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Kondo peak splitting on a single adatom coupled to a magnetic cluster

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The Kondo resonance induced by a single cobalt adatom interacting with a ferromagnetic iron nanocluster is measured using low-temperature scanning tunneling microscopy. The persistence of the Kondo resonance is evidenced along with the predicted splitting of the spectral density peak for a Kondo impurity surrounded by a spin-polarized electron bath. Reversible quenching of the split feature is observed using atom manipulation between adjacent adsorption sites. Using a Green's-function formalism, we model a double Fano resonance leading to a quantitative insight of our observations.

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The Kondo effect arises from the antiferromagnetic screening of a localized magnetic moment by the spins of the delocalized electrons of a metallic host. This many-body interaction leads to the emergence of a peak in the density of states at the Fermi level.[1] In the past decade there has been an increasing interest in the Kondo effect after it was measured in the transport properties of quantum dots[2,3] and in scanning tunneling microscopy (STM) experiments.[4,5] In the latter case, local tunneling spectra on single adatoms exhibit a Fano line shape around the Fermi level evidencing the interference between two conducting channels involving either a coupling between the tip and a discrete level or between the tip and the continuum of states of the substrate. Although most of the experimental efforts have up to now focused on magnetic impurities interacting with a nonmagnetic host, the influence of ferromagnetic electrodes on the transport properties in the Kondo regime has been subject to several theoretical works[6–10] and is of great interest in the fast growing field of spintronics. A splitting of the differential conductance signal is predicted in the case of transport through quantum dots bridging spin-polarized electrodes[8,9] or in the case of magnetic adatoms on a metallic surface scanned with a magnetic STM tip.[11,12] However, very few experimental data are available on such systems. Transport experiments have confirmed that the exchange interaction between a magnetic moment localized on a C_{60} molecule and ferromagnetic Ni electrodes can induce a splitting of the Kondo peak.[13] A similar observation in a carbon nanotube quantum dot was reported later with a backgate electrode allowing a control of the energy levels.[14] A split resonance was also measured on a single Kondo-screened adatom interacting with an unscreened adatom by an exchange interaction.[15] Recently, the splitting of the Kondo peak due to ferromagnetic proximity effect was measured in nanowire-based quantum dots.[16] Experimental data involving atomic-scale Kondo systems with a mesoscopic magnetic electrode are however still lacking.

In this Communication, we report on the first measurement of the Kondo-induced zero-bias anomaly on a single adatom coupled with a magnetic cluster. We show that for a single Co atom adsorbed on a Fe island grown on a Au(111) surface, the Kondo resonance is preserved and can split owing to the strong coupling with the ferromagnetic Fe island. Using atom manipulation we can reversibly switch the atom position between two adsorption sites where the Kondo resonance appears either as a split peak, or a single peak, evidencing the extreme sensitivity of the Kondo signature to the adsorption site. Using a Green's-function formalism, we derive the analytical expression of a double Fano resonance line shape, which reproduces well the experimental spectra. This analysis allows an estimate of the relevant physical parameters, i.e., the Kondo temperature, the splitting energy, and the coupling strength between the adatom and the substrate.

STM measurements were performed with a low-temperature microscope operating at 5 K under ultrahigh-vacuum conditions (10^{-10} mbar). The Au(111) single crystal was cleaned by repeated cycles of Ar^+ sputtering and annealing at 800 K. Fe islands were grown by thermal evaporation of 99.99% pure Fe rod on the Au(111) sample at room temperature. Co atoms were deposited similarly on the Fe/Au sample cooled at 5 K on the STM stage so that a random distribution of single adatoms is obtained. Local scanning tunneling spectroscopy was performed using a lock-in amplifier with a 4 mV rms modulation at 800 Hz. Electrolytically etched tungsten tips were prepared before measurements by controlled indentation into the Au(111) surface until the band onset of the Shockley surface state was measured so as to ensure a flat DOS for the STM tip.

Figure 1(a) shows the Au(111) substrate after room-temperature growth of Fe islands and low-temperature deposition of single Co adatoms. The monatomic high Fe nanostructures are monodomain, ferromagnetic, in-plane magnets,[17] which grow in a pseudomorphic mode on Au(111).[18,19] The inset of Fig. 1(a) displays the well-known Kondo signature in the local tunneling spectroscopy of Co adatoms adsorbed on a fcc area or a reconstruction line of the Au(111) substrate in agreement with previous studies.[5] These dI/dU spectra are well fitted by a Fano resonance, which allows to estimate the Kondo temperature T_K.[5,20] This temperature gives the strength of the Kondo interaction between the localized magnetic moment and the conduction electrons of the metallic host and thus the energy scale that must be overcome to affect the Kondo interaction. For example, a magnetic field on the order of $k_B T_K/2\mu_B$ is predicted to induce a splitting of the Kondo resonance.[7] In the Co/Au(111) system we measured a typical value $T_K \approx 80$ K, in agreement with previous observations,[5] yielding a very high critical field of around 60 T, which is a typical value for an exchange field.

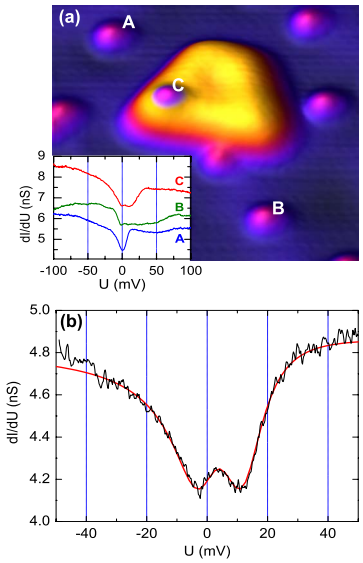

FIG. 1. (Color online) (a) 9 nm×9 nm topographic image of an iron island on Au(111) after deposition of single Co adatoms. The tunneling parameters were I=0.5 nA and U=0.1 V. Cobalt adatoms A and B are located on an fcc stacking area and on top of a reconstruction line, respectively. The adatom C is adsorbed on the Fe island. Inset: corresponding dI/dU curves. B and C spectra are vertically shifted by 0.5 nS and 2.5 nS, respectively. (b) Typical split spectrum taken above a cobalt atom adsorbed on top of an iron island, fitted by a Fano model where E_1=−6 meV, E_2=16 meV, Γ=8.8 meV, q_1=−0.25, and q_2=0.54.

Turning to the case of Co atoms adsorbed on Fe islands, we found that the Kondo signature is still observed in the dI/dU spectra. This demonstrates that the Kondo resonance is preserved in spite of the ferromagnetic host. The presence of a ferromagnetic electrode is expected to suppress the Kondo correlation if the adatom is ferromagnetically coupled to the island. The observation of the Kondo signature here shows that this effect does not dominate. A coupling between the localized moment and itinerant *sp* electrons of the island may explain these results.[21] In addition, about 30% of the 35 investigated atoms sitting on top of the Fe islands exhibit a split Fano shape [curve C in the inset of Figs. 1(a) and 1(b)]. We have investigated the influence of the adsorption site on the Kondo signature by atom manipulation induced with the STM tip. Using a continuous tunneling voltage with the tip held above a cobalt adatom, we induced a reversible switching of the atom between nearby adsorption sites on the iron island. The tunneling current recorded during such switching events displays a telegraph signal typical of a bistable system [Fig. 2(c)]. The STM topography images measured before and after the manipulation procedure allow to clearly distinguish the two corresponding positions of the adatom [Figs. 2(a) and 2(b)]. The spectra of Fig. 2 show that the split feature vanishes and reappears upon switching the atom between site *a* and site *b*. Considering the fact that the edges of the islands are aligned along the $\langle 1\bar{1}0\rangle$ directions of the underlying substrate, we found with an accuracy of less than 0.5° that the switching displacement occurs along these close-packed directions. The measured distance between the two sites is 2.6±0.1 Å, which is close to the Fe-Fe site to

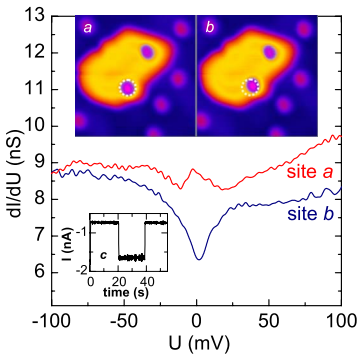

FIG. 2. (Color online) Insets (a) and (b) show 7 nm×7 nm successive topography images (U=0.1 V, I=0.9 nA) of a Co adatom before and after tip induced switching. The site *a* is reminded with a dotted circle at the same position within both images. Inset (c): telegraph noise shape of the tunneling current plotted as a function of time while a continuous −0.2 V tip-adatom bias is applied. The displayed spectra were measured on top of the Co adatom adsorbed on site *a* or site *b*.

site distance if we consider that the iron island is pseudomorphic. The apparent height of the adatom on both sites at a tunneling voltage of 0.1 V is the same (91±1 pm) indicating that there is no crystallographic defect below the adatoms. From these observations we can conclude that the adatom jumps between two adjacent sites. These data demonstrate that the Kondo signature strongly depends on the adsorption site of the adatom. While the physical origin of this behavior is still unclear, it may reveal a more complex spin structure of the Fe island than a single ferromagnetic monodomain, as recently reported for magnetic layers at surfaces.[22] For example, it can be possible that spin frustration on the hexagonal lattice leads to an effective compensation of the exchange interaction at certain Fe sites.

In order to get a deeper insight in the physics of the measured Kondo signatures, we modeled the split spectra using a double Fano resonance scheme within a Green's-function formalism.[23] In the case of single Kondo peak, the STM spectra can be described by a Fano resonance.[4,5] We stress that such a description does not include the underlying many-body physics of the Kondo effect but still provides a useful tool to understand the experimental results as we show in the following. Here we extend such an analysis to the case of a split Kondo peak that we modeled by two discrete levels that we interpret like a spin-up and spin-down Kondo resonance. A simple description can then be obtained by assuming that each state is coupled to all the continuum states with the same coupling energy. Within this assumption, the continuum can be represented by a single state which carries the total density of states of the continuum.[24] The Hamiltonian describing the interaction between two states $|\phi_1\rangle$ and $|\phi_2\rangle$ with energy levels E_1 and E_2 and a continuum $|\phi_0\rangle$ can therefore be written as

$$H = E_1|\phi_1\rangle\langle\phi_1| + E_2|\phi_2\rangle\langle\phi_2| + \Sigma(E)|\phi_0\rangle\langle\phi_0| + \sum_{i=1,2}(V_i|\phi_i\rangle\langle\phi_0| + V_i^*|\phi_0\rangle\langle\phi_i|), \quad (1)$$

where V_1 and V_2 are the energy-independent coupling matrix

elements between the discrete levels and the continuum of states of the metal host. The self-energy $\Sigma(E)$ is deduced from the Green's function of the continuum state $G_0(E) = \frac{1}{E-\Sigma(E)}$. Assuming that the substrate has a constant density of states ρ_0, we can deduce the Green's function $G_0(E)$ from the relation $-\frac{1}{\pi}\lim_{\epsilon\to 0^+} \mathrm{Im}\{G_0(E+i\epsilon)\}=\rho_0$. The real part of $G_0(E)$ is the Hilbert transform of the imaginary part and is equal to zero in this case. The Fano line shape is obtained from the calculation of the Green's function $G(E)=[EI-H]^{-1}$ (where I is the identity matrix) projected onto a linear combination of the two discrete states and the continuum state[23] $|\chi\rangle = T_1|\phi_1\rangle + T_2|\phi_2\rangle + T_0|\phi_0\rangle$ leading to the following relation:

$$\langle\chi|G(E)|\chi\rangle = -\pi\rho_0 T_0^2 \frac{\frac{E-E_1}{\Gamma_1}\frac{E-E_2}{\Gamma_2} + 2q_1\frac{E-E_2}{\Gamma_2} + 2q_2\frac{E-E_1}{\Gamma_1} - (q_1-q_2)^2 + i\left[\frac{(E-E_1)}{\Gamma_1}q_2^2 + \frac{(E-E_2)}{\Gamma_2}q_1^2\right]}{\frac{(E-E_1)}{\Gamma_1} + \frac{(E-E_2)}{\Gamma_2} - i\frac{(E-E_1)}{\Gamma_1}\frac{(E-E_2)}{\Gamma_2}}, \tag{2}$$

where $\Gamma_1=\pi\rho_0|V_1|^2$ and $\Gamma_2=\pi\rho_0|V_2|^2$. The Fano parameters $q_1=\frac{T_1V_1}{T_0\Gamma_1}$ and $q_2=\frac{T_2V_2}{T_0\Gamma_2}$ drive the line shape. They contain the ratios T_1/T_0 and T_2/T_0 that can be understood as the ratio of probabilities of transmission between the tip and the discrete states, and between the tip and the continuum state.[5] The conductance $g(E)$ is proportional to the density of states $n(E)$ of the Hamiltonian H projected onto $|\chi\rangle$, which takes the form

$$n(E) = -\frac{1}{\pi}\lim_{\epsilon\to 0^+}\mathrm{Im}\{\langle\chi|G(E+i\epsilon)|\chi\rangle\} = \rho_0 T_0^2 \frac{\left[1+\frac{\Gamma_1 q_1}{E-E_1}+\frac{\Gamma_2 q_2}{E-E_2}\right]^2}{1+\left[\frac{\Gamma_1}{E-E_1}+\frac{\Gamma_2}{E-E_2}\right]^2}. \tag{3}$$

We emphasize here that the same relation can also be deduced from the original work of Fano.[20] The Green's-function treatment described above adapted to the local transport measurement allows to clarify the assumptions and limitations of the model as we will discuss below.

In order to reproduce the experimental dI/dU spectra by a fitting procedure, we applied a double convolution of expression (3), which allows to take into account the thermal broadening due to finite temperature and the ac modulation. An *ad hoc* background was subtracted from the experimental data to obtain a featureless spectrum far from the Fermi level, which ensures the convergence of the fitting procedure. This background varies slowly with the energy and does not affect the main features of the Fano line shape in the spectra. This can be seen in the inset of Fig. 1(a), which shows raw data without background subtraction. The background usually appears as a hill-like structure in the spectrum, which corresponds to an electronic state of the Fe island, possibly a confined state of the two-dimensional electron gas within the Fe cluster. Figure 1(b) shows a typical spectrum acquired above another Co/Fe adatom. We used a fitting procedure where we assumed that $\Gamma_1=\Gamma_2\equiv\Gamma$, which allows a good quantitative description of the experimental data. The assumption of $\Gamma_1=\Gamma_2$ represents the fact that the relaxation time for both spin-split nonequilibrium Kondo events are the same. The relaxation time of nonequilibrium Kondo events is mainly related to the probability to create electron pairs by the finite-bias tunneling electrons.[25] In the small energy range concerning the observed resonances the relaxation time might not change considerably. We obtained an estimation of the splitting in energy $\Delta E=E_2-E_1=22$ meV. This corresponds to a magnetic field of 95 T within a Zeeman effect model where the splitting reads $\Delta E=2g\mu_B B$ with μ_B the Bohr magneton and assuming a Landé g factor close to the free-electron value $g\approx 2$.[26,27] The effective magnetic field deduced here is close to the exchange field experienced by an atom adsorbed onto the (111) surface of a fcc Fe crystal. This field can be estimated to be around 60 T, considering the number of closest neighbors (three Fe atoms) and the known bcc bulk Fe molecular field [eight Fe atoms, 150 T (Ref. 28)]. From the fitting procedure, we also can deduce $\Gamma=8.8$ meV leading to an estimate of the Kondo temperature of about 100 K. Over all the adatoms investigated, the splitting energies were measured typically between 10 and 20 meV, which correspond to exchange fields between 45 and 90 T. The mean value of Γ was measured at 7 meV and corresponds to $T_K=82$ K, very close to the value obtained on Co/Au(111).[5] As it can be seen in Fig. 1(b), the mean energy $E_0=(E_1+E_2)/2$ appears to be shifted from the Fermi level. The energy position of the Kondo peak is linked to the occupation number (between 0 and 2) of the atomic state that drives the magnetic moment.[1] In the case reported in Fig. 1(b), assuming that E_0 plays the same role like in the case of a single Kondo peak, we find an occupation number of 0.7 for the $3d$ state of the Co adatom which is close to experimental values previously reported.[29]

We have also investigated the spatial dependence of the Fano line shape by measuring dI/dU spectra with the STM tip located at different lateral distances d from the adatom center. Figure 3 shows an example of such a variation. On top of the adatom, the dI/dU spectrum has a symmetric structure, i.e., the two dips fall at the same conductance

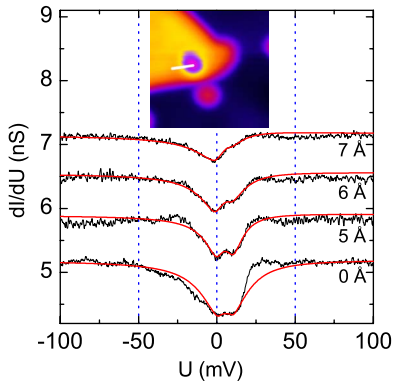

FIG. 3. (Color online) Series of dI/dU spectra measured at several lateral distances from an adatom center. Inset: corresponding 7 nm×7 nm topographic image (I=0.5 nA and U=0.1 V). The white line indicates the positions over which spectra were measured.

value. When the tip-adatom distance increases, the dip at positive bias is reduced and becomes a progressively vanishing shoulder. The same trend was observed around the adatom in two other directions (toward the center or the right corner of the island). Similar line shapes can be provided by the relation (3) as shown by calculated curves displayed in Fig. 3 where the parameters of the double Fano resonance were obtained from a fitting procedure. The resulting values of E_1, E_2, and Γ are almost constant for all the curves (3 meV deviation) and have the mean values $E_1=-1$ meV, $E_2=9$ meV, and $\Gamma=7$ meV. The variation in the line shape comes from variations in the Fano parameters q_1 and q_2. In this set of fitted curves, q_1 varies from −0.06 to −0.33 and q_2 from 0.15 to 0.74 when the distance d increases. It is worth noting that it is possible to produce the same qualitative evolution of the calculated curves by different variations in q_1 and q_2. In addition, different variations in these parameters were also obtained for measurements on different adatoms. However it is instructive to have a close look at the formula (3) from which it is easily seen that $n(E_1)=\rho_0 T_0^2 q_1^2$ and $n(E_2)=\rho_0 T_0^2 q_2^2$. As a consequence, the qualitative variation in the curves can be obtained by an increase in q_2 or at least a faster increase in q_2 with respect to q_1 when d increases. This is in contrast with the observed decrease in the Fano parameter in the case of a simple resonance.[4,5] Further theoretical investigation is needed to fully understand our observations. In particular, the Fano formula is probably limited to the measure obtained above the adatom. This can be understood by the fact that the density of states projected onto the continuum state Φ_0 is affected in this model by the discrete states.[23] However, in the limit case where the tip would be at very large distance from the adatom, one can expect that the density of states of the continuum should reach that of the unperturbed substrate. As a consequence, the model should be extended with a spatial description of the substrate in order to fully describe the experimental situation.

In conclusion, we found that the Kondo peak on a Co adatom can be split when the atom interacts with a magnetic Fe cluster. This splitting depends on the adsorption site, as demonstrated by reversible switching of an adatom between two positions. The experimental spectra can be well reproduced by a double Fano resonance calculated for two discrete levels coupled to a continuum of states.

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