Spin-polarized tunneling microscopy and the Kondo effect

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(Dated: July 14, 2018)

We present a theory for spin-polarized scanning tunneling microscopy (SP-STM) of a Kondo impurity on an unpolarized metallic substrate. The spin polarization of the SP-STM breaks the spin symmetry of the Kondo system, similar to an applied magnetic field, leading to a splitting of the Abrikosov-Suhl-Kondo (ASK) resonance. The amount of splitting is controlled by the strength of the coupling between the impurity and the SP-STM tip and also the overall spin polarization of the SP-STM.

PACS numbers: 72.15.Qm,72.25.-b

The Kondo effect has become one of the hallmarks of many-body physics [1], stimulating development of both new experimental and theoretical techniques. Apart from its original manifestation, in the form of anomalous low-temperature resistance of metals with magnetic impurities, its signature has been observed in a variety of other systems, such as transport in quantum dots [2,3] and, more recently, by scanning tunneling microscopy (STM) of single magnetic adatoms on a metallic surface [4,5,6]. In these experiments, below the Kondo temperature, an enhanced conductance near the Fermi energy is found due to the formation of a large peak in the density of states, the so-called Abrikosov-Suhl-Kondo (ASK) resonance. The imaging of a Kondo impurity with an STM does not directly resolve the ASK resonance; a more complex feature is found [4,5,7,8]. This feature is similar to the Fano resonance [9], more commonly found in atomic physics, and can be explained as resulting from an interference of two tunneling paths; one from the STM tip to the substrate and the other from the tip to the impurity-atom then to the substrate.

An STM allows one to study a Kondo system at the atomic level, and with the advent of the spin-polarized STM (SP-STM) [10,11], it is now possible to study spin resolved aspects of the Kondo effect. Because spin-dynamics is at the heart of the Kondo effect, the use of an SP-STM as a probe seems natural. However, owing to the spin symmetry of a Kondo system, one might expect nothing of interest to be found. Although, the spin symmetry is only preserved if one neglects the effects of the SP-STM tip on the Kondo system. Including the effects of the SP-STM tip on the Kondo system as a single impurity on a noninteracting conducting substrate; both are coupled by tunneling to the SP-STM, typically an antiferromagnetic coated standard STM tip, see Fig. 1. The full Hamiltonian is ( is the single-impurity Anderson model [12], describing an impurity level with on-site Coulomb term along with hybridization with the substrate. The Hamiltonian for the SP-STM is taken to be

$$H = H_{\text{And}} + H_{\text{tip}} + H_{\text{tun}},$$

where

$$H_{\text{And}} = \sum_{k,\sigma} \epsilon_k c_{k\sigma}^\dagger c_{k\sigma} + \sum_{\sigma} E_{d\sigma} d_{\sigma}^\dagger d_{\sigma} + U n_{\uparrow} n_{\downarrow}$$

$$+ \sum_{k,\sigma} V_{kd} c_{k\sigma}^\dagger d_{\sigma} + \text{H.c.}$$

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$$H_{\text{tip}} = \sum_{k,\sigma} \epsilon_k c_{k\sigma}^\dagger a_{k\sigma}^\dagger a_{k\sigma},$$

where $\epsilon_k^\prime = \epsilon_k + eV$. For the tip we allow for a different spectrum, therefore a different density of states, for each spin. The charge of the electron is $-e$ and $V$ is the applied voltage. Because tunneling can occur from the tip

FIG. 1: (Color online) Model of a Kondo system coupled to an SP-STM.
to the impurity, with amplitude $t^s_{dk}$, or to the substrate, with amplitude $w^s_{kk'}$, both processes have to be included in the tunneling term

$$H_{\text{tunn}} = \sum_{k,\sigma} t^s_{dk} d^\sigma_{ka} + \text{H.c.} + \sum_{k,k',\sigma} w^s_{kk'} c^\dagger_{k\sigma} a_{k'\sigma} + \text{H.c.}$$ (3)

We allow for the tunneling barrier to have spin dependence, inasmuch as the tunneling amplitudes $t^s_{dk}$ and $w^s_{kk'}$ can be different for respective spins, but we do not allow the barrier to flip spins. For an SP-STM, this is known experimentally [14, 17] and introduces a tip-substrate separation dependence on the spin polarization of the tunneling current. We therefore define the polarization of the tunneling current as $P^t = (\Gamma^t_1 - \Gamma^t_0) / (\Gamma^t_1 + \Gamma^t_0)$, where $\Gamma^t_\sigma = \pi \sum_k |t^s_{dk}|^2 \delta(\omega - \epsilon_{kk'})$. We will also make use of the equilibrium Hamiltonian $H_0 = H(eV = 0)$. In principle, super-exchange/RKKY effects between tip and the impurity atom are present. We address these interactions in later publications and focus here, solely on the effects of a spin-polarized current on the Kondo system.

Conductance. — Using the number operator for tip electrons, $N_{\text{tip}} = \sum_{k,\sigma} a^\dagger_{k\sigma} a_{k\sigma}$, the tunneling current is

$$I = -e \langle \dot{N}_{\text{tip}} \rangle/H.$$ This expectation value could be evaluated using the standard tunneling Hamiltonian formalism [18]; however, if vertex corrections are neglected, this only accounts for the interaction of the SP-STM and the Kondo system to lowest order. Here we follow Ref. [2] and use nonequilibrium Green’s function methods. This allows one to treat the applied voltage as the small parameter of perturbation theory, instead of the SP-STM-system coupling and the voltage, as is normally done. Assuming the density of states of the tip $\rho_{\text{tip}}$ and the bare substrate $\rho_{\text{sub}}$ are energy independent, the total current can be written as $I = I_{\text{sub}} + \delta I$, with $I_{\text{sub}} = 2e \pi \rho_{\text{sub}} V \sum_\sigma \omega |w^\sigma|^2 \rho_{\text{tip}}^\sigma$, which describes tunneling into a bare substrate, along with an additional term given by

$$\delta I = 2e \pi \sum_\sigma |w^\sigma|^2 \rho_{\text{tip}}^\sigma \int d\omega [n^F_{\text{tip}}(\omega + eV) - n^F_{\text{sub}}(\omega)]$$

$$\times \sum_\sigma \omega |w^\sigma|^2 \rho_{\text{tip}}^\sigma \left\{ (1 - q^2_\sigma) \text{Im} \left[ G_\sigma^d(\omega) \right] + 2q_\sigma \text{Re} \left[ G_\sigma^d(\omega) \right] \right\},$$ (4)

where

$$q_\sigma = t^\sigma + w^\sigma V \text{Re} \left[ G_0^R(\omega) / \omega \right]$$ (5)

is a spin dependent Fano parameter with $G_0^R(\omega) = \sum_\omega (\omega - \epsilon_k + i\eta)^{-1}$ and where $G_\sigma^d(\omega)$ is the Fourier transform of the retarded impurity Green’s function $G_\sigma^R(t) = -i\theta(t) \langle \{ d^\sigma(t), d^\dagger_{\sigma}(0) \} \rangle_{H_0}$. It should be noted that the time dependence and expectation value of the impurity’s Green’s function are with respect to the full equilibrium Hamiltonian $H_0$: $\langle O \rangle = e^{iH_0t} O e^{-iH_0t}$ and $\langle O \rangle_{H_0} = \text{Tr} e^{-\beta H_0} O / \text{Tr} e^{-\beta H_0}$. The total tunneling current contains the direct tunneling into the substrate and the impurity but also has an additional quantum interference term. It is this additional term that leads to a Fano line-shape in the conductance instead of the ASK resonance. The differential conductance $G \equiv dI/d(eV)$ is then

$$G(\omega) = \sum_\sigma G_{\text{sub}}^\sigma \left[ 1 + Y_\sigma(\omega) \right]$$ (6)

where $G_{\text{sub}}^\sigma = 2e \pi \rho_{\text{sub}} |w^\sigma|^2 \rho_{\text{tip}}^\sigma$ is the direct tunneling conductance between the tip and bare substrate, $Y_\sigma(\omega) = \Gamma_V / \omega$ for energies close to the Fermi energy $E_F$ (Eq. (6)) can be recast into the well known Fano line-shape giving

$$G(\omega) \sim \sum_\sigma G_{\text{sub}}^\sigma \left[ q_\sigma + \varepsilon_\sigma(\omega)^2 / (1 + \varepsilon_\sigma(\omega)^2) \right],$$ (7)

with rescaled energy $\varepsilon_\sigma = (\omega - E_{dc} - \text{Re} \Sigma_\sigma^d) / \text{Im} \Sigma_\sigma^d$ (where $\text{Re} \Sigma_\sigma^d$ is the self-energy of the impurity). Here we treat the Fano factor, Eq. (5), as an energy independent fit parameter; therefore, all of the energy dependence is in the impurity’s Green’s function.

Calculation of $G_\sigma^d$. — To calculate the impurity’s Green’s function a variety of methods are available, including equation-of-motion [19] or numerical renormalization group [13, 21]. Let us first use a simpler and, we believe a more, transparent method. For the asymmetric Anderson model, it is known the ASK resonance splits in the presence of spin-polarized leads/baths [13, 14, 13]. (Surprisingly the symmetric Anderson model exhibits no such splitting [13 ].) First we use scaling equations to determine the amount of this splitting [19, 20] for our model and then simply represent the ASK resonances as Lorentzians [8] centered away from the Fermi energy by the splitting value. These approximations are good for energies close to the Fermi energy and weak tip-impurity coupling, where asymmetries of the ASK peak remain small. For example see Fig. [2] These asymmetries, such as the widths and heights of the peaks, become more pronounced [13, 21] and more important for larger splittings than those predicted for our model.

For this calculation (of the impurity’s Green’s function) we neglect the tunneling into the substrate [22]. This reduces the full Hamiltonian to that of a single impurity coupled to two baths/leads; one being the substrate and the other the SP-STM tip. This two-lead-Kondo system can be down folded to a single lead (with spin-dependent hybridization) by the following canonical transformation, $c_{k\sigma} = (|V_{kd}|^2 + |t^s_{dk}|^2)^{-1/2} (V_{kd} f_{k\sigma} - t^s_{dk} h_{k\sigma})$ and $h_{k\sigma} = (|V_{kd}|^2 + |t^s_{dk}|^2)^{-1/2} (t^s_{dk} f_{k\sigma} + V_{kd} h_{k\sigma})$. This transformation requires the density of states of the
For most adatom systems, $U < D$, the bandwidth cutoff and current, with dot systems where $U$ should be cut-off at $D$; although, the Kondo temperature derived from numerical renormalization group (NRG). We have chosen a parameter set to reflect common experimental values [24]; although, changing the coupling does change the Fano parameter [24, 25].

In Fig. 4 we plot the enhancement of the conductance—the Fano parameter, Eq. (5), depends on the microscopic details of the coupling of the impurity to its environment [24]. Here we leave it as an unknown fit parameter and choose a fixed value; although, changing the coupling does change the Fano parameter [24, 25]. In Fig. 4 we plot the enhancement of the conductance (The real part of the impurity’s Green’s function can be found by the Hilbert transform of Eq. (10)). As one might expect the splitting of the ASK resonance leads to a splitting of the Fano line-shape.

We have explored the use of an SP-STM to probe the Kondo effect. By including the effect of the SP-STM on the Kondo system, we find that the spin-polarized current of the SP-STM adds a spin dependent hybridization term to the standard Anderson model. This hybridization both broadens and renormalizes the bare energy levels, of the impurity—in a spin dependent way—which in turn leads to a splitting of the ASK resonance. This

Near the Fermi energy, the spin resolved density of states of the impurity can be well approximated by Lorentzians [8] which, here, are centered away from the Fermi energy by the amount of splitting, Eq. (9).

$$
\rho^\sigma_d(\omega) = -\frac{1}{\pi} \text{Im}[G^\sigma_d(\omega)]
$$

$$
\approx \frac{1}{\pi \Gamma_V} \left[ 1 + \left( \frac{\pi T}{\sqrt{2}T_K} \right)^2 \right]^{-1} \times \left( 1 + \frac{[\omega + (\tilde{E}_{d\uparrow} - \tilde{E}_{d\downarrow})]^2}{(\pi k_B T)^2 + 2(k_B T_K)^2} \right)^{-1}
$$

where $T_K$ is the Kondo temperature and $k_B$ is the Boltzmann constant.

**Results.**—The Fano parameter, Eq. (5), depends on the microscopic details of the coupling of the impurity to its environment [24]. Here we leave it as an unknown fit parameter and choose a fixed value; although, changing the coupling does change the Fano parameter [24, 25]. In Fig. 4 we plot the enhancement of the conductance (The real part of the impurity’s Green’s function can be found by the Hilbert transform of Eq. (10)). As one might expect the splitting of the ASK resonance leads to a splitting of the Fano line-shape.

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leads be equal; therefore, we impose the spin polarization entirely by the tunneling matrix elements $t_{dk}$. The splitting of the ASK resonance can be understood as coming from the spin dependent renormalization of the bare energy levels $E_{d\sigma}$, analogous to an applied magnetic field. Using poor-man’s scaling the renormalized energies levels [19, 20, 22, 23], in our model, are given by

$$
\tilde{E}_{d\uparrow} = E_{d\uparrow} + \frac{\Gamma_V}{\pi} \ln \left( \frac{D_0}{D_1} \right)
$$

and

$$
\tilde{E}_{d\downarrow} = E_{d\downarrow} + \frac{\Gamma_V}{\pi} \left[ 1 + c \left( \frac{1 - P_t}{1 + P_t} \right) \ln \left( \frac{D_0}{D_1} \right) \right],
$$

where we have chosen (arbitrarily) a spin up polarized current, with $P_t \in [0,1]$, $c = \Gamma_0^f / \Gamma_V$, and where $D_0$ is the bandwidth cutoff and $D_1$ is the reduced bandwidth. For most adatom systems, $U < D_0$, unlike most quantum dot systems where $U \gg D_0$. Thus the scaling equations should be cut-off at $D_1 \approx U$. Therefore

$$
\tilde{E}_{d\uparrow} - \tilde{E}_{d\downarrow} \approx \frac{2\Gamma_V}{\pi} c \left( \frac{P_t}{1 + P_t} \right) \ln \left( \frac{D_0}{U} \right).
$$

Eq. (9) shows the amount of splitting is linear in the coupling strength $c$ but is also a function of the spin polarization $P_t$. Fig. 2 shows the range of the splitting. For comparison, we also plot the splitting as calculated using numerical renormalization group (NRG). We have chosen a parameter set to reflect common experimental values [24]; although, the Kondo temperature derived from these particular parameters is much larger than typical systems.
splitting of the ASK resonance is seen as a splitting of the Fano line-shape of the conductance. The amount of splitting can be controlled by the tip-adsorbate coupling and the overall spin polarization of the SP-STM. Although the splitting is relatively small, to obtain an equivalent splitting with the use of a magnetic field would require field strengths of order $10^2$ Tesla. The ability to detect such an effect requires the width of the ASK resonance, which is related to the Kondo temperature, to be of order of the splitting or smaller. All things being equal, systems such as Ti/Ag(100) with a $T_K \approx 40$K [8] or smaller would probably be needed.

Future investigations could include extending the model to include super-exchange/RKKY effects between the tip and the impurity, these become increasing important near the contact regime [28]. Replacing the non-spin-polarized substrate with a ferromagnetic one, introduces further spin dependent coupling. Experiments such as these could continue to test the effect of spin polarization of the bath(s) on the Kondo effect. Several theoretical predictions have been made in this area, [13, 14] but few experiments have been performed. [13] Also it is believed that the tunneling barrier depends on spin for an SP-STM; although, the exact nature of this is unknown and is currently an open question. This dependence could be experimentally observed by measuring the Fano line-shape splitting as a function of the tip-system separation.

This research was supported by the German Research Council (DFG) under SFB 668 and in part by Grant No. 4640.2006.2 (Support of Scientific School) from the Russian Basic Research Foundation. S.K. would like to thank Pascal Simon for stimulating discussions.

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[26] Because of the interference of tunneling paths, Eq. (6) is not proportional to a density of states, as is normally the case in tunneling [18].
[27] This neglects higher order interference effects and for the symmetric flat band model is negligible.
[28] The RKKY coupling is of order $J_{RKKY} \sim (V^2/U)(w^2/U_{tip})/\epsilon_r$. With typical on site interaction $U_{tip} \leq 5$ eV and for $w < V$ we find $J_{RKKY} < 1$ meV for the parameters considered here.

FIG. 4: (Color online) For $T = 10$ K, the normalized conductance $G(\omega) = G(\omega)/G_{sub}^\uparrow$ is shown for various values of the tip-impurity coupling $c = \Gamma_\uparrow/\Gamma V$ and with $G_{sub}^\uparrow/G_{sub}^\downarrow = 1/2$. For simplicity we set the Fano parameter $q_F = 0.2$ for both spins and impose a $T_K \approx 50$ K. Both of which are representative of Co/Cu(111) [24]. All other parameters are the same as Fig. 2.