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Direct evidence of the contribution of surface states to the Kondo resonance

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Using low-temperature scanning tunneling microscopy/spectroscopy, the Kondo resonance was observed on an isolated 5, 10, 15, 20-tetrakis-(4-bromophenyl)-porphyrin-Co (TBrPP-Co) molecule adsorbed on the Si(111)-$\sqrt{3}\times\sqrt{3}$Ag substrate. As the substrate has a two-dimensional surface state and does not have bulk states around the Fermi level (E_F), the Fano-shaped peak near E_F observed above the molecule caging a spin-active cobalt atom is a direct evidence of the contribution of the surface-state electrons to the Kondo resonance. The long decay length ($\sim$1.6 nm) of the resonance also supports for the surface-state contribution.

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I. INTRODUCTION

The Kondo effect is one of the nontrivial manifestations of correlated many-body physics.[1] The effect results from a correlation of the unpaired electrons of a magnetic impurity with conduction electrons of a host metal by spin-flip scattering processes.[2] As a result, a many-body singlet ground state is formed below a characteristic Kondo temperature (T_k), giving rise to a resonance, called the Kondo resonance, near the Fermi energy (E_F) at the site of the impurity.[2] Scanning tunneling microscopy/spectroscopy (STM/S), which has atomic-scale spatial resolution and meV energy resolution, is an ideal method to study the Kondo resonance in atomistic scale. The resonance is observed as a Fano-shaped resonance[3] near E_F in tunneling conductance spectra $dI/dV(V)$,[4,5] as described below

$$\frac{dI}{dV}(r,V) = C(V) + A(r)\frac{[\varepsilon + q(r)]^2}{1+\varepsilon^2}, \quad \varepsilon = \frac{eV-\Delta E}{\Gamma}. \quad (1)$$

Here, r is the lateral distance between the probe tip and the center of the magnetic impurity and $C(V)$ is a background conductance from the substrate. ΔE is a meV-order shift in the resonance from E_F due to the repulsion between the d levels of the substrate and the Kondo resonance. Γ, the width of the Fano resonance, is given by $k_B T_k/2$, where k_B is the Boltzmann constant. $A(r)$ is the amplitude of the resonance. q is a parameter related with an interference of the two channels contributing to the Fano line shape: one is a direct channel through the Kondo resonance localized at the magnetic impurity and the other is a channel into the conduction band of the substrate modified by the impurity.[6] The Fano resonance shape thus depends on q: it is a negative Lorentzian for $q=0$, a positive Lorentzian for $q=\pm\infty$, and most asymmetric when $q=\pm 1$.

So far, most of the STM/S studies on the single-impurity Kondo resonance were performed on noble metal (111) substrates.[4,5,7–16] Besides the bulk states, all these substrates have a surface state which has a parabolic free-electron-like energy dispersion acting like a two-dimensional electron gas (2DEG). The Kondo resonance signal is then a summation of contributions of both bulk and surface-state electrons near E_F. Therefore, the relative contribution of the surface state to the Kondo resonance has acquired great interests both experimentally and theoretically.[8,10,13–21]

Theoretical studies predict that on the noble metal (111) surfaces the amplitude $A(r)$ of the resonance due to the surface-state electrons falls off asymptotically as $1/r$ because of its two-dimensional nature, whereas $A(r)$ due to the bulk state electrons falls off as $1/r^2$.[10,18] Therefore, measuring the spatial decay of the Fano resonance has been a common method for an experimental evidence of the surface-state contribution.[10,16,18,20] Previous studies concluded that surface state is not important for the Kondo resonance in systems such as Co/Au(111), Ce/Ag(111),[18] and Co/Cu(111) (Ref. 10) from the lateral-distance dependence measurements of the resonances. However, the decay length is much longer when surface state is confined by obstacles such as the case of quantum mirage[8] and narrow terraces,[16] suggesting the importance of the surface state in these systems.

Other methods have been also reported for detecting the contribution of surface-state electrons. For example, the invariance of the Kondo temperature of Co atoms on Ag(111), 1 monolayer (ML) Ag/Cu(111), 1 ML Ag/Au(111), and 2 ML Ag/Au(111) indicates that the Kondo resonance is insensitive to the properties of surface states.[15] Similar conclusion was also deduced from a comparison of the Kondo resonances taken above Co atoms located on a terrace and near a monatomic step.[13,14] However, theoretical calculation again shows that surface state plays an significant role in Co/Cu(111) and Co/Au(111) systems.[20] Recently, a study comparing the Co-atom-induced resonances on Ag(111) surface with different terrace widths gives an evidence for a significant role of the surface-state electrons in the Kondo effect.[16] As mentioned above, both theoretical and experimental studies concluded contradictory results on different systems. One of the origins of the contradictory results could be related with a fact that surface state disappears around magnetic adsorbates in some of the systems. For instance, the onset of the surface state disappears in the tunneling spectra taken around the Ce adsorbates on Ag(111) surface[5] since the surface state is coupled with the bulk states by strongly bound adsorbates.[5,22] The absent or weakened surface states near E_F

could distort experimental results and make their interpretations contradictory.

As there is no evidence directly proving the contribution of surface states to the Kondo resonance, the challenge is to find a suitable substrate on which two-dimensional surface states dominate the density of states (DOS) around E_F. In this paper, we present a unique system: Si(111)-$\sqrt{3}\times\sqrt{3}$Ag surface, suitable for this purpose. The surface is metallic, having a quasi-free-electron two-dimensional surface state.[23] Since bulk silicon, having a band gap, does not have DOS near the E_F, the surface state dominates DOS near the E_F, which has been proved by both first-principle calculation[24] and angle-resolved photoemission spectroscopy.[23]

As a magnetic impurity, we use a 5, 10, 15, 20-Tetrakis-(4-bromophenyl)-porphyrin-Co (TBrPP-Co) molecule. Existence of a spin at the central Co atom has been confirmed by the observation of the Kondo resonance on Cu(111) surface.[25] Considering the high Kondo temperature of TBrPP-Co molecules (170 K) (Ref. 25) compared with that of Co atoms on Cu(111) (54 K),[10] Ag(111) (92 K),[11] and Au(111) (76 K) (Ref. 4) surfaces, caging the magnetic Co atom at the center of the porphyrin unit may enhance the Kondo temperature.

II. EXPERIMENTAL DETAILS

Our experiments were performed with an ultrahigh vacuum (UHV) low-temperature STM (USM-1300, Unisoku, and SPM-100, RHK) in which both of the tip and sample can be cooled by liquid He (LHe). All the STM images and the tunneling spectra shown in this paper were taken at 5 K in UHV ($\sim 7\times 10^{-9}$ Pa). The Si(111)-$\sqrt{3}\times\sqrt{3}$Ag substrate was prepared by Ag deposition (>1 ML) and subsequent annealing, whose details are found elsewhere.[26] TBrPP-Co molecules were evaporated onto the substrate using a home-made alumina cell wrapped by a tungsten wire. To avoid clustering and step-edge adsorption of the molecules and to obtain isolated adsorbed molecules on the substrate, the sample was cooled down to ~15 K by LHe during the molecular deposition. As a probe, an electrochemically etched tungsten tip was treated *in situ* using both e-beam heating and field ion spectroscopy (FIM). Removal of oxide layer on the tip was confirmed by an FIM observation of ring structures characteristic to the bare [110]-oriented tungsten tip.

III. EXPERIMENTAL RESULTS AND DISCUSSIONS

It has been known that the Si(111)-$\sqrt{3}\times\sqrt{3}$Ag surface has a surface state called S_1 near the E_F around the $\bar{\Gamma}$ point. Its parabolic and isotropic dispersion indicates a 2DEG property of the state.[23,27] Previous studies revealed that the energy level of the S_1 state shifts depending on the slight amount (a few percent of ML) of extra Ag adsorbates on the substrate[23,27] with its effective mass kept nearly constant ($0.12\ m_e$, where m_e is the free-electron mass) because of electron doping from the Ag adsorbates to the surface state. When the Ag amount is less than 1 ML and a 3×1 structure coexists, the binding energy of the state is above the E_F so that the surface becomes semiconducting.[27] When the Ag amount exceeds 1 ML, the binding energy is below the E_F and the surface state has finite DOS at the E_F.[23,27,28] To detect the surface-state contribution to the Kondo resonance, we must make sure that the surface is metallic having sufficient DOS near the E_F. To this end, we intentionally prepare a substrate with significant amount of extra Ag adsorbates on it.

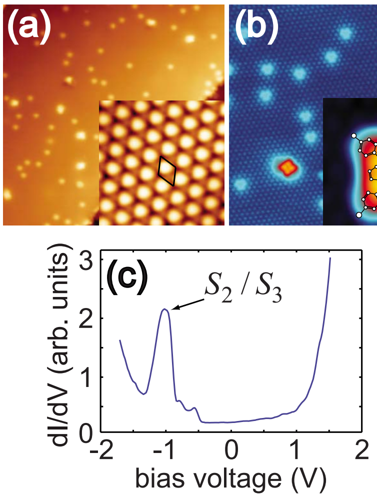

FIG. 1. (Color online) STM images of a Si(111)-$\sqrt{3}\times\sqrt{3}$Ag substrate with 0.002 ML TBrPP-Co molecules adsorbed in it. (a) large-scale STM image on which both the isolated TBrPP-Co molecules (larger dots) and Ag adsorbates (smaller dots) are found (size: 80 nm×80 nm, sample bias voltage V_s:2.0 V, tunneling current I_t:20 pA). Inset of (a) atomically resolved image of an uncovered region by the molecules. (b) Highly resolved image of the isolated TBrPP-Co molecules and Ag adsorbates. The background triangular lattice arises from the $\sqrt{3}\times\sqrt{3}$ structure of the substrate (size: 30 nm×30 nm, V_s:2.0 V, I_t:20 pA). Inset of (b) shows a planar-shape model of the TBrPP-Co molecule. (c) Typical tunneling conductance spectrum taken on the clean Si(111)-$\sqrt{3}\times\sqrt{3}$Ag region. The distinct peak at ~ -1 V corresponds to the S_2/S_3 surface states of the substrate.

Figures 1(a) and 1(b) show STM images taken after ~0.002 ML TBrPP-Co molecules deposition on the Si(111)-$\sqrt{3}\times\sqrt{3}$Ag substrate. There are two kinds of adsorbates visible on the surface: Brighter protrusions are single TBrPP-Co molecules and smaller dots are extra Ag adsorbates. The inset of Fig. 1(a) gives a zoomed image with atomic resolution. Since the experiments were performed at 5 K, the $\sqrt{3}\times\sqrt{3}$Ag surface is reconstructed in its low-temperature phase,[23,24] called the inequivalent-triangle (IET) structure. The unit cell of the structure is marked in the inset. On the Ag/Si(111) surface, all the isolated single TBrPP-Co molecules appear in a square shape, indicating a planar conformation of the molecule[29] [Fig. 1(b) and its inset]. This behavior is quite different from the adsorption of TBrPP-Co molecules on metal (111) surfaces,[25] where the isolated molecules show both planar and saddle conformations. From the STM images, we roughly counted the density of the extra Ag adsorbates and found it is around 0.007 Ag adsorbates per unit cell.

Figure 1(c) gives a tunneling conductance (dI/dV) spectrum taken on the clean $\sqrt{3}\times\sqrt{3}$Ag regions of the molecules

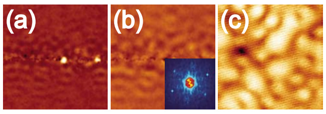

FIG. 2. (Color online) standing wave patterns observed on the same surface as that of Fig. 1. (a) and (b) dI/dV images taken in the same step-crossing area at different V_s [+393 mV for (a) and +293 mV for (b), I_t:140 pA, size: 30 nm×30 nm for (a) and 25 nm×25 nm for (b)]. The modulation amplitude and frequency are 3 mV and 713 Hz, respectively, for the dI/dV images. Inset of (b): Fourier-transformed pattern of (b). The central ring arises from the standing wave and the six surrounding dots are attributed to the $\sqrt{3}\times\sqrt{3}$ reconstruction of the substrate. (c) STM image taken with a small V_s (10.5 mV) to show DOS distribution at E_F. The image was taken on a flat terrace to avoid possible tip damage (size: 25 nm ×25 nm, I_t:100 pA).

adsorbed surface. Finite DOS near the E_F are clearly observed, suggesting that the bottom of the S_1 surface band is below the E_F and the surface is metallic. Besides that, there is a distinct peak located at ~ -1 V, characteristic to the S_2/S_3 states of the $\sqrt{3}\times\sqrt{3}$Ag substrate.[24,26,27] The observation of this peak was used to confirm the tip quality during the experiments.

However, we could not observe the onset of the S_1 surface state in the spectrum.[27] The onset is probably weak and/or strongly modified by the formation of the standing waves.

In order to further confirm the metallic S_1 state on the surface, we performed standing wave observations[30,31] at various sample bias voltages (V_s) [Figs. 2(a)–2(c)] on the same surface as Fig. 1. We took tunneling conductance (dI/dV) images with a lock-in method at $V_s=394$ mV [Fig. 2(a)] and 293 mV [Fig. 2(b)] near the same step edge expecting strong wave intensity. To make a DOS image near the E_F, an STM image was taken with a small bias voltage ($V_s=10.5$ mV) on a terrace [Fig. 2(c)]. The standing wave pattern in the DOS image taken at such small bias voltage gives a hint of the existence of metallic surface state. Besides that, from their Fourier transformed patterns [one shown in the inset of Fig. 2(b)] we extracted the wave number k of the standing waves at each bias voltage using six spots due to the $\sqrt{3}\times\sqrt{3}$ reconstruction as a reference. The obtained wave number is 1.3, 1.1, and 0.7 for Figs. 2(a)–2(c), respectively. Since the standing waves originate from the S_1 state,[23,27] we fitted these wave numbers with an equation of the parabolic dispersion: $E-E_F=\hbar^2k^2/2m^*+E_0$ with the fixed effective mass m^* of 0.12 m_e,[27] and found the bottom of the dispersion curve E_0 is located at ~ 115 mV below the E_F. In other words, we successfully prepared a substrate which has a two-dimensional metallic surface state whereas the contribution of the bulk states to the Kondo resonance is negligible since it is formed on the semiconducting bulk silicon, which does not have DOS at the E_F.[23]

In Fig. 1(b), the single molecules are surrounded by Ag adsorbates and some molecules are even located above the Ag adsorbates. For the Kondo resonance measurements, we picked up an isolated molecule whose closest Ag adsorbate is more than 5 nm away [Fig. 3(a)] to minimize the influence of the Ag adsorbates. It should be noted here that the molecule are picked up on the surface presented in Fig. 1 and that the surface state has finite DOS at E_F as demonstrated in Fig. 2. Figure 3(b) is a dI/dV spectrum taken above the molecular center using the lock-in technique, showing a clear peak near the zero bias voltage. We presumed this peak the expected Fano resonance and extracted the characteristic parameters such as the Kondo temperature T_k ($\sim 180\pm 10$ K), the interference-parameter q ($\sim 8\pm 2$), and the energy-shift ΔE ($\sim -3\pm 0.2$ mV) from a fitting with Eq. (1) at $r=0$ as drawn with a dotted line in Fig. 3(b). As mentioned before, q is a parameter related with an interference of the two channels contributing to the Fano resonance. The large acquired value of q (~ 8) here then implies that the tip is more strongly coupled to the discrete Kondo resonance rather than the metallic continuum state. Compared with the cases of other porphyrin-based molecules on various metal (111) surfaces,[25,32–34] the Kondo temperature of 180 K is rather high, indicating a significant coupling between the surface-state electrons and the spin of the magnetic atom. The T_k, which was measured on 1ML-Ag deposited Si(111), is also higher than those measured on Co atoms adsorbed on Ag(111), Ag/Cu(111), and Cu(111) surfaces.[12,15] We believe, however, that simple argument on the T_k difference based on the d-level occupation model[12] is not appropriate here since the atomic configuration[23,24] of the Ag/Si(111) surface is quite different from that of Ag(111) and so is the DOS at E_F. It has been reported that the DOS at E_F has a significant contribution to the Kondo temperature.[15]

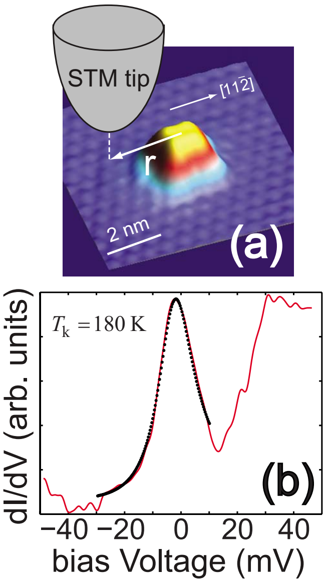

FIG. 3. (Color online) (a) three-dimensional image of an isolated TBrPP-Co molecule with the $\sqrt{3}\times\sqrt{3}$ triangular lattice. The arrow indicates the direction of the tip displacement for the measurement of the lateral-distance-dependent dI/dV spectra. (b) dI/dV spectrum taken above the center of the isolated molecule using a lock-in method with the modulation amplitude and frequency of 5 mV and 713 Hz, respectively. The dotted line shows a Fano-shaped fitting curve using Eq. (1) at $r=0$.

In the spectrum shown in Fig. 3(b), there are some background features, for instance, a steep increment in the conductance at 30 mV above the E_F. They probably originate

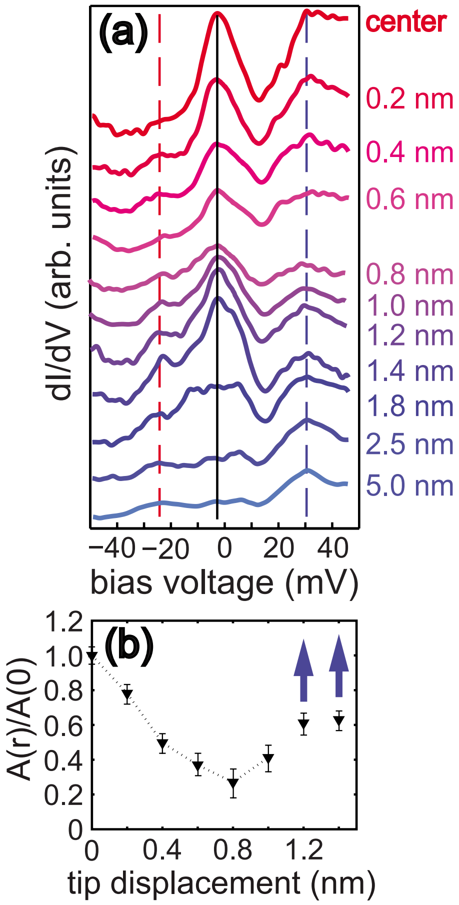

FIG. 4. (Color online) (a) series of dI/dV spectra with various tip displacements taken in a lock-in method with the modulation amplitude and frequency of 5 mV and 713 Hz, respectively. The stabilizing V_s and I_t were 130 mV and 100 pA, respectively, for $r=0-1.0$ nm and 1.8–2.5 nm, and 260 mV and 100 pA for $r=1.2$ and 1.4 nm. The vertical solid line indicates the Kondo resonance peaks located at 3 mV below the E_F. The dashed lines indicate the peaks located at around 30 and −25 mV with respect to the E_F. (b) normalized amplitude $A(r)/A(0)$ as a function of the lateral displacement r calculated from the dI/dV curves shown in (a). Since the molecule becomes unstable and easily rotatable at low bias voltages in the displacement of $r=1.2$ and 1.4 nm, the two spectra were taken at larger tip-sample gap distance by adjusting the stabilizing tunneling condition. The arrows in (b) imply that the amplitude at $r=1.2$ and 1.4 nm should be larger if the stabilization condition had been same as that of the other tip displacements.

from the electronic states of the substrate. As the Ag-induced reconstructed structure is formed on the semiconducting Si(111) surface, the background signal is much complicated comparing with that of the simple metal (111) surface.[27] A study on the spatial dependence of the spectra is, therefore, necessary to distinguish the Kondo resonance from the background signals.

Figure 4(a) shows a series of dI/dV spectra taken with various lateral tip distances (r) from the molecular center, as illustrated in Fig. 3(a), where the arrow shows the direction of the tip displacements. The peak amplitude of the resonance decreases with the lateral tip displacement, and up to 1.4 nm the peak is visible. We fitted each spectrum using Eq. (1) and found that T_k and q are nearly constant within their error margins in this distance range. The spectra taken at the distances larger than 2.5 nm, including the one taken on the clean $\sqrt{3}\times\sqrt{3}$Ag region, are almost same and do not have significant features near the E_F. We are quite sure that the long decay length of this system is not induced by the ligands the molecule has. Although it has been known that ligands significantly affect the Kondo resonance and temperature,[35] the positions the peak was observed at are far away from the ligands (1.6 nm > 1.5/2 nm), where 1.5 nm is the lateral size of the molecule. In fact, the Kondo resonance on a Co-porphyrin molecule, having ligands, adsorbed on a Pb thin film, which does not have surface states, decays quite rapidly, visible only up to 0.5 nm from its center.[33] We, therefore, believe the long decay length we observed is the surface-state contributions. Both theory and experiments suggested that the Kondo resonance amplitude due to bulk states decays rapidly in the lateral direction whereas the surface-state contribution stays longer from the magnetic impurity.[10,16,18,20] Previous studies also pointed out that the presence of any observable Kondo resonance at distances longer than 1 nm can be taken as an evidence of the surface-state contribution to the resonance.[10,16,18,20] The spectra we observed obviously demonstrate the characteristic features of the surface-state contributing Kondo resonance. In the dI/dV spectra, two additional peaks at 30 and −25 mV are marked with dashed lines. Since their intensities do not change so much with the tip displacements and they do not disappear on the clean Si(111)-$\sqrt{3}\times\sqrt{3}$Ag substrate, these two features are due to the electronic states of the substrate. As a conclusion, the distinct Fano-shaped peak near the E_F and its long decay length observed around the TBrPP-Co molecule adsorbed on the bulk-state-absent (near E_F) substrate are the direct convincing evidence that the surface state does play a significant role in the Kondo resonance.

In Fig. 4(b), we plotted the amplitude of the Kondo resonance $A(r)/A(0)$ as a function of the tip-displacement r. The amplitude decays gradually until $r=0.8$ nm. Surprisingly, the amplitude increases, reaching a maximum at $r=1.4$ nm, and then decays quickly and disappears at $r=1.8$ nm. Actually, the maximum amplitude is much larger than that shown in the plot, as marked with arrows. Because the molecule becomes unstable and easily rotatable at low-sample bias voltages[29] at $r=1.2$ and 1.4 nm, the two spectra were taken with larger stabilizing bias voltage and thus larger tip-sample gap distances. The reincreasing behavior of the resonance amplitude is similar to the case of Co adsorbates on confined Ag(111) terraces[16] and Co quantum mirage on Cu(111),[8] where the reincrement is attribute to the enhancement of the surface-states contribution due to their confinement by the narrow terrace and quantum corral. On the Si(111)-$\sqrt{3}\times\sqrt{3}$Ag surface, since only surface states contribute to the Kondo resonance, the resonance amplitude is more sensitive to the surface DOS. Although the molecules we measured are isolated and located in wide terraces, the standing waves induced by TBrPP-Co molecules and Ag adsorbates lead to significant modification in the DOS. The modulated DOS could be a possible reason of the observed peculiar dependence of the amplitude on the lateral displacement.

The peaks located at around 30 and −25 mV with respect to the E_F, as marked with the dashed lines in Fig. 4(b), are presumably due to the $\sqrt{3}\times\sqrt{3}$ surface contribution, since similar peaks are also observed on the $\sqrt{3}\times\sqrt{3}$ surface far from the molecule [e.g., $r=5.0$ nm in Fig. 4(b)]. Slight variation in the height and shape of the peaks is probably due to fluctuation in the tunneling gap distance during the spectrum measurements although we tried to measure them at constant tunneling condition. Depending on DOS between E_F and the stabilizing bias voltage, the gap distance changes and so do the measured spectra. Obviously electrical and/or mechanical vibration noises in the tunneling current also contribute to the variation in the spectra. Because of similarity of the energy level and shape and its nonlocal property, however, we believe the peaks originate from the surface, not the molecule.

IV. CONCLUSIONS

We prepared an ideal template on which only surface states contribute to DOS at the E_F for directly confirming the surface-state contribution to the Kondo resonance. The dI/dV spectrum taken above the Co-atom-caging molecule adsorbed on this bulk-state-absent (near the E_F) substrate contains a distinct peak near the E_F, which was proved to be the Kondo resonance by a series of the spectra with various tip displacements. Furthermore, the Kondo peak is visible up to 1.4 nm, in agreement with the theoretical results that the residual resonance at the lateral distance longer than 1 nm should originate from the surface-state contribution. The existence and long decay length of the Kondo resonance observed above the isolated TBrPP-Co molecule adsorbed on the Si(111)-$\sqrt{3}\times\sqrt{3}$Ag substrate presented in this paper gives the first direct evidence of the contribution of surface state to the Kondo resonance.

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[1] J. Kondo, Prog. Theor. Phys. **32**, 37 (1964).

[2] A. C. Hewson, *The Kondo Problem to Heavy Fermions* (Cambridge University Press, Cambridge, England, 1993).

[3] U. Fano, Phys. Rev. **124**, 1866 (1961).

[4] V. Madhavan, W. Chen, T. Jamneala, M. F. Crommie, and N. S. Wingreen, Science **280**, 567 (1998).

[5] J. T. Li, W.-D. Schneider, R. Berndt, and B. Delley, Phys. Rev. Lett. **80**, 2893 (1998).

[6] O. Újsághy, J. Kroha, L. Szunyogh, and A. Zawadowski, Phys. Rev. Lett. **85**, 2557 (2000).

[7] W. Chen, T. Jamneala, V. Madhavan, and M. F. Crommie, Phys. Rev. B **60**, R8529 (1999).

[8] H. C. Manoharan, C. P. Lutz, and D. M. Eigler, Nature (London) **403**, 512 (2000).

[9] V. Madhavan, W. Chen, T. Jamneala, M. F. Crommie, and N. S. Wingreen, Phys. Rev. B **64**, 165412 (2001).

[10] N. Knorr, M. A. Schneider, L. Diekhöner, P. Wahl, and K. Kern, Phys. Rev. Lett. **88**, 096804 (2002).

[11] M. A. Schneider, L. Vitali, N. Knorr, and K. Kern, Phys. Rev. B **65**, R121406 (2002).

[12] P. Wahl, L. Diekhöner, M. A. Schneider, L. Vitali, G. Wittich, and K. Kern, Phys. Rev. Lett. **93**, 176603 (2004).

[13] L. Limot, E. Pehlke, J. Kröger, and R. Berndt, Phys. Rev. Lett. **94**, 036805 (2005).

[14] L. Limot and R. Berndt, Appl. Surf. Sci. **237**, 572 (2004).

[15] M. A. Schneider, P. Wahl, L. Diekhöner, L. Vitali, G. Wittich, and K. Kern, Jpn. J. Appl. Phys. **44**, 5328 (2005).

[16] J. Henzl and K. Morgenstern, Phys. Rev. Lett. **98**, 266601 (2007).

[17] T. Kawasaka, H. Kasai, W. A. Dino, and A. Okiji, J. Appl. Phys. **86**, 6970 (1999).

[18] M. Plihal and J. W. Gadzuk, Phys. Rev. B **63**, 085404 (2001).

[19] P. S. Cornaglia and C. A. Balseiro, Phys. Rev. B **67**, 205420 (2003).

[20] J. Merino and O. Gunnarsson, Phys. Rev. Lett. **93**, 156601 (2004).

[21] C. Y. Lin, A. H. Castro Neto, and B. A. Jones, Phys. Rev. Lett. **97**, 156102 (2006).

[22] G. A. Fiete, J. S. Hersch, E. J. Heller, H. C. Manoharan, C. P. Lutz, and D. M. Eigler, Phys. Rev. Lett. **86**, 2392 (2001).

[23] S. Hasegawa, J. Phys.: Condens. Matter **12**, R463 (2000).

[24] H. Aizawa, M. Tsukada, N. Sato, and S. Hasegawa, Surf. Sci. **429**, L509 (1999).

[25] V. Iancu, A. Deshpande, and S.-W. Hla, Nano Lett. **6**, 820 (2006).

[26] M. Ono, Y. Nishigata, T. Nishio, T. Eguchi, and Y. Hasegawa, Phys. Rev. Lett. **96**, 016801 (2006).

[27] M. Ono, Ph. D. thesis, the University of Tokyo, 2006.

[28] R. I. G. Uhrberg, H. M. Zhang, T. Balasubramanian, E. Landemark, and H. W. Yeom, Phys. Rev. B **65**, R081305 (2002).

[29] Q. Li, S. Yamazaki, T. Eguchi, Y. Hasegawa, H. Kim, S.-J. Kahng, J. F. Jia, and Q. K. Xue, Nanotechnology **19**, 465707 (2008).

[30] Y. Hasegawa and Ph. Avouris, Phys. Rev. Lett. **71**, 1071 (1993).

[31] M. F. Crommie, C. P. Lutz, and D. M. Eigler, Nature (London) **363**, 524 (1993).

[32] A. Zhao *et al.*, Science **309**, 1542 (2005).

[33] Y. S. Fu *et al.*, Phys. Rev. Lett. **99**, 256601 (2007).

[34] L. Gao, W. Ji, Y. B. Hu, Z. H. Cheng, Z. T. Deng, Q. Liu, N. Jiang, X. Lin, W. Guo, S. X. Du, W. A. Hofer, X. C. Xie, and H. J. Gao, Phys. Rev. Lett. **99**, 106402 (2007).

[35] P. Wahl, L. Diekhöner, G. Wittich, L. Vitali, M. A. Schneider, and K. Kern, Phys. Rev. Lett. **95**, 166601 (2005).