Timm and Elste [15]. The transport setup consists of two metallic electrodes and a SMM, and the total Hamiltonian is written as

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

where $c^{\dagger}_{k\alpha\sigma}(c_{k\alpha\sigma})$ and $c^{\dagger}_{\sigma}(c_{\sigma})$ are the creation (annihilation) operators for the electron in the $\alpha = L, R$ leads and in the molecular orbital (for simplicity only one orbital in the molecule is assumed). The second term represents the tunneling between SMM and leads and $t_{k\alpha}$ is the tunneling amplitude. ϵ is the single-electron orbital energy, which can be tuned by the gate voltage V_g, U is the Coulomb repulsion energy, J is the exchange coupling between electron in the molecular orbital and the magnetic core, and K is the anisotropy energy of magnetic core. $\hat{n}_{\sigma} = c^{\dagger}_{\sigma} c_{\sigma}$ ($\sigma = \uparrow, \downarrow$) is the number operator, $\mathbf{s} = \sum_{\sigma\sigma'} c^{\dagger}_{\sigma}(\tau_{\sigma\sigma'}/2)c_{\sigma'}$ is the electron spin operator with $\tau_{\sigma\sigma'}$ being the Pauli matrix elements, $\mathbf{S}$ is a giant local spin operator with spin length $S \geq 1$, and S^z is the z component of the giant spin operator, where the z direction is chosen along the magnetic core's easy axis. The exchange coupling $J\,\mathbf{s}\cdot\mathbf{S}$ can be rewritten as $Js^zS^z + \frac{J}{2}s^+S^- + \frac{J}{2}s^-S^+$, with $S^{\pm} = S^x \pm iS^y$ (similarly for $s^{\pm}$) the spin-flip operators.

The eigen-system of the unperturbed SMM ($t_{k\alpha} = 0$) has already been presented in Timm's work [15] and here we just summarize the main results. There are four branches of states denoted by $|n, m\rangle^{\nu}$, where n (=0, 1, 2) is the electron number in the orbital, and m is the good quantum number for the z-component of the total spin ($S_z + s_z$). The index $\nu(=\pm)$ appears only when $n=1$. In terms of the electron state $|i\rangle$ ($i = 0, \uparrow, \downarrow, 2$) in molecular orbital and the local large spin state $|m\rangle$ ($m \in [-S, S]$), the empty branch and doubly occupied branch are

$$|0, m\rangle \equiv |0\rangle \otimes |m\rangle, \quad (2)$$

and

$$|2, m\rangle \equiv |\uparrow\downarrow\rangle \otimes |m\rangle, \quad (3)$$

with energies $\epsilon_{0,m} = -Km^2$ and $\epsilon_{2,m} = 2(\epsilon - eV_g) + U - Km^2$, where $m \in [-S, S]$. The singly occupied branches are $|1, \pm(S+1/2)\rangle = |\alpha_{\pm}, \pm S\rangle$ ($\alpha_+ = \uparrow, \alpha_- = \downarrow$) denoting the fully polarized states with energies

$$\epsilon_{1,\pm(S+1/2)} = (\epsilon - eV_g) - \frac{JS}{2} - KS^2 \quad (4)$$

for $m = \pm(S+1/2)$, and

$$|1, m\rangle^{\pm} \equiv A^{\pm}_{m}|\downarrow\rangle \otimes |m+\tfrac{1}{2}\rangle + B^{\pm}_{m}|\uparrow\rangle \otimes |m-\tfrac{1}{2}\rangle, \quad (5)$$

for $m \in [-S+\frac{1}{2}, S-\frac{1}{2}]$, where the coefficients are $A^{\pm}_{m} \equiv \mp\frac{\sqrt{2\Delta E \mp (2K-J)m}}{2\sqrt{\Delta E}}$ and $B^{\pm}_{m} \equiv \frac{J\sqrt{S(S+1)-m^2+1/4}}{2\sqrt{\Delta E}\sqrt{2\Delta E \mp (2K-J)m}}$. The corresponding energies of Eq. (5) are

$$\epsilon^{\pm}_{1,m} = \epsilon - eV_g + \frac{J}{4} - K(m^2 + \frac{1}{4}) \pm \Delta E(m), \quad (6)$$

where $\Delta E(m) \equiv [K(K-J)m^2 + (J/4)^2(2S+1)^2]^{1/2}$. In Eq. (5) $A^{\pm}_{m}$ and $B^{\pm}_{m}$ act as effective Clebsch–Gordan coefficients.

Green's function method [23,25–33] is a widely used method in the discussion of quantum transport problems, including Kondo effects discussed here. Although this method is quantitatively less accurate in predicting the intensity of the Kondo effect, it has been proven to provide correct qualitative physics at low temperatures [34]. To handle large spin involved in SMM, we use Hubbard operator Green's function method [25], which has been used in transport calculations in the regimes such as linear, non-linear response and Kondo regimes [35–38]. In this work our main task is to calculate the density of states $\rho_{\sigma} = -(1/\pi)\mathrm{Im}\,G^{r}_{\sigma}(\omega)$ [39], where $G^{r}_{\sigma}(\omega) \equiv \langle\langle c_{\sigma}|c^{\dagger}_{\sigma}\rangle\rangle^{r}$ is the Fourier transform of the retarded Green's function $G^{r}_{\sigma}(t) = -i\theta(t)\langle\{c_{\sigma}(t), c^{\dagger}_{\sigma}(0)\}\rangle$. Hence in this section we derive the local retarded Green's function of SMM in low-temperature regime when the Kondo effect manifests itself.

The standard equation of motion [40] of Green's function reads $\omega\langle\langle A|B\rangle\rangle^{r} = \langle\{A, B\}\rangle + \langle\langle[A, H]|B\rangle\rangle^{r}$. By using the Hubbard operator representation [41], the electron operators in the molecular orbital are rewritten as $c_{\sigma} = X^{0\sigma} + \delta_{\sigma}X^{\bar{\sigma}2}$, with $\delta_{\sigma} = +1(-1)$ for $\sigma = \uparrow(\downarrow)$, and in the infinite U limit the double-occupation is forbidden so the operator is simplified to $c_{\sigma} = X^{0\sigma}$; the electron spin operators are $s^z = \frac{1}{2}(X^{\uparrow\uparrow} - X^{\downarrow\downarrow})$, $s^+ = X^{\uparrow\downarrow}$ and $s^- = X^{\downarrow\uparrow}$. The local large spin operators are expressed as [25] $S^z = \sum_{m=-S}^{S} mY^{mm}$, $S^+ = \sum_{m=-S}^{S} C^{+}_{m}Y^{m+1,m}$ and $S^- = \sum_{m=-S}^{S} C^{-}_{m}Y^{m-1,m}$ with S being the large spin quantum number, where $Y^{m,n} = |Sm\rangle\langle Sn|$. Hence we rewrite the total Hamiltonian as

$$H = \sum_{k,\alpha,\sigma} \epsilon_{k\alpha} c^{\dagger}_{k\alpha\sigma} c_{k\alpha\sigma} + \sum_{k,\alpha,\sigma} (t_{k\alpha} c^{\dagger}_{k\alpha\sigma} X^{0\sigma} + H.c.) + \sum_{\sigma=\uparrow,\downarrow} \epsilon_0 X^{\sigma\sigma} - \frac{J}{2}\sum_{m=-S}^{S}(mX^{\uparrow\uparrow}Y^{mm} - mX^{\downarrow\downarrow}Y^{mm}) - \frac{J}{2}\sum_{m=-S}^{S}(C^{-}_{m}X^{\uparrow\downarrow}Y^{m-1,m} + C^{+}_{m}X^{\downarrow\uparrow}Y^{m+1,m}) - K\sum_{m=-S}^{S} m^2Y^{mm}, \quad (7)$$

where we define $\epsilon_0 \equiv \epsilon - eV_g$ and $C^{\pm}_{m} \equiv \sqrt{(S \pm m + 1)(S \mp m)}$. $X^{\sigma\sigma'}$