

Transport through anisotropic magnetic molecules with partially ferromagnetic leads: Spin-charge conversion and negative differential conductance

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We theoretically investigate inelastic transport through anisotropic magnetic molecules weakly coupled to one ferromagnetic and one nonmagnetic lead. We find that the current is suppressed over wide voltage ranges due to spin blockade. In this system, spin blockade is associated with successive spin flips of the molecular spin and depends on the anisotropy energy barrier. This leads to the appearance of a window of bias voltages between the Coulomb blockade and spin blockade regimes where the current is large and to negative differential conductance. Remarkably, negative differential conductance is also present close to room temperature. Spin-blockade behavior is accompanied by super-Poissonian shot noise, such as in nonmagnetic quantum dots. Finally, we show that the *charge* transmitted through the molecule between initial preparation in a certain spin state and infinite time strongly depends on the initial spin state in certain parameter ranges. Thus the molecule can act as a spin-charge converter, an effect potentially useful as a read-out mechanism for molecular spintronics.

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I. INTRODUCTION

The remarkable miniaturization in semiconductor-based microelectronics in the past decades rapidly approaches its natural limits. In this context, electronic transport through single molecules has received much attention.^{1–17} Since individual molecules are about two orders of magnitude smaller than chip features presently attainable, devices composed of single molecules would present an important breakthrough. Besides, transport through single molecules allows one to study nonequilibrium processes in systems dominated by quantum effects, but having a limited number of relevant degrees of freedom. This is particularly interesting in the presence of vibrations and local magnetic moments. Transport through single molecules is thus of interest for fundamental physics as well.

It is very promising to combine molecular electronics with spintronics, i.e., the idea to employ spins to store and process information, in order to build more efficient memory devices on the nanometer scale.^{18,19} The combination of these two strategies could be realized with magnetic molecules, i.e., molecules with a local magnetic moment.²⁰

Experimental work in this field has focussed on the Zeeman splitting of the Coulomb-blockade (CB) peaks as well as the Kondo effect.^{10,11} More recent transport measurements for a Mn₁₂ derivative have exhibited fine structure of the CB peaks originating from *magnetic* excitations and regions of complete current suppression due to spin selection rules.¹³ These experiments have also led to a number of theoretical works.^{13–17} Furthermore, transport through quantum dots coupled to magnetic leads has been studied extensively.^{21–24} In particular, it has been shown for paramagnetic²⁵ and ferromagnetic²⁴ leads that an external magnetic field, which lifts the spin degeneracy of energy levels, leads to voltage ranges above the CB threshold where the current is strongly suppressed due to different tunneling rates for spin-up and spin-down electrons.²⁵ However, magnetic molecules

coupled to magnetic leads have received little attention.

An essential requirement for spintronics is the ability to effectively control and detect the spin. In a recent paper we have shown that magnetic *anisotropy* is crucial for slow spin relaxation in magnetic molecules and can lead to giant spin amplification.¹⁶ If the molecule is prepared in a magnetic initial state, the current can be highly polarized for an exponentially long time interval. Thus the spin moment transmitted through the molecule depends on the initial orientation of the molecular spin and can be much larger than the initial molecular spin itself.¹⁶ This allows one to effectively *read out* the spin of a molecule coupled to two nonmagnetic leads. The strong anisotropy is expected to arise from the interplay of the ligand field (e.g., in porphyrin complexes or Mn₁₂ derivatives¹³) and spin-orbit coupling as well as from the interaction with image dipoles.

In the present paper we study the inelastic transport through an anisotropic magnetic molecule weakly coupled to one nonmagnetic and one *ferromagnetic* lead employing the rate equation approach.^{5,12–14,16} This configuration is motivated by the possibility to switch the molecule to a predetermined spin state, i.e., to *write* the spin, by applying a bias voltage alone, in zero magnetic field.¹⁶ We show that the proposed configuration leads to interesting physics beyond the effect of spin writing, including the occurrence of large negative differential conductance (NDC) at *high* temperatures. This effect is distinct from the NDC found at *low* temperatures in the fine structure of differential-conductance peaks due to inelastic processes.^{13,16} In the low-temperature transport the anisotropy leads to the appearance of a finite window of bias voltages for which the current is large, whereas it is strongly suppressed on either side due to CB and *spin blockade* (SB),^{13,15,24–29} respectively. By SB we mean the suppression of the current due to small single-electron tunneling rates, in our case due to density-of-states effects. In our system the large local spin leads to interesting modifications of SB, as discussed below.

We also find that the *charge* transmitted through a molecule prepared in a particular spin state depends strongly on this initial state in certain parameter regimes. In fact the difference of the total transmitted charge between preparation and time $t \rightarrow \infty$ is an *exponential* function of a certain energy difference over temperature. This effect is related to the giant spin amplification mentioned above,¹⁶ but appears in the charge channel.

II. MODEL AND METHODS

Our results are obtained for a molecule weakly coupled to two metallic leads. Relaxation in the leads is assumed to be sufficiently fast so that their electron distributions can be described by equilibrium Fermi functions. We assume transport to be dominated by sequential tunneling through a single molecular level with on-site energy ϵ and local Coulomb repulsion U . The full Hamiltonian of the system reads $H = H_{\text{mol}} + H_{\text{leads}} + H_t$, where¹⁶

$$H_{\text{mol}} = (\epsilon - eV_g)n + \frac{U}{2}n(n-1) - J\mathbf{s} \cdot \mathbf{S} - K_2(S^z)^2 - B(s^z + S^z) \quad (1)$$

describes the molecular degrees of freedom, $H_{\text{leads}} = \sum_{\alpha=L,R} \sum_{\mathbf{k}\sigma} \epsilon_{\mathbf{k}\sigma} a_{\mathbf{k}\sigma}^\dagger a_{\mathbf{k}\sigma}$ represents the two leads $\alpha=L,R$ (left, right), and $H_t = \sum_{\alpha=L,R} \sum_{n\mathbf{k}\sigma} (t_\alpha a_{\mathbf{k}\sigma}^\dagger c_\sigma + t_\alpha^* c_\sigma^\dagger a_{\mathbf{k}\sigma})$ describes the tunneling. Here, the operator c_σ^\dagger creates an electron with spin σ on the molecule. $n_d \equiv c_\uparrow^\dagger c_\uparrow + c_\downarrow^\dagger c_\downarrow$ and $\mathbf{s} \equiv \sum_{\sigma\sigma'} c_\sigma^\dagger \boldsymbol{\sigma}_{\sigma\sigma'} c_{\sigma'}$ are the corresponding number and spin operator, respectively. J denotes the exchange interaction between the electrons and the local spin \mathbf{S} . We assume the anisotropy of \mathbf{S} to be of *easy-axis* type, $K_2 > 0$. For simplicity we consider identical g factors for \mathbf{s} and \mathbf{S} . The external magnetic field is applied along the anisotropy axis and a factor $g\mu_B$ has been absorbed into B . Unless stated otherwise, the value of the gate voltage V_g is chosen as zero. $a_{\mathbf{k}\sigma}^\dagger$ creates an electron in lead α with spin σ , momentum \mathbf{k} and energy $\epsilon_{\mathbf{k}\sigma}$.

The eigenstates of the unperturbed Hamiltonian H_{mol} fall into sectors with $n=0,1,2$ electrons.¹⁶ Since $[S_{\text{tot}}^z, H_{\text{mol}}] = 0$, the eigenvalue m of S_{tot}^z is a good quantum number. For $n=0, 2$ we obtain

$$\epsilon(0, m) = -K_2 m^2 - Bm \quad (2)$$

and

$$\epsilon(2, m) = 2(\epsilon - eV_g) + U - K_2 m^2 - Bm. \quad (3)$$

For $n=1$ and $-S+1/2 \leq m \leq S-1/2$ there are two orthogonal states with energies

$$\epsilon^\pm(1, m) = \epsilon - eV_g - Bm + \frac{J}{4} - K_2 \left(m^2 + \frac{1}{4} \right) \pm \Delta E(m) \quad (4)$$

where $\Delta E(m) \equiv [K_2(K_2 - J)m^2 + (J/4)^2(2S+1)^2]^{1/2}$. For the fully polarized states with $n=1$ and $m = \pm(S+1/2)$ the upper (lower) sign applies if $K_2 - J/2$ is positive (negative).

Second-order perturbation theory in the hopping matrix element t_α produces a *Fermi's Golden rule* expression for the

transition rates between two many-particle states $|n\rangle$ and $|m\rangle$ of the molecule,^{5,12,14,16}

$$R_{n \rightarrow m} = \sum_{\alpha=L,R} \sum_{\sigma} \frac{2\pi |t|^2 D_\sigma^\alpha v_{\text{uc}}}{\hbar} (f(\epsilon_m - \epsilon_n - \mu_\alpha) |C_{nm}^\sigma|^2 + [1 - f(\epsilon_n - \epsilon_m - \mu_\alpha)] |C_{mn}^\sigma|^2). \quad (5)$$

Here, D_σ^α denotes the density of states of electrons with spin σ in lead α , v_{uc} is the volume of the unit cell, f denotes the Fermi function, and the ϵ_n are the eigenenergies of H_{mol} of many-particle eigenstates $|n\rangle$.

The matrix elements C_{nm}^σ are defined by $C_{nm}^\sigma \equiv \langle n | c_\sigma | m \rangle$. The occupation probabilities $P^n(t)$ of states $|n\rangle$ are obtained by solving the rate equations

$$\frac{dP^n}{dt} = \sum_m (P^m R_{m \rightarrow n} - P^n R_{n \rightarrow m}). \quad (6)$$

For the steady-state probabilities the left-hand side of this equation vanishes. In the following we assume $\epsilon=1$ eV and $U=2$ eV. Then the doubly occupied molecular states have exponentially small probabilities at the voltages and temperatures we consider. Also, the densities of states satisfy $D_\downarrow^L/D_\uparrow^L=1$ and $D_\downarrow^R/D_\uparrow^R=0.01$, i.e., lead L is nonmagnetic and lead R is a nearly half-metallic ferromagnet. For simplicity we assume $D_\downarrow^L=D_\uparrow^L$. The local spin is chosen as $S=2$.

The current through lead α reads¹⁶

$$I^\alpha \equiv \mp e \sum_{mn} (n_n - n_m) P^m R_{m \rightarrow n}^\alpha, \quad (7)$$

where the upper (lower) sign pertains to $\alpha=L$ (R), n_n denotes the occupation number of the eigenstate $|n\rangle$ and $R_{m \rightarrow n}^\alpha$ contains only those terms in the rates, Eq. (5), that involve lead α . A rate equation approach is also used to compute the current noise spectrum of the system

$$S_{\alpha\beta}(\omega) = 2 \int_{-\infty}^{\infty} dt e^{i\omega t} \langle \delta I^\alpha(t) \delta I^\beta(0) \rangle, \quad (8)$$

where $\delta I^\alpha(t) \equiv I^\alpha(t) - \langle I^\alpha \rangle$ denotes the current fluctuations in lead α and $\langle I^\alpha \rangle$ the steady-state current.³⁰

III. RESULTS

The anisotropy of the local spin partially lifts the degeneracy of the molecular energy levels with respect to the magnetic quantum number m , where m denotes the eigenvalues of the z component of the total spin \mathbf{S}_{tot} .^{13,16,17} This leads to a splitting of peaks of the differential conductance dI/dV at low temperatures, as shown in Fig. 1(a) for finite and in Fig. 1(b) for vanishing magnetic induction B .³¹ The complicated fine structure in the vicinity of the degeneracy point V_0 arises from the anisotropy of the local spin and the exchange interaction of the electrons in the molecular orbital and the local spin. Each fine structure peak corresponds to another transition becoming available for single-electron tunneling.^{13,14,16} Note that dI/dV is *asymmetric* with respect to the bias voltage, i.e., several fine-structure peaks have a significantly different intensity when the bias changes sign. This results from

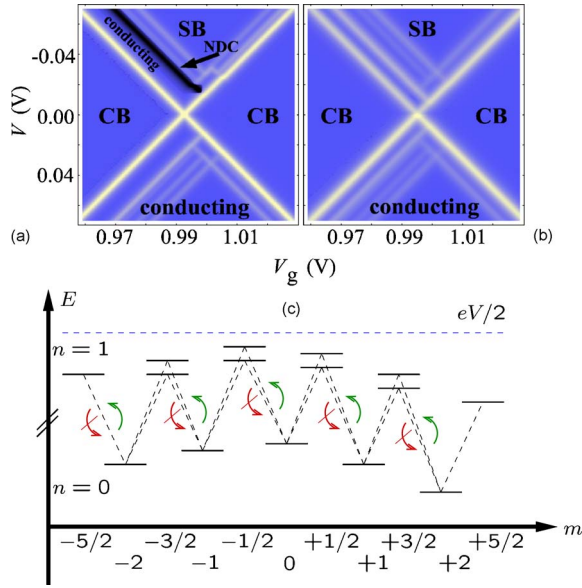


FIG. 1. (Color online) Two-dimensional plots of the differential conductance dI/dV in the vicinity of one particular degeneracy point at low temperatures. (a) dI/dV for Zeeman splitting $B = 0.05$ eV. (b) dI/dV for vanishing magnetic field. The model parameters are $J=K_2=5$ meV. When the magnetic field is switched on, the CB regime and the SB regime are separated by a finite window of bias voltage for which the conductance is high. Since it is low (governed by $D_{\downarrow}^R/D_{\uparrow}^R$) in the SB regime, NDC must occur where the conductance drops again. (c) Scheme of the molecular energy levels with all allowed transitions involving the states with $n=0$ and $n=1$ electrons in an external magnetic field. In the SB regime all transitions are *energetically* possible if the bias voltage V is large enough. However, the rates for a spin-down electrons tunneling from the molecule into the right lead (curved arrows with slash) are strongly suppressed due to the small density of states.

the breaking of spin-rotation symmetry by the ferromagnetic lead together with the spin selection rules for the tunneling processes.

The V - V_g map shows three transport regimes: At low bias the current is thermally suppressed due to CB, except close to the degeneracy point. The electron number on the molecule is constant, i.e., $n=0$ for $V_g < V_0$ and $n=1$ for $V_g > V_0$, since all transitions between different molecular charge states are energetically forbidden. At large *positive* bias the conductivity of the molecule is high, since the electrons in the right lead have enough energy to overcome the energy barrier between the $n=0$ and $n=1$ states. This is the conducting regime. At large *negative* bias the conductivity of the molecule is low. This current suppression is due to the SB mechanism explained in the following.

Two main definitions of SB are used in the literature on transport through quantum dots. The original definition refers to the phenomenon that transition probabilities for single-electron tunneling vanish between states corresponding to successive electron numbers if the total spins differ by more than $\Delta S=1/2$.^{13,15,26,28,29} The other, more general definition refers to the situation that the tunneling rate for electrons of one spin direction is strongly suppressed relative to the other, e.g., due to ferromagnetic leads^{24,27} or Zeeman splitting.²⁵ In

this case the system can be stuck in a particular molecular many-body state because the rates for leaving this state are small.

In our case the SB is related to the second mechanism,^{24,25,27} but the interaction between the electrons on the molecule and the local anisotropic spin leads to modifications: As soon as the bias is sufficiently high, all transitions between the $n=0$ and the $n=1$ multiplets are energetically possible, as shown in Fig. 1(c). Selection rules for sequential tunneling require $\Delta m=1/2$. Both spin-up and spin-down electrons hop *onto* the molecule with equal rates, since the densities of states D_{\uparrow}^L and D_{\downarrow}^L in the left (incoming) lead are equal for both spin directions. On the other hand, spin-up electrons leave the molecule much faster due to the polarization of the right (outgoing) lead. Electrons keep flowing through the molecule until a spin-down electron tunnels in. This spin-down electron can leave the molecule only with a very small tunneling rate due to the low density of states D_{\downarrow}^R . On the other hand, it can rapidly leave the molecule as a spin-up electron if the local spin is simultaneously reduced by unity. However, the number of possible spin flips is limited, depending on the initial spin state. Therefore, the molecule finally ends up in the singly charged state with minimal spin, i.e., $S_{\text{tot}}=-S-1/2$. Further electron tunneling is *blocked*, since the left lead is energetically unreachable and the right lead has a low density of states for spin-down electrons, cf. Fig. 1(c).

Another interesting feature shown in Fig. 1(a) is the appearance of a finite window of bias voltages between the CB and SB regimes for which the conductivity of the molecule is high. This obviously leads to NDC when the SB regime with low conductivity is entered. Figure 2(a) shows the current-voltage characteristics in the vicinity of the CB threshold at zero gate voltage for finite magnetic induction. The external field tilts the molecular energy levels with respect to the magnetic quantum number m due to the additional Zeeman energy, as sketched in Fig. 2(b). This removes the degeneracy of the spin multiplets. For small bias voltages and low temperatures the current is suppressed due to CB. But as soon as the transition from the ground state of the $n=0$ multiplet with $m=S$ to the lowest-energy state of the $n=1$ multiplet with $m=S+1/2$ becomes energetically allowed, cf. Fig. 2(b), the current increases to the plateau shown in Fig. 2(a). These two levels are then equally occupied. Moreover, the current through the molecule is highly spin polarized, since spin-up electrons tunnel rapidly through the molecule whereas tunneling of spin-down electrons is thermally suppressed. When the bias is further increased to allow tunneling also of spin-down electrons, the molecule in several steps goes over to the state with $n=1$ and $m=-S-1/2$ and the current is strongly suppressed by the SB mechanism discussed above. The first required transition from $n=0$ and $m=S$ to $n=1$ and $m=S-1/2$ has the highest energy so that all following transitions become active at the same bias.¹⁴ The width ΔV of the window of bias voltages with large current corresponds to twice the difference of these two excitation energies and is given by¹⁶

$$e\Delta V = 2[2SK_2 + B - \Delta E(S+1/2) - \Delta E(S-1/2)], \quad (9)$$

where $\Delta E(m) \equiv [K_2(K_2 - J)m^2 + (J/4)^2(2S+1)^2]^{1/2}$. This holds as long as $D_{\downarrow}^R/D_{\uparrow}^R \ll 1$ and the Zeeman energy B is not

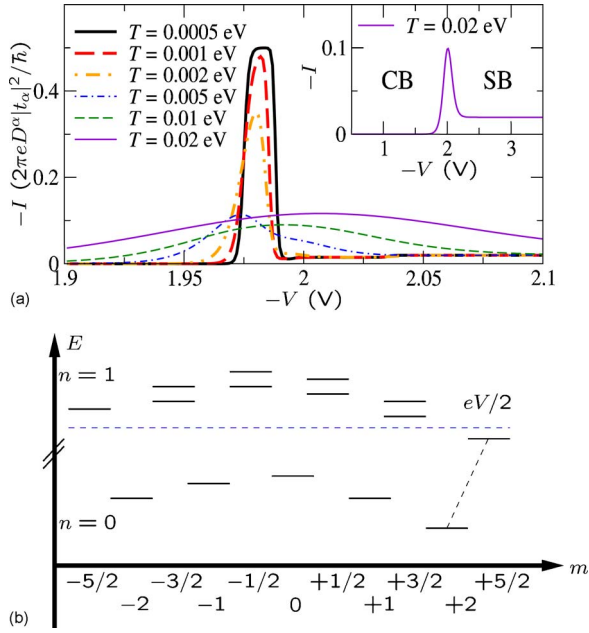


FIG. 2. (Color online) (a) Current-voltage characteristics in the vicinity of the CB threshold assuming $K_2 = J = B = 0.05$ eV. The inset shows the current for $T = 0.02$ eV over a broader voltage range. (b) Energy level scheme for the $n=0$ and $n=1$ spin multiplets as a function of the magnetic quantum number m . The bias voltage is just high enough to allow the transition between the many-particle states with spin $m=S$ and $m=S+1/2$. The steady-state current is highly spin polarized, since the tunneling of spin-down electrons is thermally suppressed. This regime corresponds to the *plateau* with enhanced current in (a).

too small—for $B \rightarrow 0$ we have to take into account that the states with $n=0$ and $m = \pm S$ become degenerate ground states. The observation of the current plateau requires the temperature to be small compared to $e\Delta V$, see Fig. 2(a).

At *high* temperatures the current steps broaden so that the plateau of high current vanishes. Nevertheless, the current remains large at bias voltages close to the transition point from CB to SB, as shown in the inset in Fig. 2(a). For vanishing magnetic induction, $B=0$, there is no enhanced current at low temperatures, as we will explain below. Nevertheless, at high temperatures a pronounced current maximum develops close to the transition from CB to SB, as shown in Fig. 3(a). For anisotropy $K_2 > 0$, the peak grows and broadens with increasing temperature. Since deep in the SB regime the current remains small, a broad region of NDC develops. This is remarkable, since NDC, which has already been observed or predicted for many systems, is usually a low-temperature effect. The high-temperature NDC is a distinct effect, since it appears even when there is no NDC at low temperatures.

Before we explain the high-temperature NDC, we note that the temperature dependence of the I - V curves is qualitatively different for vanishing anisotropy barrier and *large* exchange interaction, $K_2 \ll kT \ll J$, as shown in Fig. 3(b). Most striking is the fact that the maximum of the peaks stays constant for $T \rightarrow 0$. For this situation the energy levels are shown in Fig. 3(c). It is obvious that only a *single* energy difference is relevant as long as the higher $n=1$ quartet is not occupied. If $eV/2$ exactly equals this transition energy, the

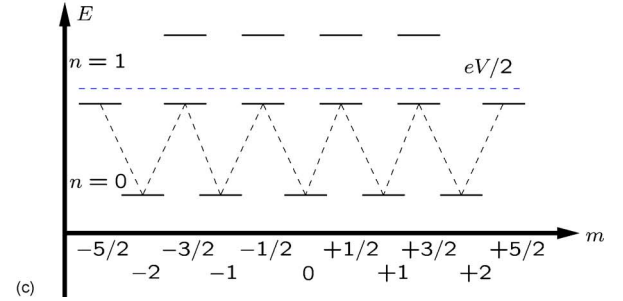
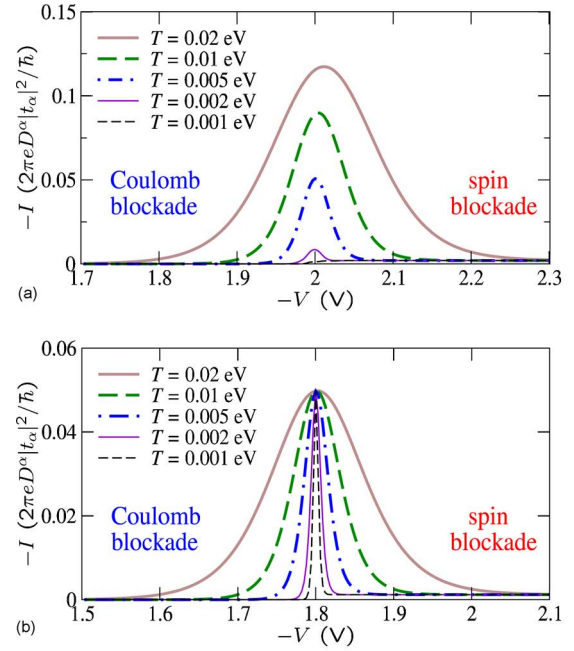


FIG. 3. (Color online) Temperature dependence of the current-voltage characteristics at high temperatures for vanishing magnetic field, exhibiting NDC. In (a) we assume $K_2 = J = 5$ meV, corresponding to the spin multiplets schematically shown in Figs. 1 and 2 except that now $B=0$. In (b) we assume a large exchange interaction $J=0.1$ eV and vanishing magnetic anisotropy $K_2=0$. (c) Energy level scheme for the $n=0$ and $n=1$ multiplets as a function of the magnetic quantum number m for the case in (b).

Fermi functions in the rates, Eq. (5), for these transitions all equal $1/2$ and are thus independent of temperature. Consequently, the steady-state probabilities and current are also independent of temperature at this bias. For lower bias all transitions from $n=0$ to $n=1$ are thermally suppressed and for higher bias the relevant transitions become fully active (the Fermi functions approach unity) and we enter the SB regime discussed above. In both regimes the current decreases for $T \rightarrow 0$.³² In the SB regime this leads to NDC. It is remarkable that the SB, a quantum effect, leads to large NDC at room temperature.

For the case of $K_2 > 0$ and, more importantly, small J , Fig. 3(a), the current at the maximum increases with temperature, since transport through the higher $n=1$ quartet contributes more and more. For the same reason the current maximum shifts to larger bias voltages. On the other hand, the current maximum vanishes for $T \rightarrow 0$. In Fig. 3(a) we have assumed vanishing magnetic induction and there is no window of en-

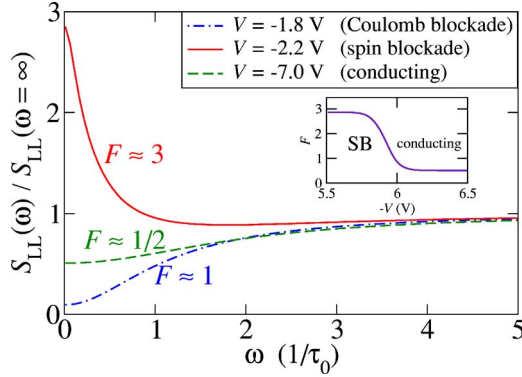


FIG. 4. (Color online) Current noise spectrum. Shown are the normalized correlation functions $S_{LL}(\omega)/S_{LL}(\omega=\infty)$ for three different bias voltages representing the three transport regimes: $V = 1.8$ V (CB), $V = 2.2$ V (SB), and $V = 7.0$ V (conducting regime). The zero-frequency limit gives the Fano factor $F = S(\omega \rightarrow 0)/2e|I|$ which tends to 3 in the SB regime, if we assume the one lead to be half metallic, $D_{\downarrow}^R/D_{\uparrow}^R \rightarrow 0$.

hanced current at low temperatures: At small bias, in the CB regime, the steady state has predominantly $n=0$ and $m=-S$ due to the asymmetric tunneling rates into the right lead. As the bias is increased the system directly crosses over to the SB regime when the bias equals the excitation energy to the state with $n=1$ and $m=-S-1/2$.

The shape of the NDC peaks in Fig. 3(b) for vanishing anisotropy can be fitted by a function of the form $1/\cosh^2 x$: Coming from the CB regime, the increase of the bias voltage energetically enables tunneling processes with a rate proportional to the Fermi function $f[(E-eV/2)/kT]$, where E denotes the excitation energy. However, the probability for the molecule being trapped in the SB state is also proportional to $f[(E-eV/2)/kT]$. Therefore, the current also contains a factor of the form $(1-f[(E-eV/2)/kT])+C$, where the first term accounts for the probability of the molecule *not* being trapped and the constant C describes the small tunneling probability for an electron with the “wrong” spin. C is determined by the ratio of densities of states $D_{\downarrow}^R/D_{\uparrow}^R$ in the ferromagnetic lead. Altogether, we obtain $I \sim 1/\cosh^2[(E-eV/2)/2kT] + f[(E-eV/2)/kT]C$, which is dominated by the $1/\cosh^2 x$ term for small $D_{\downarrow}^R/D_{\uparrow}^R$. This should be compared to Ref. 34, where the shape of the peaks in the differential conductance due the opening of additional transport channels is considered. In our case the peak occurs in the current.

Note that this simple argument does not apply to the NDC peaks shown in Fig. 3(a). For nonzero anisotropy K_2 , the ground state is no longer degenerate and the various transitions between states of the $n=0$ and $n=1$ multiplets have different energies, which leads to a more complex line shape.

Experimentally, the current suppression originating from SB could be distinguished from other mechanisms by investigating the shot noise of the system.³³ Figure 4 shows the current noise spectrum for three different bias voltages representing the CB, SB, and conducting regimes. The latter is here reached by applying a large negative bias, which overcomes the SB by allowing conduction through doubly occu-

pled states. We here consider the normalized correlation function $S_{LL}(\omega)/S_{LL}(\omega \rightarrow \infty)$ which gives information about temporal correlations of tunneling events for the left lead. According to Eq. (8), $S_{LL}(\omega)$ is the Fourier transform of the current-current correlation function. The constant part, $S_{LL}(\omega \rightarrow \infty)$, comes from the autocorrelation contribution, which is a positive δ function at vanishing time difference. In the conducting regime the noise spectrum exhibits a minimum at zero frequency, since the corresponding contribution to the correlation function is negative. Whenever the molecule is doubly occupied, further electrons cannot tunnel in due to the Pauli principle. This is the usual “antibunching” effect for fermions. The typical frequency scale apparent in Fig. 4 for the conducting case is the inverse of the typical time the molecule remains doubly occupied, which is of the order of the typical tunneling time $\tau_0 \equiv (2\pi|t_a|^2 D^L v_{uc}/\hbar)^{-1}$.¹⁶

In the CB regime, Coulomb repulsion hinders electrons from entering the molecule and the Fano factor $F \equiv S(\omega=0)/2e|I|$, which is defined as the ratio of the zero-frequency noise to the classical Schottky result,³³ is close to unity. In this case, single electrons tunnel through the system in rare, uncorrelated events, and quantum correlations are unimportant.

In contrast, in the SB regime the noise is *enhanced* at $\omega=0$. This maximum results from a *bunching* of the charge carriers, which means that several electrons tend to tunnel through the molecule within a short time interval. On the other hand, the average waiting time between such events is comparatively long. Results for the Fano factor reveal super-Poissonian shot noise, $F > 1$, in the SB regime. It reaches the value $F \approx 3$ for one halfmetallic lead, $D_{\downarrow}^R/D_{\uparrow}^R \rightarrow 0$. This factor can be understood by taking into consideration that the Fano factor contains information about the charge of the current-carrying particles and the quantum correlations between them. In the SB regime, the molecule is in the singly occupied state and has minimal spin for most of the time. Electrons from the left lead cannot hop onto the molecule until a spin-down electron is emitted into the right lead. The rate for this process is strongly suppressed. However, if the spin-down electron does leave the molecule, the probability for further tunneling processes is high, since electrons of both spin directions may then tunnel into the molecule. A current is flowing until a spin-down electron occupies the molecule again, which leads to the SB state. The Fano factor can be obtained from the following expression:³⁵

$$F = \langle N \rangle \frac{\langle t^2 \rangle - \langle t \rangle^2}{\langle t \rangle^2} + \frac{\langle N^2 \rangle - \langle N \rangle^2}{\langle N \rangle}. \quad (10)$$

Here N denotes the number of electrons tunneling through the molecule in one “bunch” and t the waiting time between such processes. Since the molecule is in the same SB state between the events, they are uncorrelated, giving an exponential distribution of times t . Since the tunneling is dominated by the three states with $n=0$, $m=-S$, and $n=1$, $m=-S \pm 1/2$, one finds a probability of $1/2^N$ for N electrons in the bunch. This leads to $\langle N \rangle = 2$ and $\langle N^2 \rangle = 6$. Since $\langle t^2 \rangle = 2\langle t \rangle^2$ for an exponential distribution, Eq. (10) yields $F = 3$ for the Fano factor. This result is of the same fundamental

origin as the super-Poissonian shot noise found for quantum dots in the SB regime in Ref. 25, which considers the case $K_2=J=0$ (no local spin) and nonmagnetic leads. In this case the SB is induced by application of a magnetic field.²⁵ We note that Eq. (10) also gives the observed value $F=1/2$ for the conducting regime.

Buřka *et al.*³⁷ have investigated a similar system, consisting of a quantum dot connected to two electrodes, where the dot and one of the leads are nonmagnetic while the second lead is ferromagnetic. In such an F - N - N junction, a *large* spin can be accumulated on the dot. This spin accumulation causes NDC and strongly enhances the current shot noise due to spin noise activated in the NDC regime.³⁷ In contrast, super-Poissonian shot noise in transport through anisotropic magnetic molecules arises from an interplay of spin-flip processes involving a small local spin and spin selection rules.

As noted above, giant spin amplification can occur in transport through anisotropic magnetic molecules coupled to *nonmagnetic* leads.¹⁶ At bias voltages close to the CB threshold the total spin transmitted from one lead to the other can become exponentially large at low temperatures, if the molecule is prepared in a magnetically polarized initial state at time $t=0$. If one lead is *ferromagnetic* the steady-state current is highly spin-polarized so that it is necessary to consider instead the *excess* transmitted spin. Analogously, one can define the *excess transmitted charge*: The molecule is prepared in a specific state $|n\rangle$ at time zero and then evolves according to the rate Eqs. (6). Since it approaches the steady state exponentially, the excess charge

$$\Delta Q_n^\alpha \equiv \int_0^\infty dt [I^\alpha(t) - \langle I^\alpha \rangle] \quad (11)$$

is finite. The main observation is that ΔQ_n^α can depend very strongly on the initial state $|n\rangle$ if the steady state shows SB. Practically it is much easier to measure the charge accumulation in the leads instead of the excess spin,³⁶ since the accumulated charge is conserved (except for leakage currents) whereas the spin is not. Moreover, it should be easier to employ the excess charge for further data processing.

Results for the excess transmitted charge as a function of bias voltage are shown in Fig. 5(a), where an initial state with $n=0$ and spin $m=S$ is assumed. The fine structure close to the CB threshold basically originates from the anisotropy of the local spin. Most striking is the exponential enhancement of ΔQ_n^α at voltages *above* the CB threshold, where the steady-state current is already nonzero (but still small due to SB). When the bias is just large enough to allow the two transitions from the $n=0$ state with extremal spin to the $n=1$ state with extremal spin, $m=\pm S \rightarrow \pm(S+1/2)$, the system initially prepared in a state with *maximal* spin, $m=S+1/2$, for an exponentially long time only performs transitions between the two extremal states connected by a solid line in the level scheme in Fig. 5(b). Thus a sizable, and completely spin-polarized, current is flowing until the molecule overcomes the anisotropy barrier by thermal activation. After that it rapidly relaxes towards the SB state, which shows a small steady-state current. On the other hand, if the molecule is prepared in the state with minimal spin, $m=-S-1/2$, it al-

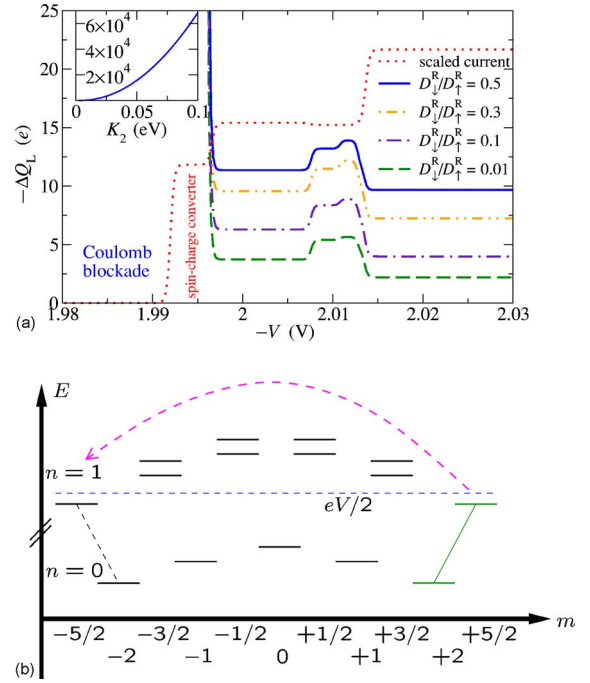


FIG. 5. (Color online) (a) Excess transmitted charge ΔQ^L as a function of bias voltage in the vicinity of the first CB step for different polarizations of the right lead: $D_\downarrow^R/D_\uparrow^R=0.5, 0.3, 0.1$, and 0.01 . We assume $J=4$ meV, $K_2=1$ meV, vanishing magnetic field and an initial state with $n=0$ and $m=2$. The steady-state current for $D_\downarrow^R/D_\uparrow^R=0.1$ is also shown. The inset shows ΔQ^L as a function of the anisotropy constant K_2 , where the bias voltage V corresponds to the arithmetic mean of the first two excitation energies. (b) Energy level scheme of the $n=0$ and $n=1$ multiplets. The molecule is prepared in the initial state with spin $m=S$.

ready starts out in the SB state and the current is always small. Thus in the first case an exponentially large excess charge is accumulated in the leads in addition to the excess spin. In this regime the proposed setup functions as a *spin-charge converter*, i.e., *spin* information can be read through a measurement of the excess transmitted *charge*, a much easier task.

On the other hand, if the bias is large enough to allow further transitions involving other levels, the system rapidly relaxes back towards SB so that ΔQ_n^α is of order e . Note that the excess transmitted charge increases for increasing anisotropy of the local spin, as can be seen from the inset in Fig. 5(a).

IV. SUMMARY AND CONCLUSIONS

We have studied transport through a single anisotropic magnetic molecule coupled to one ferromagnetic and one nonmagnetic lead. Our main findings concern the NDC, super-Poissonian noise, and a strong spin dependence of transmitted charge.

Two types of NDC occur. One appears at *low* temperatures in an external magnetic field. Here the Coulomb blockade and spin blockade regimes are separated by a finite window of bias voltages for which the current is strongly

enhanced. This is due to the interplay of bias voltage, magnetic field, and magnetic anisotropy, which allows only transitions between two specific molecular many-body states and prevents the spin flips necessary for spin blockade. The other NDC effect appears at *high* temperatures in the vicinity of the Coulomb blockade threshold. This is a distinct effect—it can occur even when there is no NDC at low temperatures. It can be viewed as a manifestation of quantum effects (the Pauli principle and spin selection rules) at room temperature.

Spin blockade is accompanied by super-Poissonian shot noise, as found earlier for nonmagnetic quantum dots. Furthermore, the total charge transmitted through a molecule

under a voltage bias depends strongly on its initial spin state in a certain parameter range. The difference in transmitted charge can diverge exponentially for low temperatures. This spin-charge conversion presents a promising method to read out the spin information in molecular-memory applications.

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