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Signature of hidden order and evidence for periodicity modification in URu_2Si_2

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The detail of electronic structures near the Fermi level in URu_2Si_2 has been investigated employing state-of-art laser angle-resolved photoemission spectroscopy. The observation of a narrow dispersive band near the Fermi level in the ordered state as well as its absence in a Rh-substituted sample strongly suggest that the emergence of the narrow band is a clear signature of the hidden-order transition. The temperature dependence of the narrow band, which appears at the onset of the hidden-order transition, invokes the occurrence of periodicity modification in the ordered state, which is shown for the first time by any spectroscopic probe. We compare our data to other previous studies and discuss possible implications.

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I. INTRODUCTION

The heavy fermion material URu_2Si_2 is an intriguing correlated-electron material, as this material exhibits superconductivity ($T_c \sim 1.5$ K) and mysterious second-order phase transition at ~17.5 K(T_{HO}).[1–3] The latter order is characterized by a large entropy release and clear anomalies have been confirmed in transport properties at the ordering temperature.[1–3] It has been suggested by various experiments that reconstruction of the Fermi surface occurs at T_{HO}.[3–9] Initially, the order at T_{HO} was ascribed to the formation of spin-density wave, but the moment detected by neutron scattering (~0.03 μ_B) is too small to account for the entropy release.[10] This puzzle has led to the proposal of various theories[11–28] but microscopic measurements have been unsuccessful in revealing the underlying nature of the phase transition. This is known as the "hidden-order" problem today,[29] and the possibility of new-type phase transition is anticipated. The hidden-order phase is closely related to large-moment antiferromagnetism (LMAF), which appears in URu_2Si_2 under pressure with ordering wave vector of (001).[30] It is worth mentioning that although the ordering wave vector of the hidden order is not fully established, neutron scattering has suggested that the vector may be incommensurate[31] or same as the case of LMAF.[32]

Recent state-of-art experiments have revealed several important aspects of the hidden order but there is much work left to be done. One of the important points that must be understood is *the evolution of electronic structures upon the hidden-order transition*. Electronic structures can be probed directly, for example, by means of angle-resolved photoemission spectroscopy (ARPES) in momentum space, or scanning tunneling spectroscopy (STS) in real space. There are several pioneering ARPES studies on URu_2Si_2 (Refs. 33–35) but they have investigated only the paramagnetic phase. In the recent high-resolution ARPES study, the shift of a heavy band to lower energies is observed around the Fermi level (E_F), where the band appears right at E_F in the vicinity of T_{HO}.[36] It is not fully established, however, if this observation is really intrinsic to the hidden-order transition since the available high-resolution data set is restricted to a single experiment (only with photon energy of 21.2 eV, thus the probed region of the momentum space is limited) so far. It is also important, for further clarification of the connection between the hidden order and the heavy band, to know how the band behaves upon the suppression of the hidden order. Moreover, the ARPES work together with the latest STS studies report the absence of band backfolding/"conventional" density-wave feature[36–38] but further thought is required to link these results with the results of neutron scattering, which have hinted some kind of periodicity modification.[31,32] From these issues, it is clear that the investigation of the electronic structures is still in the early stage, and further experimental effort is necessary.

In this paper, we report the detailed investigation of electronic structures near E_F of $U(Ru_{1-x}Rh_x)_2Si_2$, by means of state-of-art laser angle-resolved photoemission spectroscopy (laser ARPES). Laser ARPES allows us to observe the emergence of a narrow band near E_F at temperatures below T_{HO} with bright clarity. The narrow band is not observed, however, in Rh-substituted samples in which the hidden order is suppressed,[39,40] and this fact further confirms that the band is intrinsically linked to the hidden order. Moreover, the temperature dependence of the narrow band appears significantly different from the one in the previous work; we observe the energy shift and the spectral-weight enhancement of the narrow band only below T_{HO}, and the narrow band is not observed above T_{HO}. This marked observation invokes the occurrence of periodicity modification, and our observation emphasizes that the hidden-order transition is strongly coupled to the electronic degree of freedom.

II. EXPERIMENTAL METHOD

In our study, single crystals of $U(Ru_{1-x}Rh_x)_2Si_2$ (x=0 and ~0.03) were prepared by the Czochralski method employing a tetra-arc furnace.[41,42] ARPES data were collected employ-

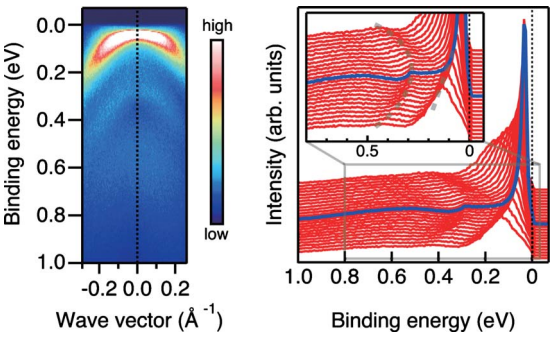

FIG. 1. (Color online) Wide scan measured at 25 K for [100] direction: (left panel) ARPES intensity plot, (right panel) corresponding energy-distribution curves. In the right panel, the bold curve corresponds to the cut at 0 Å^{-1}, and the broken lines are guide for the eyes. The maxima of a concave-up dispersive band is clearly seen at around $E_B \sim 300$ meV.

ing the system of a vacuum ultraviolet laser (photon energy of 6.994 eV) and a Gammadata-Scienta R4000WAL electron analyzer.[43] Circularly polarized light was used unless otherwise noted.[44] Samples were cleaved *in situ* to obtain (001) surface, and measurements were done in the temperature range of 7–25 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. The base pressure of main chamber was kept better than 4×10^{-11} Torr throughout. Our laser ARPES system seemed to allow much longer surface lifetime than a conventional discharge lamp system, evidenced by the fact that the signal of quasiparticle peak near the Fermi level (will be discussed later) could be obtained at even 36 h after cleavage.

III. RESULTS

In Fig. 1, we show an ARPES intensity scan up to the binding energy (E_B) of 1 eV (referred as "wide scan") measured at 25 K for [100] direction. A sharp dispersive feature is observed near E_F, which corresponds to the surface state reported in the previous work. (The near-E_F detail will be described in the next paragraph.) We also observe a concave-down dispersive feature at $E_B \sim 300$ meV, which is also evident in energy-distribution curves. The latter dispersive feature is seen around Z point in both recent band structure calculation employing much accurate lattice parameters and the result of recent soft x-ray ARPES measurement.[45] In addition, we do not observe the bottom of concave-up dispersive feature at $E_B \sim 0.7$ eV, whose absence should distinguish Z point from Γ point.[35]

Figures 2(a) and 2(b) show typical ARPES intensity plots for URu_2Si_2 at 7 K measured along [110] and [100] directions, respectively. In these plots, we find strong photoemission intensities not crossing E_F (surface state shown in Fig. 1) and a holelike dispersion, which does cross E_F. The feature of the holelike dispersion is also visible in the momentum-distribution curves (MDCs) for [110] direction, labeled as Fig. 2(d). More importantly, a sharp feature near

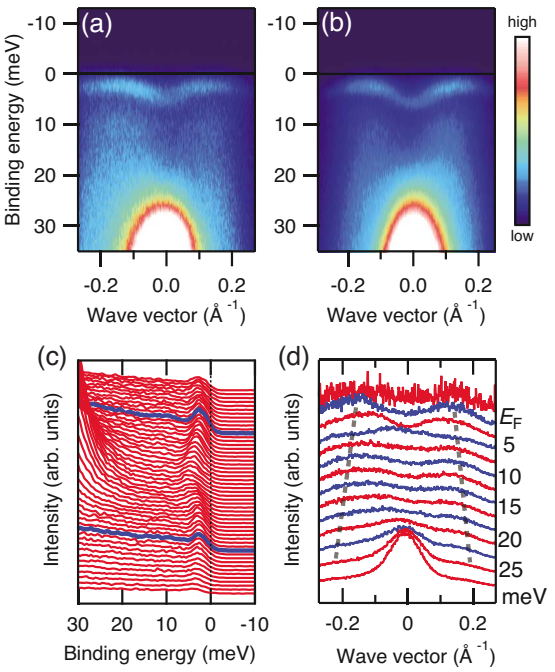

FIG. 2. (Color online) Laser ARPES data obtained at 7 K for URu_2Si_2 (a) ARPES intensity plot for [110] direction and (b) for [100] direction, (c) energy-distribution curves, and (d) momentum-distribution curves for [110] direction. The bold curves in (c) corresponds to the spectra at k_F, and the broken lines in (d) are guide for the eyes.

the Fermi level is clearly seen in both ARPES intensity plots at 7 K, as also evidenced by energy-distribution curves (EDCs) for [110] direction [Fig. 2(c)] as well. We estimate the group velocity of quasiparticles to be ~30 meV Å from the slope between 0 and 0.15 Å^{-1} in the [110] plot, being similar to the estimates in previous works.[31,36] We also estimate the Fermi vector (k_F) of the holelike band from data taken at 25 K for both [110] and [100] directions (not shown) and obtain ~0.15 Å^{-1} and ~0.12 Å^{-1}, respectively. These are close to the values previously reported.[36] Moreover, the width of the narrow band (W) is estimated to be ~3 meV, and the leading edge closest to $E_F(k_{LE})$ almost coincides the k_F of the holelike band in each direction. By using the formula employed in the previous work ($m^* = |\hbar^2 k_{LE}^2 / 2W|$; where m^* is the effective mass), we obtain a rough estimate of m^*/m_e to be ~18–29 (m_e; the free electron mass), a fairly reasonable value suggested by earlier works.[3,36]

In Fig. 3, we show the result of Fermi surface mapping performed for URu_2Si_2 at 25 K (paramagnetic phase), where a small holelike surface is observed. The positions of k_F, obtained from MDC fitting of three data sets, are superimposed in the same data. The Fermi surface may have small anisotropy in shape (may be octagonal-like) but significant deviation from a spherical shape is absent. An estimated cross-sectional area is 0.71 Å^{-2}, which should correspond to the holelike surface observed in the de Haas-van Alphen study.[41]

Another to show is ARPES data on partially Rh-substituted samples, where the substitution of Rh for Ru is known to suppress the hidden order and to induce antiferro-

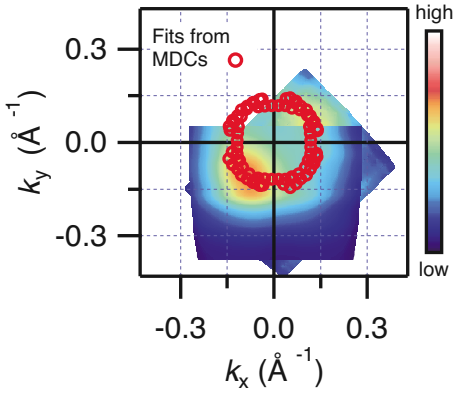

FIG. 3. (Color online) Intensity mapping at E_F performed for URu_2Si_2 at 25 K. Positions of k_F obtained from MDC analysis are also plotted for three data sets. Note that fitting data have been fourfold symmetrized.

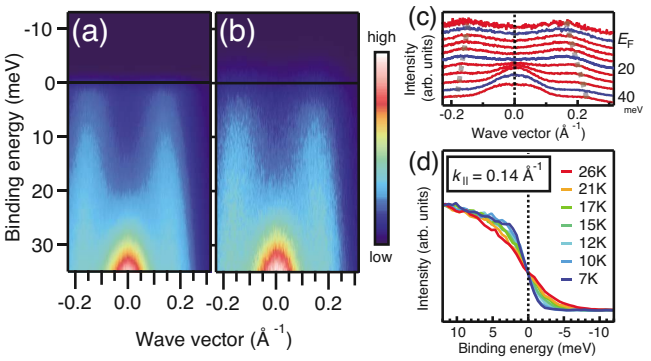

FIG. 4. (Color online) ARPES intensity plots for $U(Ru_{1-x}Rh_x)_2Si_2(x\sim0.03)$ measured for [100] direction with *s*-polarized light (a) at 7 K and (b) at 26 K. Momentum- distribution curves taken at 26 K are shown in Fig. 4(c) and describes the existence of a holelike dispersive band. Note that the broken lines in (c) are guide for the eyes. Figure 4(d) shows EDCs taken at $k_{||}$ =0.14 Å^{-1}. EDCs are normalized to the area.

magnetic region, whose fraction is dependent on the Rh concentration.[39,40] We carry out ARPES measurements on $U(Ru_{1-x}Rh_x)_2Si_2(x\sim0.03)$ along [100] direction. Figures 4(a) and 4(b) display ARPES intensity plots measured at 7 K and 26 K, respectively, where *s*-polarized light is employed. The ARPES data taken at 7 K shows intensity around $E_B\sim35$ meV and a holelike band, both of which also appear in the intensity plot at 26 K. Figure 4(c) shows that the holelike dispersion crosses E_F at $k_F\sim0.14$ Å^{-1}, being similar to what was observed for the nonsubstituted sample. However, it is surprising that the data do not show any signature of a narrow band at all. We do not observe a narrow band for circularly polarized light either (not shown), confirming that the result is not due to matrix-element effect. EDCs shown in Fig. 4(d) illustrate that the holelike band behaves similarly to the Fermi-Dirac function over the temperature range of 7–26 K.

Next, we discuss the temperature evolution of ARPES intensity plots near E_F obtained for [100] direction of URu_2Si_2 through Figs. 5(a)–5(g). Note that the intensity plots have been normalized by the resolution-convoluted Fermi functions, and the data are obtained and reproduced on single cleaves. First of all, one can see that the surface state and the holelike dispersion are present at all investigated temperatures. At temperatures above T_{HO}(18–25 K), no feature of the narrow band is observed. This is in sharp contrast to the result of previous high-resolution ARPES study which reports that the quasiparticle band lies at $E_B\sim5$ meV at 26 K and appears right at E_F at 18 K.[36] On the other hand, at temperatures below T_{HO}(7–15 K), the narrow dispersive feature is clearly seen near E_F. It is remarkable that not only the narrow band shifts to higher binding energies but also its spectral intensity increases as temperature is lowered. The above mentioned temperature dependence of the narrow band can be easily seen in the energy-distribution curves, depicted in Figs. 5(h)–5(j) (spectral cuts at $k_{||}$=0 Å (cut A), spectral cuts at $k_{||}$=−0.12 Å (cut B), and angle-integrated spectra, respectively). Note that spectra are normalized to the intensity at E_B=12 meV. One can confirm that a sharp peak is not present around E_F at temperatures above T_{HO} but shows up below T_{HO}. Also, these energy-distribution curves substantiate the shift of the band to higher binding energies as lowering temperature. The shift between 15 and 7 K is estimated to be 1.7 meV for cut A and 1.5 meV for cut B, being same within experimental error. This suggests that the entire portion of the narrow band shifts toward higher binding-energy side. In Fig. 5(k), we show the temperature dependence of integrated spectra obtained in the similar manner for [110] direction with *s*-polarized lights. One can confirm that the trends of the band shift and the spectral weight enhancement are also seen for [110] direction. More importantly, the spectrum measured at 17 K shows a small hump at $E_B\sim2$ meV which is certainly bigger than the noise level, and this signal becomes stronger at 16 K or lower temperatures. On the other hand, the spectrum obtained at 18 K does not show such peak and looks identical to the spectrum at 20 K within the experimental resolution. Therefore, the data for [110] direction also support the appearance of the narrow band at the onset of T_{HO}.

IV. DISCUSSION

From now on, we discuss the implications of present ARPES study. There are mainly two important points to discuss, namely, the nature of the narrow band and its temperature dependence above and below T_{HO}.

First of all, our data should be signified by the clear observation of a narrow dispersive band below T_{HO}. The observation of the dispersive band, which is even clear in the raw data, rules out the possibility that the band is of extrinsic origin. Moreover, our data is consistent with other works, including the recent STS studies which appears during the preparation of this manuscript.[37,38] For example, the effective mass and the group velocity is in general agreement with the values deduced from thermodynamic measurements and neutron scattering.[3,7,31] The peak position in angle-integrated data at 7 K(~3.6 meV) is also consistent with the observations in recent STS studies.[37,38] We are unable to conclude if the observed change indicates the opening of the gap solely from our data since the signal in the thermally occupied re-

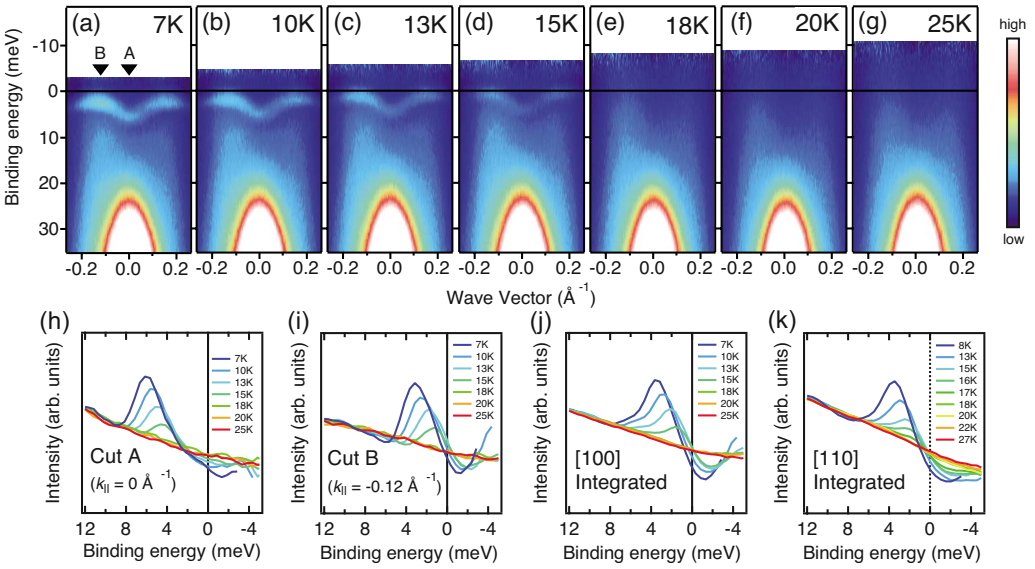

FIG. 5. (Color online) [(a)–(g)] Temperature evolution of ARPES intensity plots for URu_2Si_2 measured along [100] over the temperature range of 7–25 K, (h) EDCs for the above data taken at $k_{||}=0$ Å^{-1} (cut A) and (i) EDCs at $k_{||}=-0.12$ Å^{-1} (cut B). (j) angle-integrated spectra for [100] ARPES intensity plots, and (k) angle-integrated spectra for [110] ARPES intensity plots obtained with *s* polarization. Note that spectra are normalized by the intensity at $E_B=12$ meV.

gion (up to $4-5\ k_BT$ above E_F) does not show any hump structure. However, the gap observed by STS is reported to be particle-hole asymmetric, which may explain the diplike spectral shape near E_F formed only below T_{HO}. In addition to the above-mentioned consistency with earlier works, intrinsic nature of the narrow band is also corroborated by the data of Rh substitution. ARPES data on $U(Ru_{1-x}Rh_x)_2Si_2(x \sim 0.03)$ show the absence of the narrow band at 7 K where a clear signal of the narrow band is observed in nonsubstituted sample at the same temperature. Since 3% Rh-substituted sample is expected to be antiferromagnetic rich,[39,40] the observation strongly suggests the close link between the hidden order and the emergence of the narrow band. From these evidences, we conclude that the appearance of the narrow band must be a clear signature of the hidden-order transition.

The second important point to consider is the temperature dependence of the narrow band. It is notable in the present data that the narrow band appears only below T_{HO} and its spectral weight enhances as temperature is lowered. This behavior promptly suggests the modification of band structures but it is also unique since the change is not restricted in the immediate vicinity of k_F. The most natural way to explain this behavior is to invoke the change in symmetry upon the hidden-order transition. Then, appearance of a new band at the onset of phase transition is reasonably explained by the change in symmetry causing backfolding of the band. Spectral weight enhancement is also expected in this framework as it can occur as the order parameter grows. Note that ARPES has been capable of successful observations of band backfolding in various materials.[46–48] In the case of URu_2Si_2, strong-coupling nature of the hidden-order transition[37] favors a folded band to be observable because the ARPES intensity of a folded band is proportional to the strength of coupling.[46] More importantly, this interpretation would bridge between the data of two different experiments; while the change in symmetry associated with the hidden order transition is supported by inelastic neutron scattering,[31,32] the present work supports this idea from a spectroscopic point of view.

Next, the above discussion would raise a question if it is possible to reconcile our result and the previous spectroscopic studies which report the absence of band backfolding or conventional density-wave feature.[36–38] To consider this, we recall that the previous studies do not exclude the possibility of some form of periodicity modification along out-of-plane (*c* axis) direction, and this can consistently explain all the results. We speculate that one of the reasonable possibilities is the doubling of the unit cell along *c* axis[32] since it can even explain the discrepancy in the temperature dependence between the two high-resolution ARPES studies as well as the variety of observations. We note that upon the transition from paramagnetic phase to LMAF phase, it is well known that the unit cell doubles since the U atoms at (0, 0, 0) and at $(a/2,a/2,c/2)$ becomes inequivalent. If similar doubling of the unit cell occurs along *c* axis upon the hidden-order transition, a band at Γ would be back folded into *Z* point. In the previous ARPES study, He I measurement shows a similar narrow band, which is claimed to be near Γ point.[36] On the other hand, there are several indirect evidences, which support that laser ARPES reflects the electronic structures around *Z* point. As mentioned earlier, the feature observed near *Z* point in the recent soft x-ray ARPES study is consistent with the wide scan data.[45] Moreover while we observed a small Fermi surface, ARPES experiments,[34,45] de Haas-van Alphen experiment,[41] and positron annihilation study[49] consistently support its existence at *Z* point. The doubling of the unit cell is also consistent with the reentrant behavior of the hidden order,[50] a recent group theory analysis,[28] or symmetry-breaking scenario.[24] We also notice, during the revision process, that possible existence of superstructure along *c* axis is suggested by a recent normal emission

ARPES experiment.[51] Thus, these evidences should provide good motivation for further experimental studies to verify the proposal of unit-cell doubling. We believe that normal emission ARPES in *ultrahigh-resolution* (enough to observe the narrow band in the hidden-order state) would be an informative experiment. Nevertheless, present work demonstrates the symmetry change upon the hidden-order transition and emphasizes the strong coupling between the hidden order and the electronic degree of freedom as well as the importance to understand the information along c axis.

V. CONCLUSION

In this paper, we have shown the detailed investigation of electronic structures near E_F in URu_2Si_2. Laser ARPES measurements on $U(Ru_{1-x}Rh_x)_2Si_2$ for both $x=0$ and $x\sim0.03$ ensures the appearance of a narrow band near E_F is a clear signature of the hidden order. The temperature dependence of the narrow band, which appears only below T_{HO} and its spectral weight enhances with lowering temperature, provides the first spectroscopic evidence for the change in the symmetry upon the hidden-order transition.

Note added in proof: Recently, we became aware of related work by Oppeneer.[52]

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