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Effect of finite Coulomb interaction on full counting statistics of electronic transport through single-molecule magnet

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ABSTRACT

We study the full counting statistics (FCS) in a single-molecule magnet (SMM) with finite Coulomb interaction U. For finite U the FCS, differing from $U \to \infty$, shows a symmetric gate-voltage-dependence when the coupling strengths with two electrodes are interchanged, which can be observed experimentally just by reversing the bias-voltage. Moreover, we find that the effect of finite U on shot noise depends on the internal level structure of the SMM and the coupling asymmetry of the SMM with two electrodes as well. When the coupling of the SMM with the incident-electrode is stronger than that with the outgoing-electrode, the super-Poissonian shot noise in the sequential tunneling regime appears under relatively small gate-voltage and relatively large finite U, and dose not for $U \to \infty$; while it occurs at relatively large gate-voltage for the opposite coupling case. The formation mechanism of super-Poissonian shot noise can be qualitatively attributed to the competition between fast and slow transport channels.

1. Introduction

Electronic transport through a single-molecule magnet (SMM) has been intensively studied both experimentally [1] and theoretically [2–9] stimulated by the prospect of a new generation of molecule-based electronic and spintronic devices [10]. Recently, the current fluctuation in electron transport through single-molecule magnet or molecular junction has been attracting much interest [3,6,11–15] owing to its allowing one to identify the internal level structure of the transport system [16] and to access information of electron correlation that cannot be contained in the differential conductance and the average current [17]. These studies were mainly focused on the infinite Coulomb interaction regime [5,6]. In fact, Coulomb interaction is usually finite in a realistic mesoscopic system and thus one should consider the effects of the finite Coulomb interaction on the current correlation. In most mesoscopic systems, the negative correlation induced by Coulomb interaction may impose a time delay between two consecutive electron transfers and lead to the suppression of shot noise so that sub-Poissonian shot noise occurs. For example, in symmetrical double-barrier junctions [18] and in nondegenerate diffusive conductors [19] the 1/2 and 1/3 suppression Fano-factors have been found, respectively. However, in the presence of a strong nonlinearity of the I–V characteristics, the Coulomb interaction can yield a positive correlation and enhance the noise even to be super-Poissonian [20,21]. This phenomenon was first discovered in double-barrier tunneling diodes in the negative differential conductance (NDC) regime and a Fano factor up to 6.6 was observed [20]. In the NDC regime, Coulomb interaction and the density-shape of states in the well introduce positive correlation between consecutive current pulses, which leads to a super-Poissonian shot noise. In general, the effect of Coulomb interaction on the shot noise is more complicated. For instance, in mesoscopic coherent conductors (at sufficiently large voltages) Coulomb interaction decreases the shot noise at low transmissions and increases it at high transmissions [22]. Shot-noise measurements in mesoscopic devices can also provide information about the effective charge e^* of the current-carrying particles. For a quantum-dot system in the Kondo regime, the simultaneous presence of one- and two-quasiparticle scattering results in a universal average charge $e^* = 5/3e$ [23]. However, as shown in Ref. [24], the Coulomb interaction remarkably influences the effective backscattering charge of current-carrying particles via a correction factor ($e^* = 5/3eF$) which is less than unity. Furthermore, the effect of Coulomb interaction on the shot noise can be employed to reveal important information of the energy profile of nonequilibrium carriers injected from an emitter contact, but which cannot be obtained from shot-noise measurements in the absence of Coulomb interactions [25]. In the present SMM system with finite Coulomb energy U, an electron from one lead tunnels into the SMM and then leaves the SMM through the other lead via two kinds of transition processes: (i) between the singly-occupied and empty states, (ii) between doubly-occupied and the singly-occupied states which does

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not occur under the infinite Coulomb interaction. Therefore, it is significant to study the effect of finite Coulomb interaction on the current fluctuation in the SMM system.

An alternative way to investigate the current fluctuation, known as the full counting statistics (FCS), has been proposed in the pioneering work by Levitov et al. [26]. The method yields not only shot-noise power but also all the statistical cumulants of the number of transferred charges. The transport through mesoscopic devices is fully described by the FCS, which may provide the full information about the probability distribution $P(n,t)$ of transferring n electrons between electrode and SMM during a time interval t. The FCS is obtained from the cumulant generating function (CGF) that is related to the probability distribution by [27]

$$e^{-F(\chi)} = \sum_n P(n,t) e^{in\chi}, \tag{1}$$

where χ is the counting field. All cumulants of the current can be obtained from the CGF by performing derivatives with respect to the counting field $C_k = -(-i\partial_\chi)^k F(\chi)|_{\chi=0}$. In the long-time limit, the first three cumulants are directly related to the transport characteristics. For example, the first-order cumulant (the peak position of the distribution of transferred-electron number) $C_1 = \bar{n}$ gives the average current $\langle I\rangle = eC_1/t$. The zero-frequency shot noise is related to the second-order cumulant (the peak-width of the distribution) $S = 2e^2C_2/t = 2e^2(\overline{n^2} - \bar{n}^2)/t$. The third cumulant $C_3 = \overline{(n-\bar{n})^3}$ characterizes the skewness of the distribution. Here, $\overline{(\cdots)} = \sum_n (\cdots)P(n,t)$. In general, the shot noise and the skewness are commonly represented by the Fano factor $F_2 = C_2/C_1$ and $F_3 = C_3/C_1$, respectively.

Since the electron–electron interaction may bring correlations and entanglement of electron states, the shot noise in the mesoscopic system with Coulomb interaction has attracted the significant attention [22,24]. The study of FCS of interacting electrons in mesoscopic systems has become a challenging subject of great interest [27]. In this Letter, we study the FCS of electron transport through SMM weakly coupled to two metallic electrodes, here the first three cumulants are given in terms of numerical calculation. In the model considered here two electrodes are regarded as non-interacting Fermi gases, while the central SMM is treated as a multi-level system with finite Coulomb interaction. We found that the effect of finite Coulomb interaction on the shot noise depends not only on the internal level structure of the SMM, but also on the left–right asymmetry of the SMM-electrode coupling. In particular, our analytical results indicate that for finite Coulomb interaction the FCS, which is different from the case of $U \to \infty$, shows a symmetrical gate-voltage-dependence when both the intensities of the SMM coupling to the left and right electrodes are interchanged, which originates from the both symmetries of the effective channel energy levels and the probability distribution. Moreover, we also observed super-Poissonian noise for symmetric coupling situation. The Letter is organized as follows. In Section 2, we introduce the SMM system and outline the procedure to obtain the FCS formalism based on a particle-number-resolved quantum master equation. The numerical results are discussed in Section 3, where we analyze the occurrence-mechanism of super-Poissonian noise and discuss the effects of Coulomb interaction, the left–right asymmetry of SMM-electrode coupling, and the applied gate voltage on the super-Poissonian noise. Finally, in Section 4 we summarize the work.

2. Model and formalism

A magnetic molecule coupled to two metallic electrodes L (left) and R (right) is described by the Hamiltonian [5] $H_{total} = H_{mol} + H_{Leads} + H_T$. We assume that the SMM-electrode coupling is sufficiently weak so that the electron transport is dominated by sequential tunneling through a single molecular level with on-site energy ε_d. The molecular Hamiltonian is given by

$$H_{mol} = (\varepsilon_d - eV_g)\hat{n} + \frac{U}{2}\hat{n}(\hat{n}-1) - J\vec{s}\cdot\vec{S} - KS_z^2 - B\left(s^z + S^z\right). \tag{2}$$

Here the first two terms depict the lowest unoccupied molecular orbital (LUMO), $\hat{n} \equiv d_\uparrow^\dagger d_\uparrow + d_\downarrow^\dagger d_\downarrow$ is the number operator, where $d_\sigma^\dagger$ (d_σ) creates (annihilates) an electron with spin σ and energy ε_d (which can be tuned by applying a gate voltage V_g) in the LUMO. U is the Coulomb interaction between two electrons in the LUMO. The third term describes the exchange coupling between electron spin $\vec{s}$ in the LUMO and the giant spin $\vec{S}$, the electronic spin operator $\vec{s} \equiv \sum_{\sigma\sigma'} d_\sigma^\dagger(\vec{\sigma}_{\sigma\sigma'})d_{\sigma'}$, where $\vec{\sigma} \equiv (\sigma_x, \sigma_y, \sigma_z)$ denotes the vector of Pauli matrices. The forth term is the anisotropy energy of the magnetic molecule whose easy-axis is Z-axis ($K > 0$). The last term denotes Zeeman splitting. For simplicity, we assume an external magnetic field B is applied along the easy axis of the SMM.

The relaxation in the electrodes is assumed to be sufficiently fast so that their electron distributions can be described by equilibrium Fermi functions. The electrodes are modeled as non-interacting Fermi gases and the corresponding Hamiltonian

$$H_{Leads} = \sum_{\alpha\mathbf{k}\sigma} \varepsilon_{\alpha\mathbf{k}\sigma} a_{\alpha\mathbf{k}\sigma}^\dagger a_{\alpha\mathbf{k}\sigma}, \tag{3}$$

where $a_{\alpha\mathbf{k}\sigma}^\dagger$ ($a_{\alpha\mathbf{k}\sigma}$) creates (annihilates) an electron with energy $\varepsilon_{\alpha\mathbf{k}\sigma}$, momentum $\mathbf{k}$ and spin σ in α ($\alpha = L, R$) electrode. The electron tunneling between the LUMO and the electrodes is described by

$$H_T = \sum_{\alpha\mathbf{k}\sigma} \left(t_\alpha a_{\alpha\mathbf{k}\sigma}^\dagger d_\sigma + \text{H.c.}\right). \tag{4}$$

According to Ref. [5] the eigenstates of an isolated SMM have four branches and may be denoted as $|n,m\rangle^\nu$ where n ($n = 0, 1, 2$) is the electron number in the molecule orbital, and m is the quantum number for the Z-component of the total spin. The index $\nu(=\pm)$ appears only when $n = 1$. In term of the electron state $|i,j\rangle_{LUMO}$ ($i, j = 0, \uparrow, \downarrow$) in molecular orbital and the local spin state $|m\rangle_{GS}$ ($m \in [-S, S]$) the empty-branch and doubly-occupied states may be expressed as

$$|0,m\rangle = |0,0\rangle_{LUMO}|m\rangle_{GS}, \quad m \in [-S, S], \tag{5}$$

and

$$|2,m\rangle = |\uparrow,\downarrow\rangle_{LUMO}|m\rangle_{GS}, \quad m \in [-S, S], \tag{6}$$

respectively, and the two singly-occupied branches are $|1, \pm(S + 1/2)\rangle = |\alpha_\pm\rangle|\pm S\rangle$ ($\alpha_+ = \uparrow, \alpha_- = \downarrow$) for $m = \pm(S + 1/2)$, and

$$|1,m\rangle^\pm = a_m^\pm|\uparrow\rangle_{LUMO}\left|m - \frac{1}{2}\right\rangle_{GS} + b_m^\pm|\downarrow\rangle_{LUMO}\left|m + \frac{1}{2}\right\rangle_{GS}, \tag{7}$$

with

$$a_m^\pm = \frac{J\sqrt{S(S+1) - m^2 + 1/4}}{2\sqrt{\Delta E(m)}\sqrt{2\Delta E(m) \mp (2K - J)m}},$$

$$b_m^\pm = \mp\frac{\sqrt{2\Delta E(m) \mp (2K - J)m}}{2\sqrt{\Delta E(m)}}$$

for $-S + 1/2 \leqslant m \leqslant S - 1/2$, where $\Delta E(m) = [K(K - J)m^2 + (J/4)^2(2S + 1)^2]^{1/2}$. The corresponding energy eigenvalues of molecular eigenstates are given by [5]