Electronic structure of U$\text{(Ru}_{1-x}\text{Rh}_x)\text{Si}_2$ studied by laser angle-resolved photoemission spectroscopy

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Abstract. We have studied electronic structures of U$\text{(Ru}_{1-x}\text{Rh}_x)\text{Si}_2$ employing ultrahigh-resolution laser angle-resolved photoemission spectroscopy to understand the effect of Rh substitution. A hole-like dispersive feature, which presumably has Rh $d$-band character, was observed in U$\text{(Ru}_{1-x}\text{Rh}_x)\text{Si}_2$ for both $x = 0$ and $x = 0.03$. However, although a heavy quasiparticle band appears in the hidden-order state of URu$_2$Si$_2$ ($x = 0$), it was not observed for $x = 0.03$ over the temperature range studied. In addition, it was found that energy-distribution curves at the Fermi vector of the hole-like band for $x = 0.03$ behave similarly to the Fermi-Dirac function. We also present the mapping of Fermi surfaces formed by the hole-like band, which stay nearly unaffected by Rh substitution.

1. Introduction

A heavy fermion superconductor URu$_2$Si$_2$ shows an anomalous second-order phase transition at 17.5 K, whose order parameter is still unknown [1, 2]. Although its nature is a big mystery, the ‘hidden-order’ state below 17.5 K seems to be closely related to the antiferromagnetic phase, which appears as a bulk property under pressure [3]. Thus, the comparison between these two phases looks beneficial to understand the underlying nature of the hidden order. However, it is not always possible to carry out such comparative studies because some experimental techniques are inherently impossible to be performed under pressure.

The above problem may be compensated by the technique of chemical substitution, which are intensively carried out in the recent studies [4, 5]. In URu$_2$Si$_2$ system, the effect similar to the pressure effect can be obtained by substituting Rh for Ru, which induces the antiferromagnetic phase coexisting with the hidden order. In fact, this behavior helps various experimental techniques, such as angle-resolved photoemission spectroscopy (ARPES), to carry out comparative studies in URu$_2$Si$_2$ as a function of Rh concentration.

Motivated by recent advancements in both photoemission spectroscopy [6] and the substitution studies on URu$_2$Si$_2$ [4, 5], we have carried out ARPES experiments on U(Ru$_{1-x}$Rh$_x$)$_2$Si$_2$ ($x = 0$ and $0.03$) employing a state-of-art laser photoemission spectrometer. We observed a hole-like dispersive feature, which presumably holds Rh $d$-band character, in both...
Rh concentrations. However, we observed a heavy quasiparticle band only for \( x = 0 \) over the temperature range studied. In addition, the temperature dependence of the hole-like band for \( x = 0.03 \) follows the typical metallic behavior. We also present the mapping of Fermi surfaces formed by the hole-like band, which seems nearly unaffected by Rh substitution. These findings suggest that the emergence of the narrow band is related to the hidden-order state while some electronic states stay nearly unmodified.

2. Experimental

Single crystals of \( \text{U(Ru}_{1-x}\text{Rh}_x)\text{Si}_2 \) \((x = 0 \text{ and } 0.03)\) were prepared by the Czochralski method employing a tetra-arc furnace where the detail of synthesis has been described elsewhere \[7, 8\]. ARPES spectra were obtained employing laser ARPES system at the Institute for Solid State Physics, The University of Tokyo, where the system consists of a vacuum ultraviolet laser (photon energy of 6.994 eV) and a Gammadata-Scienta R4000WAL electron analyzer. We used circularly-polarized light unless otherwise noted. The (001) surfaces were obtained by \textit{in situ} cleaving in ultrahigh vacuum where the base pressure of the main chamber was kept better than \( 4 \times 10^{-11} \) Torr throughout the experiment. Temperature dependence of ARPES spectra were studied in the temperature range of \( 7 \text{ - } 26 \) K. Binding energies of spectra and sample temperature were calibrated in reference to the Fermi edge of a gold film evaporated near the sample, and the total energy resolution was set at 2 meV.

3. Result and Discussion

3.1. ARPES intensity plots

Figure 1 and Figure 2 illustrate ARPES data obtained for \( \text{U(Ru}_{1-x}\text{Rh}_x)\text{Si}_2 \) along (100) direction. Note that \( s \)-polarization was employed for these data, except for the one in Figure 1(c). In ARPES intensity plots at 7 K (Figure 1(a)) and corresponding energy-distribution curves (EDCs) (Figure 1(b)) for \( x = 0 \), we can observe a heavy band appearing near the Fermi level \((E_F)\). We can also see a hole-like dispersive feature, which is also confirmed to intersect \( E_F \) at 25 K with the Fermi vector \((k_F) \approx 0.12 \) Å (Figure 1(c)). In the data for \( x = 0.03 \) taken at 7 K and 25 K along (100) direction, however, we can only observe a hole-like band whose \( k_F \approx 0.14 \) Å (Figure 2). We note that these \( k_F \) values are close to the ones recently reported \[9\]. These observations show that although the existence of hole-like band is almost unvaried, the narrow band is extremely sensitive to the substitution of Rh.

3.2. Temperature dependence of energy-distribution curves

Figure 3(a) and 3(b) show the temperature dependence of EDCs for the same Rh concentrations where the cuts have been taken at the \( k_F \) of the hole-like band in each dataset. In Figure 3(a) for \( x = 0 \), one can see the development of the sharp peak with decreasing temperature. Moreover, the peak shifts toward higher binding-energy side as temperature is lowered. In Figure 3(b) for \( x = 0.03 \), however, spectra near \( E_F \) behave similarly to Fermi-Dirac function, and we are unable to detect the opening of gap. Being consistent with the observations seen in the ARPES intensity plots, remarkable difference, coming from the emergence of the narrow band, appears in energy-distribution curves.

3.3. Fermi Surface Topology

In Figure 4(a), we show the result of Fermi surface mapping performed for \( x = 0 \) at 25 K (paramagnetic phase). We observed a small hole-like Fermi surface where the positions of \( k_F \), obtained from fitting MDCs, are superimposed in the same figure. The Fermi surface may have small anisotropy in shape, but significant deviation from spherical shape is absent. An estimated cross-sectional area is \(~ 0.71 \) Å\(^2\), which is close to the hole-like Fermi surface observed in the
Figure 1. ARPES intensity plot along (100) direction measured for x = 0 sample: (a) Intensity map at 7 K, (b) EDCs at 7 K, and (c) MDCs at 25 K (where curves have been normalized to the area). Note that a concave-up dispersive feature, which does not cross the Fermi level, appears in the MDCs, and this feature is known as a surface state [9].

Figure 2. ARPES intensity plot along (100) direction measured for x = 0.03 sample: (a) Intensity map at 7 K, (b) EDCs at 7 K, and (c) MDCs at 25 K (where curves have been normalized to the area). Note that a concave-up dispersive feature, which is similar to the surface state shown in Figure 1(c), also appears in the MDCs for x = 0.03.

Figure 3. Temperature dependence of energy-distribution curves at the $k_F$ of hole-like band; (a) for x = 0, and (b) for x = 0.03. Note that spectra have been normalized to the intensity at $E_B = 12$ meV.

dHvA study [7]. Figure 4(b) illustrates the Fermi surface mapped for x = 0.03 at 26 K where s-polarization was employed. In the same way, we observed a hole-like Fermi surface, which size and shape anisotropy are comparable to the one for x = 0 within the experimental errors. This is consistent with the view that Rh substitution has small effect on the hole-like band.
Figure 4. Fermi surface mapping performed (a) for $x = 0$ at 25 K, and (b) for $x = 0.03$ at 26 K. Note that s-polarization was employed for (b), and the positions of $k_F$ obtained from fitting MDCs have been symmetrized and superimposed on the figures.

4. Conclusion
In this paper, we present the ARPES data on the system of U(Ru$_{1-x}$Rh$_x$)$_2$Si$_2$ and discuss the effect of Rh substitution. We show that substitution of Rh significantly affects the emergence of narrow band observed in the hidden-order state of URu$_2$Si$_2$. However, the effect of Rh substitution on hole-like bands appears rather small. These experimental data can be a good starting point to differentiate hidden-order state from others and would promote further systematic studies of the system of U(Ru$_{1-x}$Rh$_x$)$_2$Si$_2$.

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References
[1] Palstra T T M, Menovsky A A, Berg J V D, Dirkmaat A J, Kes P H, Nieuwenhuys G J and Mydosh J A 1985 Phys. Rev. Lett. 55 2727
[2] Chandra P, Coleman P, Mydosh J A and Tripathi V 2002 Nature 417 831
[3] Amitsuka H, Sato M, Metoki N, Yokoyama M, Kuwahara K, Sakakibara T, Morimoto H, Karawazaki S, Miyako Y and Mydosh J A 1999 Phys. Rev. Lett. 83 5114
[4] Yokoyama M, Amitsuka H, Itoh S, Kawasaki I, Tenya K and Yoshizawa H 2004 J. Phys. Soc. Jpn. 73 545
[5] Baek S-H, Graf M J, Balatsky A V, Bauer E D, Cooley J C, Smith J L and Curro N J 2010 Phys. Rev. B 81 132404
[6] Hüfner S 2007 Very High Resolution Photoelectron Spectroscopy (Berlin: Springer)
[7] Ohkuni H et al. 1999 Phil. Mag. B 79 1045
[8] Haga Y, Sakai H and Kambe S 2007 J. Phys. Soc. Jpn. 76 051012
[9] Santander-Syro A F, Klein M, Boarin F L, Nuber A, Lejay P and Reinert F 2009 Nature Phys. 5 637