Theory of frequency-dependent spin current noise through correlated quantum dots

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We analyze the equilibrium and nonequilibrium frequency-dependent spin current noise and spin conductance through a quantum dot in the local moment regime. Spin current correlations are shown to behave markedly differently from charge correlations: Equilibrium spin crosscorrelations are suppressed at frequencies below the Kondo scale and are characterized by a universal function that we determine numerically for T=0temperature. For asymmetrical quantum dots dynamical spin accumulation resonance is found at the Kondo energy, $\omega \sim T_K$. At higher temperatures surprising low-frequency anomalies related to overall spin conservation appear.

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I. INTRODUCTION

Coherent detection and manipulation of spin currents in nanostructures has recently attracted wide attention due to possible promising applications in future storage technologies and quantum computing.^{1,2} Many proposals have been made to build spin batteries to inject spin-polarized current, and then filter, manipulate, and detect it.³ Often one makes use of ferromagnetic electrodes in these circuits⁴ while in other cases the application of an external magnetic field⁵ or the presence of a ferromagnetic resonance process⁶ enables one to filter and detect spin currents. Quantum dots play a special and important role in this regard: in these devices, the strong electron-electron interaction enables one to manipulate the spin of a single electron⁷ and such quantum-dot devices provide a possible route to quantum computing.⁸

However, to use spin circuits efficiently, it would be of crucial importance to characterize the noise in them. In addition, the structure of the noise provides valuable information on interactions and correlations. In fact, a lot of attention has been devoted to noise analysis in correlated mesoscopic circuits for this reason.⁹⁻¹¹ Due to progress in experimental technology, it is now possible to measure ac conductance properties as well as frequency-dependent noise in these circuits down to very low temperatures and even in the Kondo regime.^{12–14} Furthermore, with efficient spin-filtering methods³ measuring spin-resolved current noise in such circuits is also within reach. Surprisingly, while a lot is known about the properties of ordinary noise in the correlated regime, much less is known about the structure of spin current noise. So far, only spin correlations in the sequential tunneling^{15,16} and perturbative regimes¹⁷ have been analyzed, and these works focused almost exclusively on shot noise.

Here we carry out a detailed analysis of the full frequency spectrum of the spin-dependent current noise in the Kondo regime. We show that equilibrium spin current correlations are characterized by two universal functions, which we determine numerically for T=0 temperature using the method of numerical renormalization group (NRG),¹⁸ and compute analytically for large frequencies. At finite temperatures, we

analyze spin correlations using a perturbative approach. We find in all regimes that correlations between electrons of the same and opposite spins behave markedly differently. In the perturbative regime these remarkable differences emerge at frequencies below the Korringa relaxation rate: while a dip appears in the frequency-dependent noise of opposite spin directions, a large peak develops for the parallel spin components. These surprising features are all intimately related to spin conservation.

II. MODEL

Focusing on the Kondo regime, we shall assume that there is a single spin S=1/2 electron on the quantum dot, which couples to the electrons on the leads through the Kondo interaction¹⁹

$$H_{\rm int} = \sum_{r,r'=L,R} \sum_{\sigma,\sigma'} \frac{j}{2} v_r v_{r'} \mathbf{S} \psi_{r\sigma}^{\dagger} \sigma_{\sigma\sigma'} \psi_{r'\sigma'}.$$
 (1)

Here σ stands for the three Pauli matrices, the fields $\psi_{r\sigma} = \int_{-D}^{D} c_{r\sigma}(\varepsilon) d\varepsilon$ destroy electrons of spin σ in leads $r \in \{L, R\}$, and their dynamics are governed by the noninteracting Hamiltonian, $H_0 = \sum_{r\sigma} \int \varepsilon c_{r\sigma}^{\dagger}(\varepsilon) c_{r\sigma}(\varepsilon) d\varepsilon$.²⁰ The coupling *j* in Eq. (1) is the usual dimensionless coupling, which incorporates already the density of states in the leads, and is related to the Kondo temperature as $T_K \approx De^{-1/j}$, with *D* the cut-off energy appearing in $\psi_{r\sigma}$. The dimensionless hybridization parameters are given by $v_L = \cos(\phi/2)$ and $v_R = \sin(\phi/2)$ with ϕ parametrizing the asymmetry of the dot: $\phi = \pi/2$ corresponds to a symmetrical quantum dot with maximum transmittance.

III. EQUILIBRIUM NOISE

In view of the special structure of Eq. (1), it is natural to introduce the "even" and "odd" linear combinations, $\Psi \equiv \cos(\phi/2)\psi_L + \sin(\phi/2)\psi_R$, and $\tilde{\Psi} \equiv \sin(\phi/2)\psi_L - \cos(\phi/2)\psi_R$. Although only Ψ couples to the spin in H_{int} , changing the chemical potential in one of the leads couples the fields $\tilde{\Psi}$ and $\Psi,$ and both contribute to the spin noise.

To compute the noise, we first define the spin component σ of the current in lead *r* through the equation of motion, $J_{r\sigma} \equiv e\dot{N}_{r\sigma} = ei[H_{\text{int}}, N_{r\sigma}]$. The corresponding current is found to have two distinct (even and odd) parts, $J_{r\sigma} = I_{r\sigma} + \tilde{I}_{r\sigma}$, with

$$\begin{split} I_{r\sigma} &= ej \gamma_r i (F_{\sigma}^{\dagger} \Psi_{\sigma} - \Psi_{\sigma}^{\dagger} F_{\sigma}), \\ \widetilde{I}_{r\sigma} &= ej \widetilde{\gamma}_r i (F_{\sigma}^{\dagger} \widetilde{\Psi}_{\sigma} - \widetilde{\Psi}_{\sigma}^{\dagger} F_{\sigma}) \end{split} \tag{2}$$

and the prefactors defined as $\gamma_{L/R} = [1 \pm \cos(\phi)]/4$ and $\tilde{\gamma}_{L/R} = \pm \sin(\phi)/4$. The operator $F_{\sigma} = (\mathbf{S}\sigma\Psi)_{\sigma}$ denotes the so-called composite fermion operator²¹ and represents the universal (Kondo) part of the dot electron.

The operator identity, $I_{r\uparrow} + I_{r\downarrow} = 0$, and the simple even-odd decomposition of $J_{r\sigma}$ imply that, in equilibrium and in the absence of external magnetic field, the 16 components of the symmetrized noise $S_{rr'}^{\sigma\sigma'} = \frac{1}{2} \langle \{J_{r\sigma}(t), J_{r'\sigma'}(0)\} \rangle$, depend on just two universal functions, *s* and *s*. Maybe the most interesting left-right noise component, $S_{LR}^{\sigma\sigma'}$, can be expressed, e.g., as

$$S_{LR}^{\sigma\sigma'}(\omega) = -\frac{e^2}{2\pi}T_K\sin^2(\phi)[\delta_{\sigma\sigma'}\tilde{s}(\omega) + \sigma\sigma's(\omega)],$$

where $e^2/2\pi = e^2/h$ denotes the universal conductance unit, and the dimensionless functions *s* and *s* depend exclusively on the ratios ω/T_K and T/T_K . The function *s* is related to the even current component, and it governs the correlations between spin-up and spin-down carriers, however, its contribution cancels in the charge noise and charge conductance, which are exclusively determined by the odd component of the current, incorporated in *s*.

In equilibrium, the fluctuation-dissipation theorem relates $S_{rr'}^{\sigma\sigma'}(\omega)$ to the real part of the spin conductance through the dot, Re $G_{rr'}^{\sigma\sigma'}(\omega) = -\frac{1}{\omega} \operatorname{coth}(\omega/2T) S_{rr'}^{\sigma\sigma'}(\omega)$, which can therefore also be expressed in terms of two dimensionless universal conductance functions, $g(\omega, T)$ and $\tilde{g}(\omega, T)$. The left-right conductance, e.g., reads

Re
$$G_{LR}^{\sigma\sigma'}(\omega) = \frac{e^2}{2\pi} \sin^2(\phi) [\delta_{\sigma\sigma'} \tilde{g}(\omega, T) + \sigma\sigma' g(\omega, T)].$$

Using tedious but straightforward manipulations, we can express g and \tilde{g} in terms of the spectral functions $\mathcal{Q}_F(\omega, T)$ and $\mathcal{Q}_{\mathcal{I}_{\sigma}\mathcal{I}_{\sigma'}}(\omega, T)$ of the composite fermion and of the "current" operator $\mathcal{I}_{\sigma} \equiv i(F_{\sigma}^{\dagger}\Psi_{\sigma} - \Psi_{\sigma}^{\dagger}F_{\sigma})$

$$\widetilde{g}(\omega,T) = \frac{1}{2\omega} \int d\omega' \frac{\varrho_F(\omega',T)}{\varrho_F(0)} [f(\omega'-\omega) - f(\omega'+\omega)],$$
$$g(\omega,T) = -\frac{1}{2\omega\varrho_F(0)} \varrho_{\mathcal{I}_{\uparrow}\mathcal{I}_{\uparrow}}(\omega,T)$$
(3)

with $f(\omega)$ denoting the Fermi function.²² Since F_{σ} and \mathcal{I}_{σ} are local operators, we can compute g and \tilde{g} (and thus s and \tilde{s}) by using the powerful method of NRG.^{18,23}

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FIG. 1. (Color online) Zero-temperature universal functions s and \tilde{s} computed by NRG. Inset: universal spin-conductance functions g and \tilde{g} .

IV. T=0, EQUILIBRIUM RESULTS

The T=0 temperature universal functions $\tilde{s}(\omega/T_K)$ and $s(\omega/T_K)$, and the conductance functions $g(\omega/T_K)$ and $\tilde{g}(\omega/T_K)$ are displayed in Fig. 1. The high-frequency behavior of *s* and \tilde{s} can be captured by doing perturbation theory in *j* and summing up the leading logarithmic corrections to give

$$\widetilde{s}(\omega/T_K) \approx -\frac{3}{2}s(\omega/T_K) \approx \frac{3\pi^2}{16} \frac{|\omega|}{T_K} \frac{1}{\ln^2(|\omega|/T_K)}$$

for $\omega \gg T_K$. Though they look similar at high frequencies, s and s behave markedly differently in the Fermi-liquid regime, $\omega \ll T_K$, where $\widetilde{s} = \widetilde{\alpha} |\omega| / T_K + \cdots$ while $s = \alpha (|\omega|/T_K)^3 + \cdots$ with α and $\tilde{\alpha}$ universal constants of the order of unity. The ω^3 scaling of s is related to spin conservation: in the absence of external spin-relaxation mechanism, the total number of spin-up electrons can fluctuate between two values, N_{\uparrow} and N_{\uparrow} + 1. Since the spin-up electrons couple to the spin-down electrons only at a single point (the quantum dot), no steady spin current can be generated for the spin down electrons by injecting spin-up electrons in one of the leads. Thus the spin conductance $G_{rr'}^{\uparrow\downarrow}(\omega)$ must vanish at $\omega = 0$, and by analyticity, $G_{rr'}^{\uparrow\downarrow} \sim \omega^2$. In equilibrium, however, the spin current noise is simply related to the spin conductance by the fluctuation-dissipation theorem, implying a $|\omega|^3$ scaling of s at T=0. This argument carries over to finite temperatures too, where it leads to an asymptotic behavior, $s \sim T\omega^2$ in the absence of external spin relaxation. We should emphasize that, in our calculations, spin relaxation is due to the interaction part of the Hamiltonian which, however, conserves the total spin, and leads to the vanishing of $\uparrow \downarrow$ spin noise component at $\omega=0$. Introducing some source of an external spin relaxation, however, leads to a violation of spin conservation, and amounts in a finite $S_{LR}^{\uparrow\downarrow}(\omega=0) \neq 0.^{22}$

The fundamental difference between $\uparrow\uparrow$ and $\uparrow\downarrow$ correlations shows up even more strikingly in the spin conductance (see Fig. 1): while $G_{LR}^{\uparrow\uparrow}(\omega)$ is dominated by $\tilde{g}(\omega)$ and behaves qualitatively the same way as the conductance through the dot, $G_{LR}^{\uparrow\downarrow}(\omega) \sim g(\omega)$ exhibits a resonance at a frequency $\omega \approx 0.5T_K$.²⁴ This can be understood in a simple and intuitive way: the spin conductance $\uparrow\downarrow$ is generated by flips of the localized spin. For $\omega > T_K$, the coupling to the conduction electrons gets stronger with decreasing ω and increases the conductance. At very small energy scales, $\omega \ll T_K$, however,

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FIG. 2. Diagrams contributing to the noise. The reduced density matrix of the spin evolves along the upper and lower Keldysh contours. Triangles denote the bare current vertices and dots indicate the exchange interaction. Arrows correspond to conduction electron propagators. To leading order, the current-current correlation function is given by the connected diagrams (top), $S_{\rm conn}(\omega)$, and the "disconnected" diagrams (second line), $S_{\rm disc}(\omega)$, with Π the propagator describing the evolution of the reduced density matrix of the spin (third line). The dressed current vertex $W_{r\sigma}$ is given by diagrams similar to those of the self-energy, Σ (last line), with one of the dots replaced by a triangle.

the impurity spin is quenched, and with the above mechanism being absent, the $\uparrow\downarrow$ conductance must vanish.

V. $T \neq 0$, PERTURBATIVE REGIME

Computation of the finite-temperature noise requires care: Usual finite-temperature NRG broadening procedures lead to an unphysical finite linear coefficient for $s(\omega)$, conflicting with our exact finite T result, $s(\omega) \sim \omega^2$. Therefore, for $T \neq 0$, other methods must be used. For $T \gg T_K$, we carried out a systematic expansion in j for the time dependence of the reduced density matrix of the spin and the spin current noise using the formalism of Refs. 25 and 26. Details of this involved calculation shall be published elsewhere,²² here we just outline the main results.

Naively, to calculate the noise in leading order, one would just compute the first (connected) noise diagram of Fig. 2, $S_{\rm conn}(\omega)$. This diagram accounts for short time current correlations mediated by electron-hole excitations in the leads, and dominates indeed the noise at high and intermediate frequencies, $\omega \gtrsim T$. At small frequencies, however, a resummation of the perturbation series is necessary, because there the "disconnected" contribution, $S_{disc}(\omega)$, turns out to be of the same order in j as $S_{\text{conn}}(\omega)$, and becomes also important: This contribution accounts for correlations between subsequent incoherent tunneling processes, generated by the impurity spin itself. These correlations are due to the mere fact that a spin-flip process where a conduction electron's spin is flipped from up do down, $\uparrow \rightarrow \downarrow$, must be followed by a process $\downarrow \rightarrow \uparrow$. To account for them, one needs to solve a Dyson equation for the propagator Π of the reduced density matrix





FIG. 3. (Color online) Top: sketch of consecutive spin-flip processes. Bottom: equilibrium and nonequilibrium noise spectra $S_{LR}^{\sigma\sigma'}(\omega)$ in the perturbative regime, max $\{T, \omega, E_K\} > T_K$, as computed from a diagrammatic approach. In the nonequilibrium case a simple current bias was assumed, $V_L^{\sigma} \equiv V$ and $V_R^{\sigma} \equiv 0$, while j=0.07 and $\phi=\pi/2$ in both cases.

of the spin, as sketched in Fig. 2. In this approach, spin relaxation is characterized by the relaxation rate, $\Gamma(\omega)$, appearing in the self-energy Σ of the propagator Π (Ref. 25)

$$\Gamma(\omega) = T \sum_{r,r'} j^2 v_r^2 v_{r'}^2 \hat{L}\left(\frac{\omega}{T}, \frac{\mu_r - \mu_{r'}}{T}\right),\tag{4}$$

where Re $\hat{L}(x,y) = \pi y + \pi [x \operatorname{sh}(x) - y \operatorname{sh}(y)] / [\operatorname{ch}(x) - \operatorname{ch}(y)]$ and Im $\hat{L}(x,y) = \frac{1}{\pi} \int dx'$ Re $\hat{L}(x',y) / (x-x')$. In the $\omega \to 0$ limit, $\Gamma(\omega)$ can be identified as the Korringa relaxation rate, $E_K \equiv \Gamma(0)/2$, of the impurity spin, which for a simple voltage-biased quantum dot reads

$$E_K = \pi j^2 T \left[\frac{1 + \cos\phi}{2} + \frac{1 - \cos\phi}{2} \frac{eV}{2T} \operatorname{coth} \frac{eV}{2T} \right]$$

In the voltage-biased case, we can express the left-right component of the spin current noise in a compact form

$$S_{LR}^{\sigma\sigma'}(\omega) = -\frac{e^2}{2\pi} \operatorname{Re} \left\{ \frac{\sigma\sigma'}{16} \frac{\Gamma^2(\omega) - R^2(\omega)}{-i\omega + \Gamma(\omega)/2} + \frac{3 - \sigma\sigma'}{32} Tj^2 \sin^2(\phi) \left[\hat{L}\left(\frac{\omega}{T}, \frac{V}{T}\right) + \hat{L}\left(\frac{\omega}{T}, \frac{-V}{T}\right) \right] \right\}$$
(5)

with $R(\omega) = j^2 \cos(\phi) \hat{L}(\omega/T, 0)$. The symmetrized noise is shown in Fig. 3: at high frequencies, $\omega \ge E_K$, the noise is dominated by the result of simple-minded perturbation theory, corresponding to the second line of Eq. (5). This part of the correlation function describes *short-time* correlations within a single tunneling process, generated by the dynamics of electron-hole excitations in the leads. However, at time scales $t \sim 1/\omega > 1/E_K$, consecutive incoherent tunneling processes start to correlate by the constraint mentioned before. These correlations are captured by the first term in Eq. (5), coming from the "disconnected" part of the noise (see Fig. 2). As a consequence, for $\omega < E_K$, a large dip appears in the noise component $S_{LR}^{\uparrow\downarrow}$, while a bump emerges in $S_{LR}^{\uparrow\uparrow}$. For zero bias, V=0, we find that $S_{LR}^{\uparrow\downarrow}(\omega=0, V=0)=0$, in agreement with the fluctuation-dissipation theorem and the observation that the linear spin conductance between spin-up and spin-down electrons must vanish. This has a simple physical explanation: a spin- \uparrow electron injected from the left can give rise to a spin- \downarrow outgoing electron on the right with a certain probability (see Fig. 3). However, before such a process occurs again, another spin-flip process must take place, where the dot spin is flipped back. In equilibrium, this second process (on the average) removes *exactly* the same amount of \downarrow spin from the right lead as injected in the first process. Therefore, no equilibrium $\uparrow \downarrow$ dc spin conductance is possible.

Remarkably, the above correlations only show up in the spin current noise, and cancel out in the charge current noise, $S_{LR} \equiv \sum_{\sigma,\sigma'} S_{LR}^{\sigma\sigma'}$. Being the result of rather classical correlations between subsequent incoherent processes, these low-frequency features can also be captured by a much simpler rate equation approach (see Refs. 17 and 22), which, however, is unable to account for the high-frequency part of the noise at $\omega > T$.

Although the above results are perturbative in *j*, they carry over to the whole regime $\max\{T, \omega, E_K\} > T_K$ with the small modification that *j* must be replaced by the renormalized coupling $j \rightarrow j(T, \omega, eV) \approx 1/\ln(\max\{T, \omega, E_K\}/T_K)$.

VI. FERMI-LIQUID REGIME

In the Fermi-liquid regime, $\omega, T, eV \ll T_K$, one can compute the spin current correlations by describing the dot in terms of scattering states that interact at the impurity site.

This is a rather cumbersome approach for finite frequencies. However, observing that correlations between spin-up and spin-down electrons are generated only through the residual electron-electron interaction, simple phase space arguments immediately give that the T=0 shot noise is just given by $S_{LR}^{\uparrow\downarrow}(V) = (e^2/2\pi)\gamma \sin^2(\phi)(eV)^3/T_K^2$, while for equilibrium we recover the numerically observed result, $S_{LR}^{\uparrow\downarrow}(\omega) = (e^2/2\pi)\alpha \sin^2(\phi)|\omega|^3/T_K^2$, with γ and α two universal numbers. The discussion of the finite temperature and finite frequency noise and the precise determination of these universal constants is very complicated, and shall be considered in a future publication.

VII. CONCLUSIONS

Analyzing the full frequency dependence of the spin current noise through a quantum dot in the Kondo regime we found that $\uparrow\downarrow$ correlations are strongly suppressed at frequencies below the Kondo temperature and below the Korringa relaxation rate as compared to $\uparrow\uparrow$ correlations due to overall spin conservation. In the $\uparrow\downarrow$ conductance a resonance is predicted at $\omega \sim T_K$. Observing these striking features is within reach with present-day noise-measurement techniques.

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