The surface density of states of layered $f$-electron materials

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We theoretically analyze the surface density of states of heavy fermion materials such as CeCoIn$_5$. Recent experimental progress made it possible to locally probe the formation of heavy quasi-particles in these systems via scanning tunneling microscopy, in which strongly temperature-dependent resonances at the Fermi energy have been observed. The shape of these resonances varies depending on the surface layer, i.e. if Cerium or Cobalt terminates the sample. We clarify the microscopic origin of this difference by taking into account the layered structure of the material. Our simple model explains all the characteristic properties observed experimentally, such as a layer-dependent shape of the resonance at the Fermi energy, displaying a hybridization gap for the Cerium-layer and a peak or dip structure for the other layers. Our proposal resolves the seemingly unphysical assumptions in the preceding analysis based on the two-channel cotunneling model.

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Recent advances in scanning tunneling microscopy (STM) provide the ability to analyze strongly correlated materials with high spatial and energy resolution, and thus enable to visualize strongly correlated states. One of the more recent successes in the field of STM has rendered it possible to examine heavy fermion materials. Aynajian et al. studied CeMIn$_5$ (M=Co, Rh) and succeeded in using the STM for locally observing the gradual formation of heavy quasi-particles, which are ubiquitous for heavy fermion materials, when lowering the temperature. While the $f$-electrons in CeCoIn$_5$ strongly influence the material properties, which give rise to an effective electron mass 10 - 50 times of the bare electron mass, the $f$-electrons in CeRhIn$_5$ seem to be decoupled from the conduction electrons. Thus, the comparison of both materials enables the authors to distinguish the influence of the strong correlations in the $f$-electrons. Furthermore, the cleaving process used in these experiments leads to the exposure of multiple termination surfaces which are distinguished by different chemical components of the material. The STM spectra measured on the Ce-layer of CeCoIn$_5$ reveal that around the chemical potential a dip is formed, which is the signature of the hybridization gap between $f$- and conduction electrons. On the other hand, spectra taken on the Co-surface of CeCoIn$_5$ show an enhancement of the spectral weight for energies below the chemical potential at low enough temperatures.

These experimental results are explained by the authors using the two-channel cotunneling model, where it is assumed that the electrons of the STM tip can simultaneously tunnel into the $f$-electron levels (conduction electron levels) with the amplitude $t_f$ ($t_c$), respectively. By tuning the ratio $t_f/t_c$, they construct the spectra having a dip or a peak at the chemical potential. In order to explain the experimental findings, the STM spectra on the Ce-layer (Co-layer) are modeled by a large tunneling into the conduction electron levels ($f$-electron levels). However, as already noticed by Aynajian et al., this assumption seems to be unphysical and is contrary to the general expectation that a tunneling into the strongly localized $f$-orbitals should be weak. Particularly, the claim of a strong tunneling from the STM tip into the $f$-electrons of Ce, when the STM tip is not located above Ce but above Co, seems to be questionable.

In this paper, we propose an alternative and consistent explanation for the STM spectra, which is based on the layered structure of CeCoIn$_5$, and resolve the apparent inconsistency encountered in the previous analysis based on the two-channel cotunneling model. We will qualitatively explain all the essential features in the experimental findings of Aynajian et al. by assuming a tunneling only between the STM-tip and the spd-conduction electrons, where tunneling processes into the $f$-electron orbitals play an insignificant role on both of the Ce- and Co-layers. Our scenario demonstrates that the characteristic dip-peak structure in the STM profile reflects the intrinsic properties of the material, which is contrasted to the previous scenario where the interplay of the STM tip and the substrate would give such a structure.

General idea: The essential point in our scenario is that CeCoIn$_5$ is built up of layers of different constituents; $f$-electrons only exist in the Ce-layers, while in the Co- and In- layers only conduction electrons in spd-bands are present. A key observation is that the Kondo effect in the $f$-orbitals of Ce, which leads to the formation of heavy quasi-particles at low temperatures, is not only observable in the orbitals of Ce, but also significantly influences the electrons of Co layers via the proximity effect between the Ce- and Co-layers. The penetration of the Kondo effect indeed results in the formation of a strongly correlated resonance at the Fermi energy in the local density of states (LDOS) of Co-electrons. An important point is that the resulting resonance in the LDOS...
of Co-electrons takes a different shape from that in the Ce-layer; due to the propagation of electrons from the Ce-layer to the Co-layer this shape should be changed.

The mechanism proposed here for explaining the STM profile is based on the fact that CeCoIn$_5$ is a naturally-layered heavy fermion compound, implying that the observed dip-peak profile reflects the intrinsic properties of the layered material. In this respect, the present scenario is quite different from that of the two-channel cotunneling model, which relies on the interference effect between two possible tunneling processes to produce such a structure. We note here that in a recent related study we have analyzed the paramagnetic state of artificially layered f-electron superlattices, where we have shown that the Kondo effect can be observed even in the conduction electrons of the spacer-layers in between the f-electron layers. Also, in a slightly different context, such a layer-dependent shape of the Kondo resonance has been observed in STM spectra, which are taken on a Copper surface above a single magnetic impurity which is buried below the surface. These recent studies naturally motivate us to demonstrate here that the STM measurements can probe the penetration of the Kondo effect of the Ce-f-orbitals into the normal-metal layers at the surface of CeCoIn$_5$.

Another point we wish to stress here is that to explain the experimental findings we do not need a fine tuning of the tunneling into c- and f-electron orbitals, $t_f$ and $t_c$, which has been previously used to reproduce the observed resonance. We thus avoid an unrealistic strong tunneling between the STM-tip and the f-electron orbitals, which is expected to be weak, because f-electron orbitals are strongly localized.

Model and method: We now demonstrate the above scenario by explicitly performing the numerical calculation for a simplified model. The crystal structure of CeCoIn$_5$, which is shown in Fig. 1, is built up of three different two-dimensional (2D) layers. The Ce- and Co-atoms form a regular three-dimensional cubic lattice, while the In-atoms occupy positions within the Ce-layers and in layers which are located between the Ce- and Co-layers. The measurements in ref. were taken on the Ce- and the Co-layer. In order to simulate the STM spectra, we construct a minimal model which is able to qualitatively reproduce the results. We assume a single spd-orbital on each of the Ce-atoms and the Co-atoms and a direct hopping between these two orbitals. In order to include the effects induced by the strong correlations in the f-orbitals of Ce, we furthermore couple the conduction electrons of the Ce-atom to localized spins. We note that a more elaborate model for CeCoIn$_5$ should incorporate the multi-orbital structure of the f-electrons of Ce. Our model thus consists of a regular cubic lattice, which is made of two different 2D layers: a square lattice of Ce-atoms including f- and conduction-electrons (f-electrons are approximated as localized spins), and the other layers, which include only non-interacting electrons, corresponding to the Co-layers. Furthermore, using open boundary conditions and varying the number of layers in our system, we are able to change the nature of the surface layer in order to simulate the STM spectra which are taken on various terminating layers of CeCoIn$_5$. Thus, our theoretical model reads

$$H = H_{Ce} + H_0 + H_{inter}$$

$$H_{Ce} = t \sum_{<i,j>,\sigma} c_{i\sigma}^\dagger c_{j\sigma} + J \sum_i \hat{s}_i \cdot \hat{S}_i$$

$$H_0 = t \sum_{<i,j>,\sigma} c_{i\sigma}^\dagger c_{j\sigma}$$

$$H_{inter} = t \sum_{i<z,z'>\sigma} c_{i\sigma}^\dagger c_{i'\sigma}.$$

We assume that all hopping constants are equal for simplicity. The correlation effects of the f-electrons are taken into account by an antiferromagnetic coupling of the conduction electrons to the localized spins in the Ce-layer, $J \hat{s}_i \cdot \hat{S}_i = c_{i\sigma}^\dagger c_{j\sigma} \hat{\sigma}_{\sigma\sigma'} \cdot \hat{S}_i \cdot (\hat{\sigma}_{\sigma\sigma'}$: Pauli matrices).

We furthermore assume that the STM-tip can be modeled as a point probe. The conductance between the f-electron material and the STM tip is then proportional to the LDOS of the surface layer at the location of measurement. We will thus study in this paper the surface LDOS of CeCoIn$_5$, which can be directly compared to the STM spectra in reference.

For solving this model, we use the inhomogeneous dynamical mean field theory (IDMFT) in combination with the numerical renormalization group (NRG). Within DMFT, one describes the lattice model as a self-consistent solution of a quantum impurity model. This approximation becomes exact in the limit of infinite dimensions. However, in many previous calculations it has been shown that DMFT can capture the essential physics of heavy fermion systems, i.e. the low energy physics including heavy quasi-particles as well as magnetic phases can be described by the DMFT. A detailed description of the IDMFT for superlattices can be found in reference.
Figure 2: (Color online) Temperature dependent LDOS close to the Fermi energy, $\omega = 0$, for a system with a Ce-surface-layer. The temperatures are scaled so that the Kondo temperature of the simulated system is $50K$ corresponding to CeCoIn$_5$. The right inset shows the shape of the whole simulated band. The left inset shows the simplified lattice, we have used to simulate the spectra.

Figure 3: (Color online) Same as Fig. 2, but for a lattice corresponding to the Co-layer in CeCoIn$_5$.

**Simulated LDOS**: We simulate a slightly doped system with $\langle n_{Ce} \rangle = 1.1$ and $\langle n_{Co} \rangle = 1.2$ conduction electrons per Ce- and Co-atom, respectively. We choose this doping in order to imitate the slope of the high temperature spectra of the experimental data, where the Kondo effect does not influence the system. We believe that the asymmetries at high temperatures in the experimental data are due to the lattice structure of CeCoIn$_5$ and are not caused by the interplay of the STM tip and the substrate of CeCoIn$_5$. By taking into account all Ce-, Co-, and In- energy bands which are close to the Fermi energy, a better agreement between the simulated and measured spectra at high temperatures can be expected. However, the inclusion of all energy bands is out of reach in this study. Nevertheless, our simplified model can qualitatively explain the measured spectra.

We set the antiferromagnetic coupling between the conduction electrons and the localized moments in the Ce-layers to $J = 2t$, and treat systems including up to 30 layers. The shape of the Kondo resonance at the Fermi energy as well as the temperature dependence, when scaled with the Kondo temperature, do not depend on the coupling strength. The coherence temperature for the formation of heavy quasi-particles for this interaction strength is approximately $T_K \approx \frac{1}{4}t$. In order to compare the temperature dependence of our results to the experimentally measured STM spectra, we adjust our energy scale so that $T_K = 50K$ roughly corresponding to the coherence temperature of CeCoIn$_5$.

In Figs. 2 and 3 we show the simulated LDOS of the conduction electrons for the Ce- and Co-surface layers. Fig. 2 corresponds to the Ce-layer including $f$-electrons. For the Ce-layer, a hybridization gap opens at the Fermi energy when the temperature is lowered. The formation of heavy quasi-particles by the hybridization between the $f$- and $spd$-electrons leads to a transfer of spectral weight from the Fermi energy to energies above the Fermi energy in the LDOS of the conduction electrons. The resulting pseudo-gap at the Fermi energy as well as the peak above the Fermi energy and the temperature dependence of these resonances agree fairly well with the measured STM spectra on the surface layer A in Aynajian et al., which has also been identified as Ce-layer.

In Fig. 3 we show the LDOS of the non-interacting layer which corresponds to the Co-layer of CeCoIn$_5$ in our calculations. A resonance due to the Kondo effect of the $f$-electrons in the Ce-layer is also visible in this non-interacting layer. However, the shape of this resonance is completely different. Instead of a pseudo-gap at the Fermi energy, we observe an enhanced spectral weight below the Fermi energy in the LDOS of the conduction electrons. Furthermore, we observe that the spectral weight above the Fermi energy is decreased when lowering the temperature. The temperature dependence of this spectral weight transfer is similar to that of the formation of the hybridization gap in the Ce-layer. Around the coherence temperature, the spectral weight below the Fermi energy suddenly increases. We want to stress that this enhanced spectral weight (peak) is observed in the LDOS of the conduction electrons. We do not assume any direct tunneling into the $f$-electron levels. The Kondo effect in the Ce-layers influences the LDOS of the conduction electrons of the nearest neighboring layer. This resonance qualitatively agrees with the STM spectra taken on the Co-surface of CeCoIn$_5$, where a peak is found slightly below the Fermi energy.

We note that the results shown here are calculated via a simplified crystal structure, i.e. a 3D cubic lattice instead of a realistic crystal structure of CeCoIn$_5$ and neglecting the multi-orbital structure of the $f$-electrons of Ce. The inclusion of the multi-orbital structure will lead to further anisotropies in the shape of the hybridiza-
tion gap and a possible fine structure at finite temperatures. Nevertheless, this simplified model is sufficient to qualitatively reproduce the experimentally observed resonances. The main point is that the resonances resulting from the Kondo effect of the Ce-atoms can be observed in the LDOS of the conduction electrons of the neighboring non-interacting layers. The shape of these resonances at the Fermi energy thereby depends on the distance between the probed layer and the f-electron layer. If one uses a realistic crystal structure, one can expect that the STM can also quantitatively be reproduced by such calculations.

Before concluding the paper, some comments are in order on the theoretical background. The appearance of a Kondo resonance in the LDOS of the conduction electrons of the non-interacting layers can be understood using the Dyson equation. The LDOS, which is probed in STM measurements, corresponds to the imaginary part of the interacting Green’s function. The change in the LDOS at position $x$ and frequency $\omega$ originating from correlation effects in the Ce-layer, can be written as

$$\Delta \rho(x, \omega) = -\frac{1}{\pi} Im \left( G^0(x, x_{Ce}, \omega) T(x_{Ce}, \omega) G^0(x_{Ce}, x, \omega) \right),$$

where $G^0(x, x_{Ce})$ is the free Green’s function for the non-interacting conduction electrons. All interaction effects of the f-electrons are included in the T-matrix, $T(x_{Ce}, \omega)$, which for the case of heavy fermions develops a resonance at the Fermi energy below the coherence temperature. This resonance is carried via the non-interacting electrons from the Ce-atoms, $x_{Ce}$, to the place of the STM measurements, $x$.

**Conclusions:** We have analyzed the surface LDOS of layered heavy fermion systems such as CeCoIn$_5$. We have shown that resonances originating from the Kondo effect of the f-electrons occur at the Fermi energy. These resonances can be observed not only in the conduction electrons of the Ce-atoms, but also in the LDOS of the conduction electrons of all other layers, such as the Co- and the In-layers. The shape of this resonance is quite different for different layers and depends on the distance from the f-electron layer. Therefore, the STM results in Aynajian et al. can be unambiguously explained by tunneling into the conduction electrons of different layers which have different distance to the Ce-layer. For explaining the measured spectra, we do not need a fine tuning done by Aynajian et al. for separate tunneling into the c- and f-electrons. Because f-orbitals are strongly localized, a direct tunneling into these orbitals should be very small and would be negligible in the Co-layers. Taking a realistic crystal structure of CeCoIn$_5$ or other heavy fermion materials into account, we believe that measured STM spectra can be quantitatively reproduced. This is left as a future project.

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