STM conductance of Kondo impurities on open and structured surfaces

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I. INTRODUCTION

Scanning Tunneling Microscopy (STM) has proven to be one of the most useful tools to measure and characterize low energy electron spectroscopy, at least on open surfaces where the microscope tip has a direct access to the physical region under study. In a series of recent works, Kondo impurities on the surface of noble metals have been studied using the STM. In the Kondo problem there is a characteristic temperature \( T_K \) that separates the low temperature from the high temperature regimes. In the low temperature regime \( (T < T_K) \), the spectral density of a Kondo impurity develops a resonance at the Fermi energy, known as the Kondo resonance, and consequently locally changes the low energy spectroscopic properties of the system. When the microscope tip is placed on top of the impurity, the characteristic conductance vs voltage \( V \) shows a typical Fano-like structure that is identified as the fingerprint of the Kondo effect. In this way, the Kondo behavior of several transition metal impurities on different noble metal surfaces has been identified. In particular, the temperature dependence of the Kondo effect has been measured and, by studying Co impurities on different Cu surfaces, the scaling of the Kondo temperature with the host electron density has been shown.

STM has also been used to create nanostructures on noble metal surfaces by manipulation of single atoms. This allows to positionate impurities at special points of the surface or to engineer the environment of the impurity. In recent experiments the Kondo effect for a Co impurity inside a quantum corral has been observed.

The Fano like structure of the conductance observed when the STM tip is on top of a Kondo impurity, can be explained with a simple phenomenological model which includes an impurity resonant state at the Fermi energy. This resonance, that represents the Kondo resonance, has a width \( k_B T_K \). In this model, the differential conductance though the tip \( G(V) = \frac{dI}{dV} \) is given by:

\[
G(V) = g_0 + a \frac{q^2 - 1 + 2xq}{1 + x^2}.
\]

Here \( g_0 \) is a background conductance, \( a \) is a constant, \( x = (eV - \varepsilon_F)/k_BT_K \) and \( q \) is the Fano parameter that depends on the electronic structure and on the tunneling matrix elements.

For a more quantitative estimation, a many body calculation including the electron - electron interactions leading to the Kondo resonance is needed. Moreover, if the impurity is placed on a surface that is structured at a nanoscopic scale, the local density of states may strongly depend on energy, and effect that changes the shape of the Kondo resonance and, consequently of the voltage dependence of the STM conductance. A nanostructure with a characteristic length \( L \) has a characteristic energy scale \( \Delta \sim \hbar v_F/L \) where \( v_F \) is the Fermi velocity. In systems with \( \Delta \sim k_BT_K \) we expect strong effects. Quantum corrals on the Cu (111) surface can be engineered with the desired energy scale \( \Delta \), however it is not clear how much the surface states and bulk states contribute to the Kondo effect. Despite of some notorious effects like the observation, in elliptical corrals, of the so-called mirages, some experiments indicate that the surface states play almost no role in the development of the Kondo effect. If so, the Kondo screening would not be affected by the corral that confines only surface states. A detailed analysis of the STM conductance can be used to elucidate this point, however to do that the surface nanostructure and the Kondo correlations have to be treated on the same footing.

In what follows we present results for the tip conductance on top of a Kondo impurity. Using the numerical renormalization group, we evaluate the finite temperature conductance for an impurity on open and clean surfaces and on structured ones.

II. THE MODEL AND RESULTS

In this section we present the model and briefly discuss how the STM conductance is calculated using Wilson’s renormalization group. We then present the results for different situations. Our starting point is an extended Anderson model for magnetic impurities described with the following Hamiltonian.
where the operator $d_{\sigma}^\dagger$ creates an electron with spin $\sigma$ at the impurity orbital with energy $\varepsilon_d$ and Coulomb repulsion $U$, $c_{k\sigma}$ creates an electron in an extended state with quantum numbers $k$ and $\sigma$ and energy $\varepsilon_k$. The last term is a Falicov-Kimball like term which takes into account a Coulomb repulsion between the electrons at the impurity orbital and the ones at extended states. The quantum number $k$ includes the band index and crystal momentum. In the case of impurities on the Cu (111) surface there is a surface band that is relevant for determining the STM conductance.

The STM conductance at low temperatures is given by

$$ G(V) = \frac{4\pi e^2}{h} \rho(\varepsilon_F + eV), $$

where $eV$ is the voltage drop from the tip to the sample, and $\rho$ is the tip density of states that is assumed to be constant around the Fermi energy. In the linear response regime, the quantity $\rho(\varepsilon_F + eV)$ is given by

$$ \rho(\varepsilon_F + eV) = -\frac{1}{\pi} \text{Im} G(\varepsilon_F + eV). $$

Here $G(\omega)$ is the retarded Green function of the operator $t_c \psi_0^\dagger(R) + t_d d_{\sigma}^\dagger$, $t_c$ and $t_d$ are matrix elements for tunneling of an electron from the tip to the conduction band states and impurity orbital respectively and $\psi_0^\dagger(R)$ is the field operator that creates an electron on the surface at coordinate $R$. In what follows we consider that the tip is on top of the impurity and take $R \equiv R_i$ to be the impurity coordinate.

The Green function is calculated using the numerical renormalization group (NRG). For a general description of the method we refer to Refs. [13] and [14]. Here we give only some details that are specific to our calculation. The starting point of the NRG is to reduce the Hamiltonian (5) to that of a linear chain

$$ H_{AM} = \sum_{\sigma} \varepsilon_d d_{\sigma}^\dagger d_{\sigma} + U d_{\uparrow}^\dagger d_{\uparrow} + d_{\downarrow}^\dagger d_{\downarrow} + \sum_{k,\sigma} \varepsilon_k c_{k\sigma}^\dagger c_{k\sigma} + \sum_{k,\sigma} (V_k c_{k\sigma}^\dagger d_{\sigma} + V_k d_{\sigma} c_{k\sigma}) $$

$$ + \sum_{k,k',\sigma} U_{kk'} c_{k\sigma}^\dagger d_{\sigma} c_{k'\sigma'} + c_{k'\sigma'}^\dagger c_{k\sigma} + \sum_{\sigma,n=0} \lambda_n (f_{n\sigma}^\dagger f_{n+1\sigma} + f_{n+1\sigma}^\dagger f_{n\sigma}) $$

The states described by the operators $f_{n\sigma}^\dagger$ are Wilson’s orbitals centered at the impurity with the state $|0,\sigma\rangle$ associated with $f_{1\sigma}^\dagger$ being a Wannier like orbital centered at the impurity coordinate. The parameters $\lambda_n$ generate a logarithmic discretization of the conduction band, and $V_0$ describes the impurity potential scattering. The Hamiltonian (6) is diagonalized numerically using an iterative controlled scheme. At each iteration step $N$, a truncated Hamiltonian with $N$ orbitals is used and the many body states within an energy scale $\omega_N$ are evaluated. The thermodynamic and dynamical properties at each energy scale can then be calculated recursively.

As it is usually done to describe the Kondo problem, Hamiltonian (6) includes a single channel that is due to all the conduction bands. In the case of the Cu(111) surface there is a surface band and bulk states that contribute to the density of states at the Fermi energy. Wilson’s orbitals are assumed to be a linear combination of surface and bulk states, in particular the Wannier state $|0,\sigma\rangle$ contains a surface and a bulk component determined by the respective hybridization matrix elements $V_k$.

To calculate the STM conductance when the tip is on top of the impurity, we take the field operator $\psi_0^\dagger(R)$ to be $f_{1\sigma}^\dagger$ and calculate the retarded Green function of the operator $t_c f_{1\sigma}^\dagger + t_d d_{\sigma}^\dagger$. The matrix element $t_c$ depends on how much the Wannier orbital $|0,\sigma\rangle$ contains of the surface or bulk states. To be more specific on the nature of the Wannier orbital $|0,\sigma\rangle$ and on the matrix element $t_c$, a detailed model of the electronic structure of the metal host and of the electron transfer from the tip to the surface is required.

The retarded Green function of Eq. (6) is calculated as in Ref. [14] and

$$ \rho(\varepsilon_F + eV) = -\frac{1}{\pi} [G(\omega + i0^+) - G(\omega - i0^+)]_{\omega=\varepsilon_F+eV} $$

$$ = \sum_{\lambda,\lambda'} |M_{\lambda,\lambda'}|^2 \frac{2(e^{-E_{\lambda}/k_BT} + e^{-E_{\lambda'}/k_BT})}{Z(T)} \times $$

$$ \delta(\varepsilon_F + eV - (E_{\lambda'} - E_{\lambda})) $$

where $Z(T)$ is the grand partition function

$$ Z(T) = \sum_{\lambda} e^{-E_{\lambda}/k_BT} $$

and $M_{\lambda,\lambda'} = \langle \lambda | t_c f_{1\sigma}^\dagger + t_d d_{\sigma}^\dagger | \lambda' \rangle$. At each energy scale $\omega_N$ the discrete spectra is smoothed by replacing the delta functions by a smooth distribution. In what follows we use a logarithmic Gaussian that has been proven to give the best results.

**A. Impurity on a clean surface**

Here we present the results for a Kondo impurity on an open surface. In an energy scale of several $k_BT_K$, the bare density of conduction states is constant around the Fermi energy. Then the calculation can be done with the usual band discretization first proposed by Wilson.

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impurity, the temperature local spectroscopic properties are dominated by the parameters: 1, 3 and 4.

Results obtained for the conductance are shown in Figs. 1 and 2.

The STM conductance depends on the following parameters: $k BT_K$ that gives the low energy scale where the local spectroscopic properties are dominated by the impurity, the temperature $k BT$ and the Fano parameter $q$ that combines the tunneling matrix elements and some aspects of the band structure. If the Kondo resonance is represented by a simple resonant state, the Fano parameter $q$ is proportional to the matrix element $t_d$ that describes the direct tunneling into the localized state. In this case, $q$ accounts for the interference between the two tunneling channels. In general, real systems have a non zero $Re \Sigma(0)$ and there is no need to assume a direct tunneling to the localized state to have an asymmetric ($q \neq 0$) Fano resonance. In the NRG approach, the use of a symmetric conduction band simplifies the calculation and most of the numerical work on the Kondo effect has been done with this symmetry. We have introduced the potential scattering term $W_0$ to break the electron-hole symmetry of the band. The Fano resonance then depends on the conduction band parameters and in agreement with Ref. 13, with a non zero $W_0$ we obtain asymmetric line shapes even for $t_d = 0$.

In Fig. 1(a) the conductance vs applied voltage is shown for $\varepsilon_d = -U/2$ and $U_{dc} = 0$, all parameters are given in units of half the bandwidth $D = 1$. In Fig. 1(b) the results for $W_0 = 0$ and a set of the ratios $t_d/t_c$ are shown, this ratio determines the structure of the conductance. In Fig. 2(b), by independently varying $W_0$ for $t_d = 0$ we show that in a full NRG many body calculation, the low energy STM conductance is still given by a single parameter $q$ as given by equation (8). As $W_0$ is increased the resonance narrows indicating a lowering of the Kondo temperature. This is due to a decrease in the low density of states at the Fermi energy. The increase of $W_0$ also induces an increase of the Fano parameter. For simplicity, in what follows we take symmetric band ($Re \Sigma(0) = 0$) where the Fano parameter is given by $q = t_d/t_c \pi \rho V$, where $\rho$ is the density of states of the band.

Our calculation, that up to now mainly reproduces known results, shows that the NRG gives reliable results when used to evaluate the STM conductance. In what follows we present some results that include the behavior of the conductance when the energy of the localized orbital approaches the Fermi energy. The effect of the Falicov-Kimball term $U_{dc}$ and the temperature dependence of the conductance.

We start by discussing the conductance dependence on the localized orbital energy $\varepsilon_d$. As $\varepsilon_d$ is increased and approaches the Fermi energy, first the Kondo temperature increases and then the impurity enters an intermediate valence regime. In the conductance shown in Fig. 2 these effects are clearly observed. A notorious effect for intermediate values of $\varepsilon_d$ is the occurrence of an asymmetry for finite voltages: away from the Fermi level the values of $G(V)$ are smaller for $V < 0$ than for $V > 0$. This effect is due to the localized orbital resonance that is close to the Fermi energy. As $\varepsilon_d$ increases even more (not shown) the resonance crosses the Fermi level, the impurity occupation is reduced and the conductance shows a broad structure with a width that is given by the resonance width $\Gamma$ rather than by the Kondo temperature.

As we now show, the effect of a non zero electron-electron interaction $U_{dc}$ is equivalent to a shift in the impurity level. The parameter $U_{dc}$ is irrelevant, that means that...
in the Kondo regime the zero temperature fixed point does not depend on this parameter and the low temperature thermodynamic behavior is universal as for \( U_{dc} = 0 \). However this interaction may change some details in the low energy spectral structure for energies of the order or larger than \( k_B T_K \). We have analyzed the effect of \( U_{dc} \) on the STM conductance and the results are summarized in Fig. 3. As \( U_{dc} \) increases the Fano line shape changes and two different effects can be observed: i) the width of the conductance minimum increases indicating an increase in the Kondo temperature and ii) the voltage dependence of the conductance becomes more asymmetric. These two points are consistent with an effective shift the localized level toward the Fermi energy. The asymmetry in \( G(V) \) obtained when the resonant level approaches the Fermi level has not been discussed in detail in the literature, however experimental results for \( Co \) on \( Cu(111) \) and for \( Ti \) on \( Ag \) show this type of effect. In Ref. 3 the raw experimental has a marked asymmetry in the conductance that was attributed to a broad resonance lying just above the Fermi energy. Our results may explain the asymmetry as due to the same localized orbital that generates the Kondo resonance.

Now we present finite temperature results and analyze the temperature dependence of the conductance. To do this we calculate the finite temperature spectral densities following Refs. 1 and 2 and use the finite temperature extension of Eq. (3)

\[
G(V, T) = -\frac{4\pi e^2}{h} \rho_0 \int d\varepsilon \frac{\partial f(\varepsilon - eV)}{\partial \varepsilon} \rho(\varepsilon),
\]

where \( f(\varepsilon) \) is the Fermi function. In Fig. 3(a) the conductance for a small \( q \) and \( W_0 = U_{dc} = 0 \) is shown for different temperatures. As the temperature increases the width of the low voltage structure increases. In agreement with the experimental results of Ref. 1, our results show a width that increases according to the low temperature Fermi liquid theory. In the inset of Fig. 3 (a) the numerical width is shown for different temperatures, the thick continuous line is a fit with the Fermi liquid expression \( \gamma = \frac{\sqrt{2\pi B_K}}{k_B T} \sqrt{(\pi k_B T)^2 + 2(k_B T K)^2} \). In Fig. 4(b) the temperature dependence of a system with \( \varepsilon_d = -0.2 \) is shown. There is remarkable resemblance between some of these curves and the experimental results of \( Ti \) on \( Ag \).

### B. Impurity on a structured surface

We now present results for the STM conductance when the Kondo impurity is on a structured surface. The main effect of a nanostructure on the surface is to produce an energy dependence of the local density of states on a scale \( \Delta \) that may be comparable with \( k_B T_K \). If this happens the Kondo effect may be very much affected as discussed in previous works. Possible realizations of this situation may be obtained by placing impurities in quantum corrals, on small islands or in any structure with a characteristic size of the order of the hundred Å. The problem then becomes much more complicated mainly because of two reasons: on the one hand, the nanostructure of the host and the Kondo correlations have to be treated on the same footing. On the other hand, a detailed model for the metallic surface and the effect of the nanostructure on the bulk and surface states is needed. In general, in surfaces like the \( Cu(111) \) where there is a surface band, we expect structures like the quantum corrals or small islands to confine surface states keeping the bulk states nearly unaffected. If so, these structures would change the Kondo effect according to the participation of the surface states on the magnetic screening of the impurity. This point is still controversial: while the
Another important aspect of the Kondo effect in nanoscale systems is the role of the size of the nanostructure. We distinguish between two cases: large structures with \( \Delta \) out of scale and small structures with \( \Delta \approx k_B T_K \). For the latter, the Kondo temperature increases and the Fano structure becomes visible on the open surface result. For the former, the superstructure is not visible since \( \Delta \) is out of scale. In this case, the superstructure is not visible since \( \Delta \) is out of scale. For the at-resonance case (Fig. 6(b)) case and large barriers, the Kondo temperature increases and the Fano structure is broadened. This is due to an increase of the local density of states at the Fermi energy \( \epsilon_F \). The Kondo temperature has an exponential dependence with the density of states and consequently the broadening is very important for intermediate or large barriers. For the off-resonance case (Fig. 6(b)), the density of states decreases and the structure narrows, but large barriers a narrow Fano structure with a large \( q \) mounted on a background with a pronounced minimum at \( \epsilon_F \) is obtained.

As stated above, a nanostructure introduces a new length scale and the corresponding energy scale \( \Delta \). Since the number of microscopic parameters of the model is large, we just distinguish to cases: large structures with \( \Delta \ll k_B T_K \) and small nanostructures with \( \Delta > k_B T_K \). Another important aspect of the Kondo effect in nanostructures concerns the position of the Fermi level relative to the structure of the local density of states. In this respect we consider the two extreme cases: the Fermi energy lying at a resonance (at-resonance case) or between two resonances (off-resonance case). The quantum corrals on \( Cu(111) \) with characteristic sizes of the order of 100 Å correspond to the small nanostructure category. The elliptical corrals of Ref. [9] have the Fermi energy at a resonance.

Figure 5 illustrates the case of a relatively large nanostructure (\( \Delta \sim k_B T_K \)) for the at-resonance and off-resonance cases. The calculation was done with a small \( q \) and an intermediate barrier. The low temperature STM conductance shows a Fano like structure modulated by a superstructure of peaks separated approximately by the characteristic energy \( \Delta \). For the at-resonance case the central valley slightly broadens while for the off-resonance case narrow structures at low voltages are obtained. As the barrier height decreases the amplitude of the oscillations in the host local density of states decreases and the conductance continuously approaches the open surface result.

Figure 6 shows the results for a small system (\( \Delta > k_B T_K \)). In this case the superstructure is not visible since \( \Delta \) is out of scale. For the at-resonance (Fig. 6(b)) case and large barriers, the Kondo temperature increases and the Fano structure is broadened. This is due to an increase of the local density of states at the Fermi energy \( \epsilon_F \). The Kondo temperature has an exponential dependence with the density of states and consequently the broadening is very important for intermediate or large barriers. For the off-resonance case (Fig. 6(b)), the density of states decreases and the structure narrows, but large barrier a narrow Fano structure with a large \( q \) mounted on a background with a pronounced minimum at \( \epsilon_F \) is obtained.

FIG. 5: Linear conductance spectra for an impurity in a confined system \( T_K \sim \Delta \) and \( \delta/\Delta \sim 0.35 \) (a) at-resonance (b) off-resonance. Other parameter are \( \epsilon_d = -U/2 \), \( V = 0.2 \), \( t_d/t_c = 0.1 \), and \( W_0 = U_{dc} = 0 \). For comparison the lineshape for an open surface is shown ( thin line).

FIG. 6: Linear conductance spectra for an impurity in a confined system with \( t_d/t_c = 0.1 \), \( T_K \sim \Delta/10 \) and different widths \( \delta \) of the resonant states (a) at-resonance without barrier (thin solid line) and \( \delta/\Delta \sim 0.63, 0.42, 0.21 \) (thick solid line) (b) off-resonance without barrier (thin solid line) and \( \delta/\Delta \sim 0.63, 0.42 \) (thick solid line).
III. SUMMARY AND DISCUSSION

We have analyzed the STM conductance $G(V)$ for Kondo impurities on clean and structured surfaces. Starting with an Anderson model we used the NRG to calculate the physical properties and voltage dependence of the conductance. For clean surfaces we showed that the numerical approach gives reliable results and reproduces the Fano type STM response. In particular, we have studied in some detail how $G(V)$ depends on different microscopic parameters and on temperature. As the impurity level is shifted away from the symmetric case $\varepsilon_d = -U/2$, either by changing its energy or by means of a Falicov-Kimbal interaction, the Fano line is distorted. For a set of physical parameters, both the shape and the temperature dependence of the conductance are in good agreement with experimental observations. The temperature dependence is also in good agreement with the low temperature Fermi liquid theory.

To discuss the STM conductance of impurities in structured surfaces we used an effective one channel model. In surfaces like the Cu(111) one, where there is a surface band, this effective channel represents a linear combination of surface and bulk states. Since a surface structure perturbs much more the surface modes than the bulk modes, the sensitivity of the Kondo effect to the structure gives an idea of the surface mode participation in the Kondo screening. We have shown that small structures like the quantum corrals, that are quite efficient in confining surface electrons, should produce important changes in the Kondo temperature and on the STM response if the surface modes were active in the Kondo physics. The experiments show that the Fano line has essentially the same width when the impurity is on a clean surface or inside a corral, only small changes in the parameter $q$ are observed. This fact is a clear indication that the surface modes can sense the Kondo effect but are not important in determining it. This conclusion is in agreement with estimation of the microscopic parameters for surface and bulk states.

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