Band structure and Fermi surface of URu_2Si_2 studied by high-resolution angle-resolved photoemission spectroscopy

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We have performed a high-resolution angle-resolved photoemission spectroscopy (ARPES) on single-crystal URu_2Si_2 to study the band structure and the Fermi surface in the paramagnetic phase. The valence-band structure consisting of the Ru $4d$ and Si $3p$ states shows a qualitatively good agreement between the ARPES experiment and the band-structure calculation. In the vicinity of the Fermi level (E_F), we observed a less dispersive band, which crosses E_F midway between the Z and X points in the Brillouin zone. Comparison with the band calculation as well as with the de Haas–van Alphen (dHvA) result suggests that the experimental band near E_F is assigned to the largest hole pocket centered at the Z point, which has a strong U $5f$-Ru $4d$ hybridized character. The observed remarkable narrowing of the near-E_F band compared with the band-structure calculation suggests a strong renormalization effect due to the electron correlation of U $5f$ electrons. [S0163-1829(99)01343-0]

I. INTRODUCTION

The heavy fermion superconductor URu_2Si_2 has attracted much attention because of its anomalous physical properties as represented by the coexistence of the antiferromagnetic ordering ($T_N = 17.5\,\mathrm{K}$) and the superconductivity ($T_c = 1.5\,\mathrm{K}$).[1–5] The electronic specific heat coefficient (γ) shows a drastic change at T_N from $\gamma = 180$ to 50 $\mathrm{mJ/mol\,K^2}$ (Ref. 6), suggesting the development of a charge- or a spin-density wave below T_N (Ref. 2). The complicated electronic and magnetic properties of URu_2Si_2 at low temperatures have been regarded to originate in the fundamental character of U $5f$ electrons.[5] The U $5f$ states located near the Fermi level (E_F) are expected to strongly hybridize with the Ru $4d$ orbital near E_F and produce the complicated Fermi surface(s), leading to the various electronic and magnetic orderings at low temperatures. The Fermi surface of URu_2Si_2 has been intensively studied by the de Haas-van Alphen (dHvA) effect measurements,[7–9] which have reported several independent oscillations ascribable to the Fermi surfaces in URu_2Si_2. However, the experimental results are not necessarily consistent with each other and it is hard at present to discuss the Fermi-surface topology in detail. An alternative information from a different experimental technique has been required.

Angle-resolved photoemission spectroscopy (ARPES) is a unique and powerful experimental technique to study the electronic band structure of materials.[10] ARPES has been employed on various materials and successfully established the valence-band structure. However, the energy resolution was not necessarily enough to discuss the detailed electronic structure near E_F, in particular for the so-called f-electron materials where $4f$ or $5f$ electrons produce the complicated narrow ''bands'' near E_F. A recent remarkable improvement in the energy and angular resolutions in ARPES has enabled the precise probing of the momentum-resolved electronic structure near E_F. Using high-resolution (HR) ARPES, we have studied the detailed band structure near E_F of some $4f$- and $5f$-electron materials such as CeSb (Ref. 11) and UPt_3 (Ref. 12) and demonstrated the high capability of HR-ARPES to study the Fermi-surface topology and its evolution as a function of temperature.

In this paper, we report the result of our HR-ARPES study on URu_2Si_2. We have successfully determined the whole valence-band structure consisting of mainly the Ru $4d$ and Si $3p$ states. The result shows a good qualitative agreement with the band-structure calculation. Beside the highly dispersive Ru $4d$ and Si $3p$ bands, we also found a less dispersive narrow band near E_F, which crosses E_F midway between two high-symmetry points in the Brillouin zone and forms a large Fermi surface. We compare the present ARPES result with the band-structure calculation as well as with the dHvA to discuss the Fermi-surface topology and the renormalization effect due to the strong electron correlation in the band structure.

II. EXPERIMENT

URu_2Si_2 single crystals (typically 3–4 mm in diameter and 80 mm in length) were grown by the Czochralski method with a tetra-arc furnace. The starting materials were

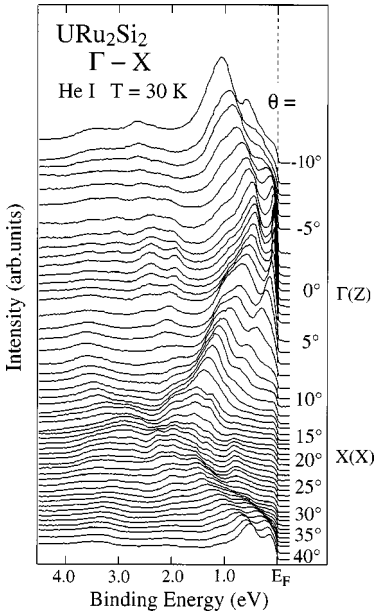

FIG. 1. ARPES spectra of URu_2Si_2 measured at 30 K (paramagnetic phase) with He I resonance line (21.2 eV) in the ΓX-XRZ plane in the Brillouin zone. Polar angle (θ) with respect to the surface normal is indicated.

99.95% pure U, 99.99% pure (4N) Ru and 5N Si. The obtained crystals were annealed at 900 °C for a week under vacuum and then characterized by x-ray diffraction to confirm the single crystallinity.

Photoemission measurement was carried out using a home-built high-resolution photoemission spectrometer, which has a hemispherical electron energy analyzer and a highly bright discharge lamp. The base pressure of the spectrometer is 3×10^{-11} torr and the angular resolution is $\pm1°$. The energy resolution was set at 30–50 meV for quick data acquisition because of the relatively fast degradation of sample surface. A clean mirrorlike surface of URu_2Si_2 (001) plane was obtained by *in situ* cleaving at 30 K just before measurement and kept at the same temperature during the measurement. Since we observed degradation of the sample surface as evident by the gradual increase of background in the spectrum, we recorded all the spectra within twenty hours after cleaving. We have checked that the spectral feature was unchanged within this time interval. We measured three sets of ARPES spectra using different samples and confirmed the reproducibility. The Fermi level of the samples was referred to that of a gold film evaporated on the sample substrate and its accuracy is estimated to be less than 5 meV.

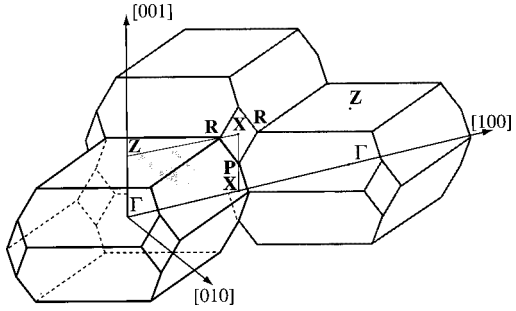

FIG. 2. Brillouin zone of URu_2Si_2 with a body-centered tetragonal crystal structure. ARPES measurement was performed in the ΓX-XRZ plane.

III. RESULTS AND DISCUSSION

Figure 1 shows a set of ARPES spectra of URu_2Si_2 measured with the He I resonance line (21.2 eV) at 30 K for the ΓX-XRZ plane in the body-centered tetragonal Brillouin zone (Fig. 2). Polar angle θ measured from the surface normal of the cleaved (001) plane is denoted on spectra. We find that the position and intensity of structures in ARPES spectra are very sensitive to the polar angle. For example, we find several prominent peaks (bands) located near E_F around $\theta=0°$, which show a remarkable energy dispersion toward

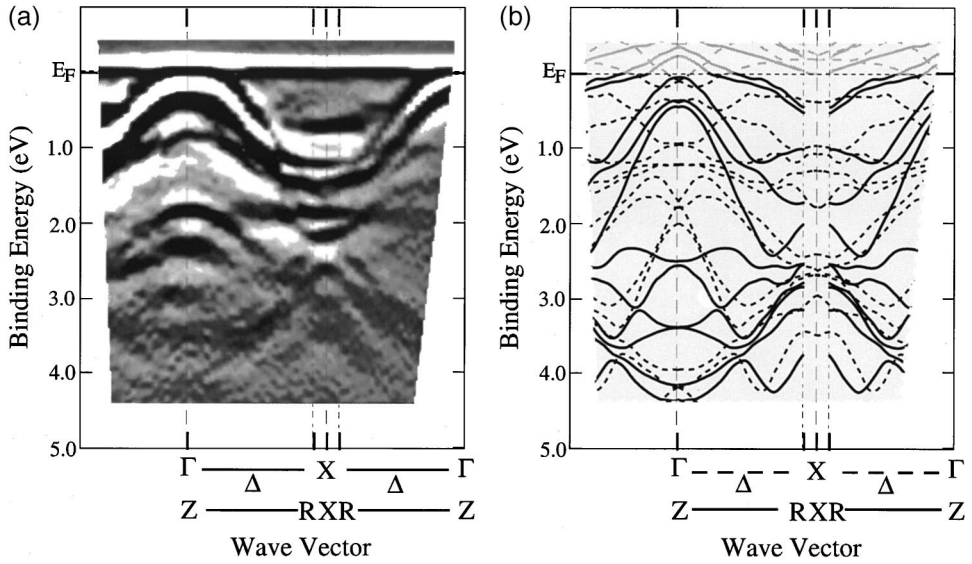

FIG. 3. Comparison of the valence-band structure of URu_2Si_2 between (a) the ARPES experiment and (b) the band calculation (Ref. 16). The band-structure calculation has not presented the result between R and X points. Note the qualitatively good agreement in the band dispersions between the experiment and the calculation.

the higher binding energy with increasing or decreasing the polar angle. Since the polar angle of 0° corresponds to the high-symmetry point $\Gamma(Z)$ in the Brillouin zone (see Fig. 2), we find that these bands show a symmetric dispersive feature with respect to the $\Gamma(Z)$ point. These dispersive bands appear to have a bottom at 1.5–2 eV binding energy around $\theta=22°$, which roughly corresponds to the Brillouin-zone boundary (X point). With further increasing the polar angle ($\theta=30°-40°$), these bands again approach E_F as if the bands have a mirror symmetry with respect to $\theta=22°$. This indicates that the bands enter the next Brillouin zone and the ARPES spectrum traces the band dispersions from the $X(X)$ point to the $\Gamma(Z)$ point in the next Brillouin zone (see Fig. 2). Thus, we find that the measured ARPES spectra follow well the periodicity of the bulk Brillouin zone and reflect the electronic structure of URu_2Si_2.

In order to see more directly the dispersive feature of bands as well as to compare it with the band-structure calculation, we have mapped out the ''band structure'' of URu_2Si_2 from the ARPES spectra. The results are shown in Fig. 3(a) for the ΓX-XRZ plane. The experimental band structure has been obtained by taking the second derivative of ARPES spectra after moderate smoothing and plotting the intensity in a square-root scale by gradual shading as a function of the wave vector and the binding energy.[10,13] Using this numerical method, we can avoid artificial errors observed in a case of picking up the peak position by hand and also we can delete a background effect due to the spurious secondary electrons, which masks the real-energy position of peaks. In Fig. 3(a), dark areas correspond to the experimentally determined bands. We set the gray-scale level so as to have the apparent bandwidth in the gray-scale image being almost equal to the full width at half maximum of the corresponding peak in ARPES spectra.

Before comparing the experimental results with the calculation, we briefly explain what ''band structure'' the ARPES results correspond to. In the present experimental setup (see Fig. 2), we observe the electronic structure of the ΓX-XRZ plane in the Brillouin zone. In the photoemission process, the momentum of photoelectrons parallel to the surface is conserved owing to the existence of translational symmetry. In contrast, the above is not the case for the momentum perpendicular to the surface because of the surface potential. More importantly, the momentum itself has an uncertainty due to the very short escape depth of photoelectrons. Thus the momentum of photoelectron perpendicular to the surface becomes broad when the photoelectron escapes from the surface. This means that the ARPES spectrum reflects the electronic structure averaged out in the direction perpendicular to the surface (ΓZ direction in this case, see Fig. 2). As a result, the high-symmetry lines in the Brillouin zone are likely to appear as prominent structures in the ARPES spectrum because the density of states on the high-symmetry lines is relatively large. This indicates that the experimental band structure obtained by ARPES is compared with the band-structure calculation performed for the high-symmetry lines.[13,14] Of course, this interpretation is valid when the momentum broadening is comparable to or larger than the Brillouin zone (BZ) size in the perpendicular direction. The transition-matrix element and the final state effects may change the peak position in the spectrum.[15] Thus, it is noted

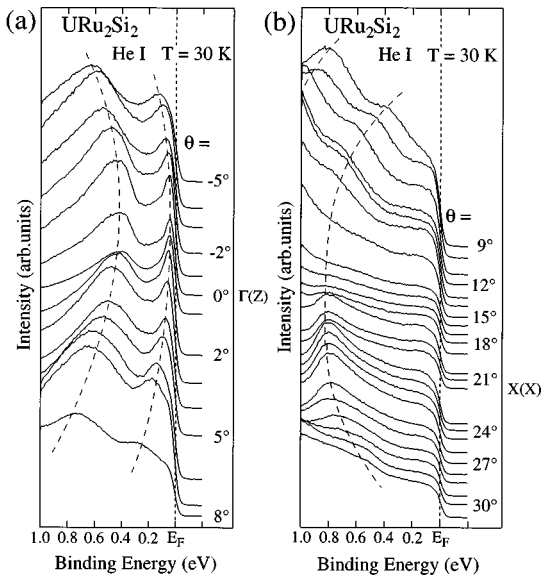

FIG. 4. ARPES spectra near E_F of URu_2Si_2 measured at 30 K for the momentum regions around (a) $\Gamma(Z)$ and (b) $X(X)$ points in the Brillouin zone. The energy dispersions of the Ru $4d$ bands are roughly traced with dashed lines.

here that the present interpretation method is based on the assumption of a large momentum broadening and a small transition matrix and final state effect. In this study, we have observed some indirect experimental evidences for the substantial momentum broadening perpendicular to the surface. In Fig. 3(a), we find that (1) almost of all the experimental bands are symmetric with respect to the $X(X)$ point at the Brillouin-zone boundary and (2) the experimental band dispersions around the $\Gamma(Z)$ point in the next Brillouin zone are almost the same as those in the first Brillouin zone. These behaviors of the experimental band dispersions are not expected if the momentum perpendicular to the surface is strictly conserved or the momentum broadening is much smaller than the size of Brillouin zone. In return, these experimental facts suggest that the above interpretation is valid for the case of URu_2Si_2. In addition, the observed bands show the periodicity matching well with the bulk Brillouin zone, which indicates that the bands are of bulk origin.

In Fig. 3(b), we show the result of the band-structure calculation[16] performed for the two parallel high-symmetry lines, ΓX (dashed lines) and ZR (solid lines). According to the calculation, the main body of the valence band consists of the Ru $4d$ and Si $3p$ states while the less dispersive bands near E_F have a strong U $5f$-Ru $4d$ hybridized character. As found in Fig. 3, the overall feature of the valence-band structure shows a qualitatively good agreement between the experiment and the calculation. For example, we find three prominent bands from E_F to 1 eV binding energy at the $\Gamma(Z)$ point in the experiment, which show a remarkable downward energy dispersion toward the $X(X)$ point. By comparing the experimental band dispersions with the calculation, the two experimental bands located closer to E_F are ascribed to the highly dispersive bands on the ZRX high-symmetry line in the calculation, while the third experimental band located at 0.9 eV at the $\Gamma(Z)$ point is assigned to a less dispersive calculated band on the ΓX high-symmetry line. In the higher binding energy region, we also find a good

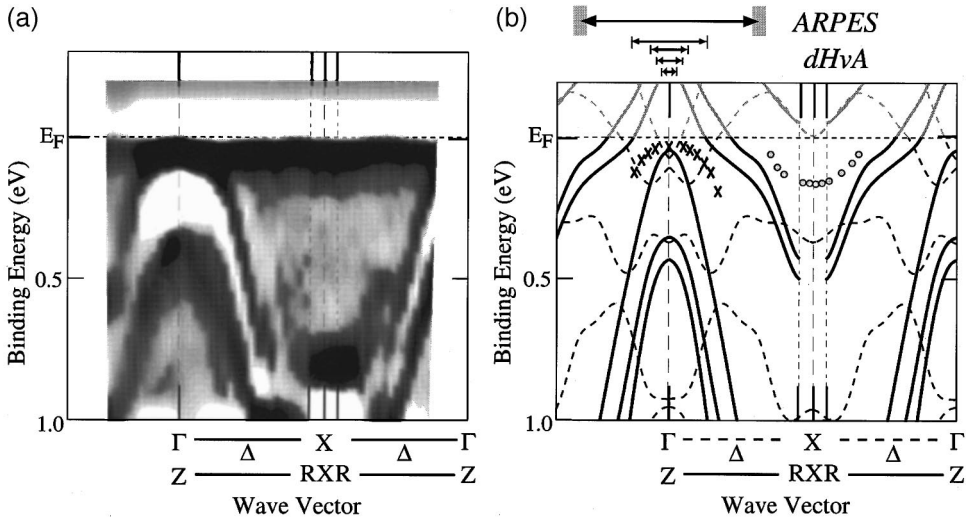

FIG. 5. Comparison of the band structure near E_F of URu_2Si_2 between (a) the ARPES experiment and (b) the band calculation (Ref. 16). The peak positions of the topmost Ru $4d$ band and the U $5f$-Ru $4d$ hybridized band in the ARPES spectra (Fig. 6) are superimposed in the calculation with crosses and open circles, respectively. The Fermi momenta observed by the present ARPES and the dHvA experiments (Refs. 7–9) are shown in a upper panel of the calculation. The uncertainty of the Fermi momentum by ARPES due to the finite angular resolution is shown by shading.

correspondence in the band dispersion between the experiment and the calculation, although the experimental bands become less clear owing to the intrinsic lifetime broadening at the higher binding energy. In spite of the good qualitative agreement, there are some quantitative differences between the experiment and the calculation. For example, we have not observed any bands in the energy range of 1.0–2.0 eV at the $\Gamma(Z)$ point as shown in Fig. 3(a), while the band structure calculation predicts several bands in the same energy and momentum region. However, it is stressed here that the band-structure calculation is a good approximation to interpret the overall valence-band structure. In the energy region near E_F, on the other hand, a strong hybridization from the U $5f$ states is expected.

In Fig. 4, we show ARPES spectra near E_F measured with a smaller energy interval. Figures 4(a) and (b) correspond to the momentum regions near the $\Gamma(Z)$ and $X(X)$ points in the Brillouin zone, respectively. The dispersive bands marked by dashed lines in Fig. 4 correspond to the highly dispersive Ru $4d$ bands, whose dispersive feature is found to be fairly well reproduced in the band calculation as shown in Fig. 3(b). In Fig. 4(a), we find that the topmost Ru $4d$ band is situated about 30 meV away from E_F even when it approaches closest to E_F at $\theta=0°$ and thus does not seem to cross E_F as predicted by the band calculation.[16] In contrast to the Ru $4d$ band which disperses to the higher binding energy from the $\Gamma(Z)$ to the $X(X)$ point, we recognize additional broad structures just below E_F, where a substantial contribution from the U $5f$ states is expected. For example, the topmost Ru $4d$ peak has a broad shoulder at the lower-binding energy side at $\theta=7°$-$8°$. Furthermore, around the $X(X)$ point in Fig. 4(b), we find a broad structure in the vicinity of E_F, where several dispersive U $5f$-Ru $4d$ hybridized bands are predicted by the band-structure calculation. Figure 5 shows comparison of the experimental band structure near E_F obtained by the ARPES with the band structure calculation.[16] The experimental band structure has been obtained with the same procedure as in Fig. 3. As described above, the overall feature of the Ru $4d$ bands near the $\Gamma(Z)$ point shows a qualitatively good agreement between the experiment and the calculation. In contrast, while the band-structure calculation predicts the comparably dispersive U $5f$-Ru $4d$ hybridized bands near E_F around the $X(X)$ point, the experimental band structure exhibits a broad vague structure from E_F to 200 meV binding energy in the same momentum region. This may suggest that the real U $5f$-Ru $4d$ hybridized bands are considerably narrow and located very close to E_F.

In order to check this point, we have measured ARPES spectra near E_F in detail with a higher energy resolution and a smaller energy interval. The result is shown in Fig. 6. We again find a very sharp peak near E_F around the $\Gamma(Z)$ point

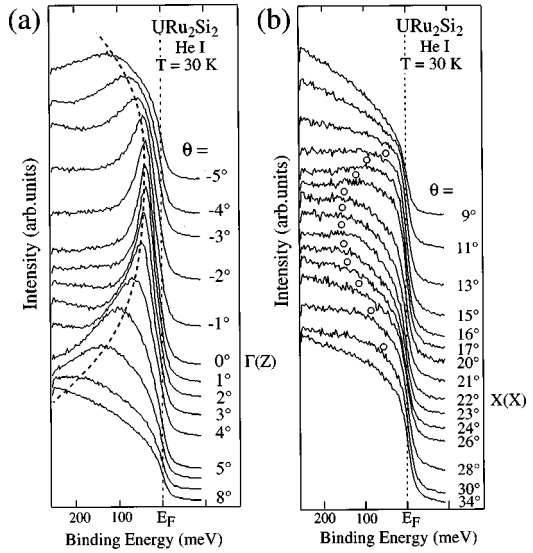

FIG. 6. ARPES spectra in the very vicinity of E_F of URu_2Si_2 measured at 30 K for the momentum regions around (a) $\Gamma(Z)$ and (b) $X(X)$ points in the Brillouin zone. The energy dispersion of the U $5f$-Ru $4d$ hybridized band around the X point is roughly traced with open circles.

[Fig. 6(a)], which has been assigned to the topmost Ru $4d$ band. It is clear in Fig. 6(a) that the E_F position is situated below the midpoint of the leading edge of ARPES spectrum near E_F even when the peak approaches closest to E_F ($\theta = 0°$).[17] This confirms that the Ru $4d$ band is totally occupied consistent with the band calculation. However, we notice that the curvature of the Ru $4d$ band near E_F around the $\Gamma(Z)$ point is considerably flatter in the experiment than in the calculation. This indicates that the band-structure calculation underestimates the hybridization strength between the Ru $4d$ and the U $5f$ states near the Z point, suggesting that the topmost ''Ru $4d$ band'' has a substantial U $5f$ character near E_F through the hybridization. In fact, as found in Fig. 5(a), the experimental second Ru $4d$ band located at 0.4 eV at the $\Gamma(Z)$ point has a sharper curvature compared with the topmost ''Ru $4d$ band,'' while the both have almost the same curvature in the calculation. This difference reinforces the above interpretation that the topmost ''Ru $4d$ band'' is flattened near E_F through the hybridization with the U $5f$ states. This suggests that the U $5f$-Ru $4d$ hybridized bands around the $X(X)$ point predicted from the band-structure calculation would be much flattened or narrowed because of the substantial U $5f$ character in the bands.

In the ARPES spectra near E_F around the $X(X)$ point in Fig. 6(b), we do not find a sharp peak as observed around the $\Gamma(Z)$ point, but instead recognize a systematic variation of the photoemission intensity near E_F with the polar angle (momentum). As found in Fig. 6(b), the intensity near E_F is considerably suppressed at $\theta = 9°$ but it suddenly increases when the polar angle is increased to $\theta = 11°$ and 13°. This suggests that a band may enter the occupied states with crossing E_F around $\theta = 11° - 13°$. With further increasing the polar angle, the photoemission intensity near E_F gradually increases as evident by the change of the curvature of the ARPES spectrum near E_F: we clearly find a small peak around 100 meV in the spectrum of $\theta = 16°$. We have confirmed the reproducibility of this small systematic variation of the ARPES intensity near E_F for several measurements with different samples. However, since the structure is weak and broad, the apparent position might be distorted by external effects such as a relatively large background. We have roughly traced the dispersion with open circles in Fig. 6(b). The band appears to have a bottom at about 150 meV binding energy around $\theta = 22°$ and reenters the unoccupied states around $\theta = 30°$. Since the momentum of $\theta = 22°$ corresponds to the $X(X)$ point in the Brillouin zone, the band shows an energy dispersion symmetric with respect to the $X(X)$ point. This may support the intrinsic nature of this small structure near E_F. We have superimposed the energy dispersion with open circles in Fig. 5(b) to compare with the band calculation. As is clear from the comparison, the observed band is assigned to the U $5f$-Ru $4d$ hybridized band(s), which has a bottom at 400–500 meV at the $X(X)$ point and forms the holelike Fermi surface(s) centered at the Z point in the calculation.[16] Here, we again find a considerable narrowing of the band dispersion near E_F in the experiment. This indicates that the strong-electron correlation between U $5f$ electrons causes the band narrowing through the renormalization effect, which is not included in the band calculation.

Finally we briefly comment on the Fermi surface of URu_2Si_2. By following the energy dispersion of the experimental band near E_F around the $X(X)$ point denoted by open circles in Fig. 5(b), we roughly estimate the E_F-crossing point, namely the Fermi momentum (k_F) as $k_F \sim 0.5\ \text{Å}^{-1}$ from the Z point. The dHvA measurements on URu_2Si_2 (Refs. 7–9) have observed at least four independent oscillations ascribable to the different Fermi surfaces. Since the dHvA frequency does not show a remarkable angular dependence, all of the four Fermi surfaces observed by the dHvA experiments are considered to be almost spherical. Under this assumption, we have estimated the Fermi momentum for each dHvA frequency as $k_F \sim 0.05$, 0.08, 0.09, and 0.2 Å^{-1} and show the size of Fermi surfaces with arrows in the upper panel of Fig. 5(b), together with the Fermi momentum observed by the present ARPES experiment for comparison. It is noted here that only one of the four dHvA branches with the largest k_F has been observed by the three independent dHvA experiments[7–9] and assigned as an electronlike Fermi surfaces at the Γ point or a small holelike Fermi surfaces centered at the Z point[9] [see Fig. 5(b)]. The largest holelike Fermi surface has not been observed by the dHvA measurements. We find in Fig. 5(b) that the largest Fermi momentum observed by the dHvA experiments is smaller than that observed by the ARPES. This suggests that the present ARPES measurement has observed the largest holelike Fermi surface centered at the Z point, which has a strong U $5f$-Ru $4d$ hybridized character. In contrast, we could not observe the Fermi surfaces with a smaller volume predicted around the $\Gamma(Z)$ point. In order to resolve these smaller Fermi surfaces and compare in detail with the dHvA result as well as with the band-structure calculation, a precise ARPES experiment with higher energy and momentum resolutions is desired.

IV. CONCLUSION

We have experimentally determined the band structure of URu_2Si_2 in the paramagnetic phase using the high-resolution angle-resolved photoemission spectroscopy, and compared the result with the band-structure calculation as well as the dHvA measurements. We found that the main body of the valence-band structure due to the Ru $4d$ and the Si $3p$ states consists of several highly dispersive bands qualitatively in good agreement with the band calculation. In the vicinity of E_F, we found a less dispersive band which crosses E_F midway between the Z and X points in the Brillouin zone. Comparing with the band calculation and the dHvA results, we found that this band is assigned to the U $5f$-Ru $4d$ hybridized band, which forms a largest holelike Fermi surface centered at the Z point. We also found that the strong-renormalization effect due to the electron correlation of U $5f$ electrons remarkably narrows the bands near E_F.

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[17] We have performed numerical simulations to check the E_F crossing of the Ru $4d$ band, using parameters of a finite energy resolution and a temperature effect (Fermi-distribution function). We have calculated ARPES spectra for several cases where the Ru $4d$ band at Γ point is (1) 30 meV below E_F, (2) 10 meV below E_F, (3) just on E_F, and (4) 10 meV above E_F. We found that in the latter two cases, (3) and (4), the E_F position is located above the midpoint of the leading edge of ARPES spectra at E_F, while in case (1), the E_F position is situated below the midpoint. Since the present experimental result is consistent with the case (1), we have concluded that the Ru $4d$ band at Γ point is located below E_F.