Probing the Kondo screening cloud via tunneling-current conductance fluctuations

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We show that conductance fluctuations or noise in the conductance of a tunneling current into an interacting electron system is dominated by density-density and (or) spin-spin correlations. This allows one to probe two-particle properties (susceptibilities) and collective excitations by standard experimental tunneling methods. We demonstrate this theoretically, using a many-body calculation for the single-adatom Kondo problem. An example of the two-particle correlations around a single magnetic adatom in the Kondo regime, as would be viewed by a scanning tunneling microscope, is given. The spatial dependence of the local spin and charge correlations of the substrate exhibits a clear signature of the Kondo screening cloud.

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Noise spectroscopy has become a valuable tool to study electronic systems. The intrinsic noise, i.e., the fluctuations of a signal due to inherent uncertainties generated by an electronic system is not a simple set of random uncorrelated events but contains fundamental information about electron-electron correlations, that is not reflected in a time-averaged measurement. A famous application was the experimental verification of the fractionally charged quasiparticles in the quantum-Hall regime, through shot-noise measurements.[1,2] The use of current-current correlations, such as shot-noise or Johnson-Nyquist (thermal) noise, has a long history and has been a prevalent topic of both experimental and theoretical work, along with related topics, such as conductance fluctuations of mesoscopic wires, mostly studying the effects of disorder.[3–5]

In this Brief Report, we show that in the weak local-tunneling limit, the conductance fluctuations of a tunneling current into an interacting system are determined by *local* density-density and spin-spin correlations. While on a macroscopic scale two-particle properties, such as the compressibility or magnetic susceptibility, are easily measured, few (if any) techniques exist to extract these quantities locally at a microscopic scale. For instance, almost 50 years after the discovery and explanation of the Kondo effect, the prediction of spatially extended spin correlations around a magnetic impurity—the Kondo cloud—has never been observed experimentally. This screening cloud and the associated screening length ξ_K, has attracted a lot of attention. Renormalization-group analysis[6,7] confirms the presence of such a scale but experiments to detect the screening cloud through the Knight shift[8] have not done so. Although these results are somewhat controversial, other proposed experiments have been difficult or impossible to realize.

Here, we propose to use a scanning tunneling microscope (STM) to measure the spatial dependence of the conductance fluctuations and to extract information on the spin-spin correlations. We demonstrate that a clear signature of the Kondo screening cloud can indeed be obtained via a single local probe, which we expect to stimulate future experimental work along these lines.

The atomic spatial resolution of a scanning tunneling microscope makes it a natural choice to study systems on a microscopic scale. The combination of an STM and noise spectroscopy has already been used to develop the field of electron-spin-resonance scanning tunneling microscopy.[9] With a similar experimental setup in mind, the total Hamiltonian is taken to be $H=H_{\rm STM}+H_{\rm sub}+H_{\rm tun}$, where $H_{\rm sub}$ is the general interacting Hamiltonian of the substrate and $H_{\rm STM}=\Sigma_{\mathbf{k}\sigma}(\epsilon_{\mathbf{k}\sigma}-\mu-eV)a^{\dagger}_{\mathbf{k}\sigma}a_{\mathbf{k}\sigma}$ is the Hamiltonian of the STM. As usual, we assume that the STM is a noninteracting Fermi gas with an energy-independent density of states near the Fermi energy $\epsilon_F=\mu$. The chemical potential of the STM is displaced by eV, where the charge of the electron is $-e$ and V is the applied voltage. The tunneling is determined by $H_{\rm tun}=\Sigma_{\mathbf{k},\mathbf{k}',\sigma}[Ta^{\dagger}_{\mathbf{k}\sigma}b_{\mathbf{k}'\sigma}+\text{H.c.}]$, with tunneling amplitude T, which within the local tunneling approximation is taken to be independent of momenta $\mathbf{k},\mathbf{k}'$. The operators $b^{\dagger}_{\mathbf{k}'\sigma}$ and $b_{\mathbf{k}'\sigma}$ are the mode creation and annihilation operators for the substrate. The current operator is defined as $\hat{I}=-e\partial_t\hat{N}_{\rm STM}$, where $\hat{N}_{\rm STM}=\Sigma_{\mathbf{k}\sigma}a^{\dagger}_{\mathbf{k}\sigma}a_{\mathbf{k}\sigma}$ is the particle number operator for the STM. Assuming $[\hat{N}_{\rm STM},H_{\rm sub}]=0$, by Heisenberg's equation of motion, $\hat{I}=ie[\hat{N}_{\rm STM},H_{\rm tun}]$ (with $\hbar=1$).

Evaluating the commutator within the tunneling Hamiltonian formalism leads to the common expression for the current operator $\hat{I}=ie\Sigma_{\sigma}\Sigma_{\mathbf{k},\mathbf{k}'}[Ta^{\dagger}_{\mathbf{k}\sigma}b_{\mathbf{k}'\sigma}-\text{H.c.}]$.[10] The non-equilibrium expectation value of $\hat{I}$, which determines the experimentally measured current, is obtained within linear response (LR) by treating the tunneling as the perturbation and assuming the STM and substrate are separately in thermodynamic equilibrium. If the system is decoupled in the infinite past, the current operator within LR is given as

$$\hat{I}_{\rm LR}(t)=\hat{I}+i\int_{-\infty}^{t}dt'[H_{\rm tun}(t'),\hat{I}(t)], \tag{1}$$

where $\hat{O}(t)=e^{iH_0t}\hat{O}e^{-iH_0t}$ and $H_0=H_{\rm STM}+H_{\rm sub}$. The expectation value of Eq. (1) with respect to H_0, $\langle\hat{I}_{\rm LR}\rangle_{H_0}=\text{Tr}\,\hat{I}_{\rm LR}e^{-\beta H_0}/\text{Tr}\,e^{-\beta H_0}$, gives the current to leading order in the tunneling amplitude T. The linear conductance, in this approximation, is given by $G_{\rm LR}=\partial_V\langle\hat{I}_{\rm LR}\rangle_{H_0}$. We define a conductance operator by $\hat{G}_{\rm LR}=\partial_V\hat{I}_{\rm LR}$, such that $\langle\hat{G}_{\rm LR}\rangle_{H_0}=\partial_V\langle\hat{I}_{\rm LR}\rangle_{H_0}$. With this operator expression for the conductance, the fluctuations or specifically the spectral density of the conductance, is defined as

$$S(\mathbf{r},\omega) = \frac{1}{2}\int dt e^{i\omega t}\langle\{\delta\hat{G}_{\text{LR}}(\mathbf{r},t),\delta\hat{G}_{\text{LR}}(\mathbf{r},0)\}\rangle_{H_0}, \quad (2)$$

where $\delta\hat{G}_{\text{LR}}=\hat{G}_{\text{LR}}-\langle\hat{G}_{\text{LR}}\rangle$. A lengthy calculation assuming rotational invariance leads to the following result for the low-temperature zero-frequency limit of Eq. (2):

$$S(\mathbf{r},\omega=0) = \pi^2 e^4|\mathrm{T}|^4[\rho_{\text{STM}}(eV)]^2\chi^{\text{ch}}_{\text{sub}}(\mathbf{r},\omega=0) + 32\pi^2 e^4|\mathrm{T}|^4 P^2[\rho_{\text{STM}}(eV)]^2\chi^{\text{sp}}_{\text{sub}}(\mathbf{r},\omega=0) \quad (3)$$

with $\rho_{\text{STM}}=\Sigma_\sigma\rho^\sigma_{\text{STM}}$, $P=(\rho^\uparrow_{\text{STM}}-\rho^\downarrow_{\text{STM}})/\rho_{\text{STM}}$, and

$$\chi^{\text{ch}}_{\text{sub}}(\mathbf{r},t) = \langle\delta\hat{n}(\mathbf{r},t)\delta\hat{n}(\mathbf{r},0)\rangle_{H_{\text{sub}}}, \quad (4a)$$

$$\chi^{\text{sp}}_{\text{sub}}(\mathbf{r},t) = \langle\hat{s}^z(\mathbf{r},t)\hat{s}^z(\mathbf{r},0)\rangle_{H_{\text{sub}}}, \quad (4b)$$

where $\chi^{\text{ch}}_{\text{sub}}$ and $\chi^{\text{sp}}_{\text{sub}}$ are the local charge and spin susceptibilities, respectively, with the density and spin-density operators of the substrates $\hat{n}$ and $\hat{s}^z$.[11] The compressibility [Eq. (4a)] is given in terms of the density variation; $\delta\hat{n}=\hat{n}-\langle\hat{n}\rangle$. From Eq. (3) one sees that for a spin-polarized STM, i.e., $P\neq 0$, the spin susceptibility may dominate, as the overall prefactor can be an order of magnitude larger. This comparison is of course system dependent as charge and spin susceptibilities can vary greatly. Equation (3) has the important consequence that in addition to the single-particle density of states routinely extracted from STM experiments, also two-particle correlations can be determined.

Note that in contrast to Eq. (2), the current noise in the weak tunneling limit, which is given by the current-current correlation function, leads to the well-known shot-noise relation. The zero-frequency shot noise is proportional to the current itself which goes as the tunneling amplitude squared while Eq. (3) is proportional to $|\mathrm{T}|^4$. This seemingly contradictory result is resolved by the fact that in general the complete characterization of the fluctuations or noise of a signal is not solely determined by a second-order moment, such as a current-current correlation but by all higher moments as well.[12,13] To obtain a similar result to Eq. (3) from the current signal itself requires the choice of a higher-order moment, i.e., the current-current-current-current correlation or kurtosis of the current noise. Thus, the conductance fluctuations of a tunneling current are related to the kurtosis.

Among the many possibilities, one could spatially resolve the low-energy correlations near one or more Kondo impurities, where the geometry of the nanocluster, as well as the direct and indirect exchange processes between atoms are in strong competition[14] with the Kondo correlations. Here, we present one intriguing application of this. We show that the correlations around a single magnetic atom, the so-called Kondo cloud, can be observed in such measurements. We now turn to a calculation of the local susceptibilities [Eqs. (4a) and (4b)] for such a system.

The Kondo effect has been extensively studied both theoretically and experimentally,[15] more recently by using an STM to image single or multiple magnetic adatoms on a metallic surface.[16–21] Experimentally, for the most part, the focus has been on measuring the formation of the Abrikosov-Suhl-Kondo resonance in the density of states while in the Kondo regime. Theoretically, many other quantities have been explored, such as the nonlocal spin correlations between the impurity and conduction electrons $\chi^{\text{sp}}_{\text{NL}}(\mathbf{r},t) =\langle\hat{s}^z(\mathbf{r},t)\hat{S}^z_{\text{imp}}(0)\rangle$, which is typically used to study the Kondo screening.[6,7,22–24] The Kondo cloud could be explored by measuring $\chi^{\text{sp}}_{\text{NL}}(\mathbf{r},t)$ but the nonlocality would require a two-probe setup: one to measure the impurity and one for the bath. Currently, and in the foreseeable future, such a setup is not feasible. In the following we show that the screening length can be deduced from local measurements of only the bath electrons.

To calculate the required bath correlation functions [Eqs. (4a) and (4b)], in the presence of magnetic impurities, we used a numerically exact scheme, briefly outlined below.[25] In principle, this formalism allows one to calculate all physical quantities of the impurities and the substrate. We believe this is the ideal framework to apply to real experimental systems.[22,23,26,27] The general Hamiltonian of a noninteracting substrate with n bands coupled to N-atomic impurities with amplitude $V^n_{\mathbf{k}\sigma,\alpha s}$, including direct exchange $J_{\alpha,\alpha'}$ between impurities is

$$\begin{aligned}H_{\text{sub}} &= \sum_n\sum_{\mathbf{k},\sigma}(\epsilon^n_{\mathbf{k}\sigma}-\mu)b^\dagger_{n,\mathbf{k}\sigma}b_{n,\mathbf{k}\sigma} + \sum_{\alpha=1}^{N}\sum_s E_{\alpha s}c^\dagger_{\alpha s}c_{\alpha s} \\ &+ \frac{1}{2}\sum_{\alpha=1}^{N}\sum_{s_1\ldots s_4}U^\alpha_{s_1\ldots s_4}c^\dagger_{\alpha s_1}c^\dagger_{\alpha s_2}c_{\alpha s_3}c_{\alpha s_4} \\ &+ \frac{1}{2}\sum_{\alpha\neq\alpha'}^{N}J_{\alpha,\alpha'}\hat{\mathbf{S}}_\alpha\cdot\hat{\mathbf{S}}_{\alpha'} \\ &+ \sum_{\alpha=1}^{N}\sum_n\sum_{\mathbf{k},\sigma,s}[V^n_{\mathbf{k}\sigma,\alpha s}b^\dagger_{n,\mathbf{k}\sigma}c_{\alpha s}+\text{H.c.}],\end{aligned} \quad (5)$$

where $c^\dagger_{\alpha s}(c_{\alpha s})$ is the electron creation (annihilation) operator for an impurity, with a complete set of quantum numbers s. Here $\hat{\mathbf{S}}_\alpha$ is the total spin of an adatom, $E_{\alpha s}$ are the bare energy levels, and $U^\alpha_{s_1\ldots s_4}$ is the Coulomb interaction. In principle, all of the above parameters, along with the dispersion of the metal $\epsilon^n_{\mathbf{k}\sigma}$, could be obtained from an *ab initio* calculation, e.g., density-functional theory. With respect to Hamiltonian (5), the generating functional, with action S_A, for the entire system can be written as a functional integral over Grassmann variables, including source terms A_i and $\bar{A}_i$ for the bath electrons and each impurity; $Z_A =\int D[\bar{c},c]D[\bar{b},b]e^{-S_A}$. Because the host metal is assumed to be noninteracting, i.e., Gaussian, the bath electrons can be integrated out exactly, leading to a reduced generating functional $Z_A\sim\int D[\bar{c},c]e^{-S^{\text{eff}}_A}$, with an effective action for the impurity sites. The propagator of the bath or any correlation function can be obtained by suitable functional differentiation with respect to the sources. In doing so, general correlation functions of the system are expressed in terms of the local impurity correlation functions. While there are a variety

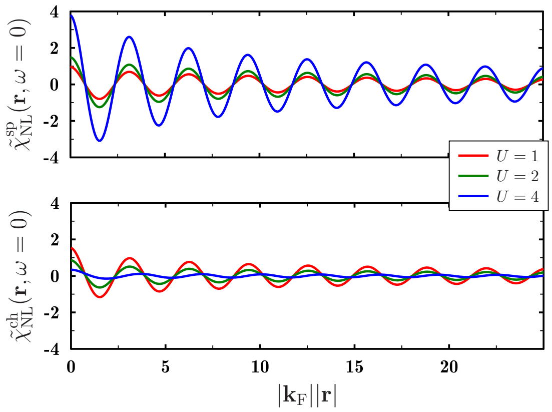

FIG. 1. (Color online) The zero-frequency nonlocal spin (top) and charge (bottom) susceptibilities, for the single-impurity spin-$\frac{1}{2}$ symmetric Anderson model as a function of the distance from the impurity site and for an inverse temperature of $(k_{\rm B}T)^{-1}=\beta=200$. Here, $\tilde{\chi}_{\rm NL}=[\frac{2(|\mathbf{k}_{\rm F}||\mathbf{r}|)^2}{\pi N_\sigma \Gamma}]\chi_{\rm NL}$. All other parameters are given in Table I. (The gray-scale line shade corresponds to different on-site Coulomb interactions U, decreasing from dark to light.)

of computational impurity solvers available, we used the numerically exact continuous-time quantum Monte Carlo method of Ref. 28 for the evaluation of the frequency-dependent two-particle Green's function of the impurity.

For simplicity and clarity, instead of the full Hamiltonian (5), we take the model of a single-orbital adatom on a metallic surface described by the symmetric single-impurity spin-$\frac{1}{2}$ Anderson model,[15] with an onsite interaction U and a three-dimensional parabolic dispersion for the bath. We will neglect the direct tunneling into the impurity. With this contribution our results would be modified only for $\mathbf{r}\approx 0$. When studying Kondo correlations, usually the nonlocal equal-time spin-spin correlations between the impurity and the bath, $\chi^{\rm sp}_{\rm NL}(\mathbf{r},t)=\langle \hat{s}^z(\mathbf{r},t)\hat{S}^z_{\rm imp}(0)\rangle$ are considered. Here we instead focus on the zero-frequency component of the spin and charge correlations for different values of U. Figure 1 shows that their spatial periodicity $\lambda=\pi k_{\rm F}^{-1}$ is the same as that of Friedel or Ruderman-Kittel-Kasuya-Yoshida oscillations.[31] With increasing U, charge fluctuations of the impurity and the corresponding correlations are suppressed as expected while spin correlations are enhanced. Note that in contrast to the equal times correlations,[24] ferromagnetic and antiferromagnetic contributions appear in approximately equal magnitudes. This shows that the impurity spin is screened dynamically, as higher-frequency components on a scale set by the Kondo temperature $T_{\rm K}$ eliminate the ferromagnetic correlations.

Although being of theoretical interest, measurements of such nonlocal quantities is currently unfeasible. Alternatively we propose to measure the local spin or charge correlations of the bath through conductance fluctuations. Figure 2 proves that a signature of the Kondo cloud also appears in these local quantities of the bath, which appear in Eqs. (4a) and (4b). The spatial dependence of the zero-frequency components is shown in the insets. The period of the oscillations of both spin and charge is smaller compared to the nonlocal quantities of Fig. 1 by a factor of 2, with a relative phase

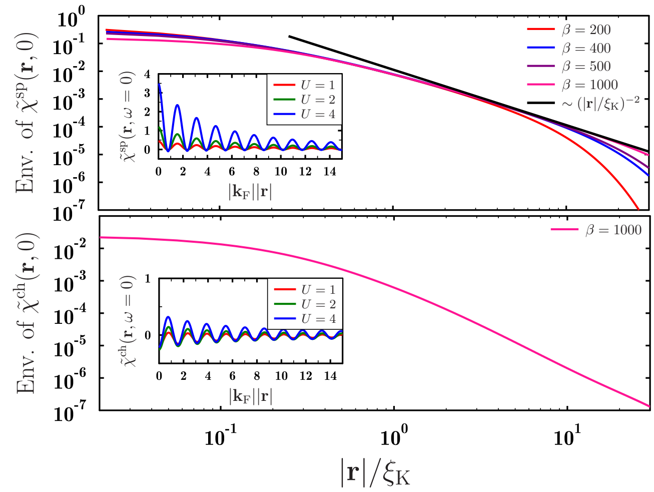

FIG. 2. (Color online) The insets show the local spin (top) and charge (bottom) correlations, Eqs. (4a) and (4b), for the single-impurity spin-$\frac{1}{2}$ symmetric Anderson model as a function of the distance from the impurity site and for an inverse temperature of $(k_{\rm B}T)^{-1}=\beta=200$. The envelope of these susceptibilities for fixed $U=1$ and different temperatures T, $\beta=(k_{\rm B}T)^{-1}$, are also shown for each. The horizontal axis has been rescaled by the Kondo length; $\xi_{\rm K}=\hbar v_{\rm F}/(k_{\rm B}T_{\rm K})$. The deviations for the spin susceptibility at small distances are purely numerical, coming from a high-energy frequency cutoff. Here, $\tilde{\chi}=[\frac{2|\mathbf{k}_{\rm F}|^2|\mathbf{r}|^2}{\pi N_\sigma \Gamma}]^2\chi$. (In the inset figure, the gray-scale line shade corresponds to different on-site Coulomb interactions U, decreasing from dark to light. While the main figure shows the inverse temperature β increasing from light (high temperature) to dark (low temperature) in gray scale.)

shift of $\pi/2$ between spin and charge. The figure clearly shows that the envelopes of these oscillations show a nonalgebraic decay, which changes to a power law at $\mathbf{r}\approx\xi_{\rm K}$, and ultimately, at finite temperature, the correlations are exponentially cut off by the thermal length $\xi_T\sim\hbar v_{\rm F}/k_{\rm B}T$. The appearance of the thermal length can be seen for the largest values of $\mathbf{r}$ in Fig. 2, as the power law changes over into an exponential. At zero temperature the decay remains a power law for $\mathbf{r}\gg\xi_{\rm K}$. The physical interpretation is that at zero temperature the magnetic impurity is almost fully screened by conduction electrons within $\xi_{\rm K}$, thus outside of this length-scale correlations are weak and decay rapidly but within $\xi_{\rm K}$ correlations of the bath, mediated by the impurity, remain nontrivial.

Figure 2 also shows a signature of the Kondo screening appearing in the density-density correlations. Normally, for Kondo systems, charge degrees of freedom are of little interest since the physics of the Kondo effect is entirely in the spin sector. Nonetheless these degrees of freedom remain coupled. This is also consistent with Ref. 32, where the Kondo length is shown to appear in the Friedel oscillations of the density.

Finally it should be noted that, because we have neglected interactions in the bath, there is an infinite phase coherence length L_ϕ of the bath electrons. In any real system, L_ϕ, which itself is strongly temperature dependent, can be of the same order as $\xi_{\rm K}$ ($\xi_{\rm K}\approx 100$ nm for adatom systems). If $L_\phi<\xi_{\rm K}$ the Kondo correlations are lost before the screening length is reached, resulting in an underscreened impurity. In general, the detection of the Kondo cloud as outlined here would necessitate systems with a higher Kondo temperature (smaller ξ_K) and lower operational temperatures (larger L_ϕ).

In conclusion, we have shown that the conductance fluctuations of a tunneling current into an interacting system are determined by the charge and spin susceptibilities of the system. We have also shown that one application of this is to use an STM to detect the Kondo screening cloud. We have furthermore developed a general method to exactly calculate n-point correlations for experimentally relevant setups consisting of multiple adatoms or correlated "sites." Extension of these results to finite frequency and multiple impurities would allow one to study, for example, the singlet-triplet excitations of two or more antiferromagnetically coupled adatoms.

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