Electronic Structure of ThPd$_2$Al$_3$: an impact of the U 5$f$ states in the electronic structure of UPd$_2$Al$_3$

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The electronic structure of ThPd$_2$Al$_3$, which is isostructural to the heavy fermion superconductor UPd$_2$Al$_3$, was investigated by photoelectron spectroscopy. The band structure and Fermi surfaces of ThPd$_2$Al$_3$ were obtained by angle-resolved photoelectron spectroscopy (ARPES), and the results were well-explained by the band-structure calculation based on the local density approximation. The comparison between the ARPES spectra and the band-structure calculation suggests that the Fermi surface of ThPd$_2$Al$_3$ mainly consists of the Al 3$p$ and Th 6$d$ states with a minor contribution from the Pd 4$d$ states. The comparison of the band structures between ThPd$_2$Al$_3$ and UPd$_2$Al$_3$ argues that the U 5$f$ states form Fermi surfaces in UPd$_2$Al$_3$ through hybridization with the Al 3$p$ state in the Al layer, suggesting that the Fermi surface of UPd$_2$Al$_3$ has a strong three-dimensional nature.

I. INTRODUCTION

The coexistence of unconventional superconductivity and antiferromagnetic ordering with relatively large magnetic moment (0.85 $\mu_B$/U) is the most characteristic feature of the heavy fermion compound UPd$_2$Al$_3$. There are many electronic structure studies for UPd$_2$Al$_3$; however, the nature of the U 5$f$ state in UPd$_2$Al$_3$ remains contradictory. The dHvA study reported that the observed dHvA branches are mostly explained by the band structure calculation treating the U 5$f$ states as being itinerant [2]. Its electronic structure has also been studied by photoelectron spectroscopy [3–7], and the itinerant nature of the U 5$f$ states has been reported. A recent Compton scattering study has also reported that the U 5$f$ states have an itinerant character at temperatures lower than $T = 20$ K [8], which is consistent with the ARPES study [3]. Meanwhile, a resonant photoemission study on UPd$_2$Al$_3$ has also suggested the itinerant nature of the U 5$f$ state, but there exists the correlated satellite structure due to the strong electron correlation effect [9]. In contrast, the transport properties of UPd$_2$Al$_3$ are very similar to those of heavy fermion Ce-based compounds, and they have been essentially understood based on the very localized U 5$f$ picture. For example, the temperature dependence of the magnetic susceptibility has been interpreted based on the crystalline electric field scheme [10]. To understand these physical properties consistently, the dual model of the U 5$f$ states has been proposed for this compound [11]. In this scenario, the U 5$f$ states are formally divided into two subsystems: an itinerant $f^1$ component and a localized $f^2$ component. The result of the recent point contact spectroscopy study on UPd$_2$Al$_3$ was also interpreted along with this scenario [12], but the microscopic information of the U 5$f$ states is still lacking. To understand the electronic structure of UPd$_2$Al$_3$, it is essential to identify the contribution of the U 5$f$ state to gain a greater insight regarding its nature in this compound.

In the present study, we have studied the electronic structure of ThPd$_2$Al$_3$, which is the $f^0$-reference compound of UPd$_2$Al$_3$, by photoelectron spectroscopy. We compared the angle-resolved photoelectron spectroscopy (ARPES) spectra between ThPd$_2$Al$_3$ and UPd$_2$Al$_3$, and the contribution from the U 5$f$ states to the band structure of UPd$_2$Al$_3$ was clarified. Furthermore, ThPd$_2$Al$_3$ is a superconductor with $T_C = 0.2$ K [13]. It is the only case where an uranium heavy fermion superconductor has an isostructural counterpart of the thorium compound, which is also superconductor. Thus, ThPd$_2$Al$_3$ is in itself an important target material to study its electronic structure. We found that the band structure and the topology of the Fermi surface of ThPd$_2$Al$_3$ are essentially explained by the band-structure calculation based on the local density approximation.

II. EXPERIMENTAL PROCEDURE AND BAND-STRUCTURE CALCULATION

The ThPd$_2$Al$_3$ single crystal was grown by Czochralski method by pulling speed of 12 mm/h in a tetraarc furnace under Ar protective atmosphere. Final shape of single crystal was a cylinder with a diameter of $2 - 3$ mm and a length of 15 mm. The high quality of single crystal was confirmed by Laue method showing sharp spots and RRR ratio = 37 (the extrapolated value $\rho_0 = 0.7 \mu\Omega$ cm) after 2 weeks annealing at 900°C in an evacuated quartz ampule. Photoemission experiments were performed at the soft X-ray beamline BL23SU of SPring-8 [14]. The overall energy resolution in angle-integrated photoemission (AIPE) experiments at $h\nu = 800$ eV was approximately 150 meV, and that in the ARPES experiments at $h\nu = 660$ eV was approximately 90 meV. Clean sample surfaces were obtained by cleaving the samples in situ perpendicular to the c axis under ultra-high vacuum conditions. The vacuum during the course of measurements was typically < $2 \times 10^{-8}$ Pa, and the sample surfaces were stable for the entire duration of the measurements (about 2 days) since no significant changes had been observed in the ARPES spectra during these periods. The positions of the ARPES cuts in the momentum space were determined by assuming a free-electron final state with an inner potential of
V_0 = 12 eV.

In the band-structure calculations, relativistic linear augmented plane wave (RLAPW) calculations [15] within the LDA [16] were performed, treating all U 5f electrons as itinerant. In this approach, the Dirac-type Kohn-Sham equation has been formulated, and the spin-orbit interaction is exactly taken into account [17]. To compare the results of the calculation with the ARPES spectra, we have simulated the ARPES spectral functions on the basis of the band-structure calculation. In the simulation, the following effects were taken into account: (i) the broadening along the k_z direction due to the finite escape depth of the photoelectrons, (ii) the lifetime broadening of the photohole, (iii) the photoemission cross sections of orbitals, and (iv) the energy resolution and the angular resolution of the electron analyzer. The details are outlined in Ref. [18].

III. RESULTS AND DISCUSSION

Figure 1 shows the ARPES valence-band spectrum of ThPd_2Al_3 measured at \( h\nu = 800 \text{eV} \) and its comparison with that of UPd_2Al_3. According to the photoionization cross-sections, the contributions from the Pd 4d and U 5f states are dominant in this photon energy [19].

The color coding of each band represents the contribution of the U 5f states in UPd_2Al_3. This is consistent with the result of the resonant photoemission experiment for UPd_2Al_3 in which a similar similar sharp peak was observed at the Fermi energy of UPd_2Al_3 [9]. A further important point to note is that all three Pd 4d peaks in the valence-band spectrum of UPd_2Al_3 are located in higher binding energies than those in the valence-band spectrum of ThPd_2Al_3. A very similar rigid shift of the transition metal d bands has been observed in the valence-band spectra of URu_2Si_2 and ThRu_2Si_2 [20]. The amount of energy shift is approximately 200 meV, which is also very similar to the case between ThRu_2Si_2 and URu_2Si_2. This energy shift of the d band is in contrast with the case of the U 5f localized compound UPd_3 where no shift was observed in the Pd 4d bands between ThPd_3 and UPd_3 [21]. The rigid shift in the Pd 4d states between UPd_2Al_3 and ThPd_2Al_3 suggests that the U 5f states are involved in the valence-band structure of UPd_2Al_3, and they are not impurity-like contributions as expected when the U 5f states are almost localized.

Next, we discuss the band structure and Fermi surface of ThPd_2Al_3 measured by ARPES. Figure 2 summarizes the result of the ARPES study of ThPd_2Al_3. Figure 2 (a) shows the ARPES spectra of ThPd_2Al_3 along the K-M-K' high-symmetry line measured at \( h\nu = 660 \text{eV} \). The spectra consist of two different types of energy dispersions, namely very dispersive bands at \( E_F \leq E_B \leq 2.5 \text{eV} \) and less dispersive bands with enhanced intensities distributed at \( E_B \geq 2.5 \text{eV} \). Since the photoionization cross section of the Pd 4d states is more than 30 times enhanced than those of Al 3s, 3p and Th 6d states [19], the prominent dispersions at \( E_B \geq 2.5 \text{eV} \) are the contributions from the Pd 4d states. Furthermore, the dispersive bands at \( E_F \leq E_B \leq 2.5 \text{eV} \) reflect the contributions mainly from the Al and Th states.

Figure 2 (b) shows the calculated band structure and the simulation of the ARPES spectra based on the band-structure calculation. The solid lines and the density plot represent the calculated energy dispersions and the simulation, respectively. The color coding of each band represents the contribution from the Pd 4d states. The experimental band structure is well-explained by the band-structure calculation. For example, the parabolic dispersions centered at the Γ point that forms the Fermi surface are in good agreement with the calculated bands 16-18. There are also parabolic dispersions along the K-M-K high-symmetry line, which are also well-explained by the calculated band 15. At higher binding energies, the nearly flat Pd 4d bands in the experimental ARPES spectra agree with the bands 2-13 in the calculation. Although the Pd 4d bands are not clearly resolved one by one in the experimental spectra, the overall features agree very well between experimental ARPES spectra and the calculated band structure. Note that the contribution from the Pd 4d states is distributed in higher binding energies (\( E_B \geq 2.5 \text{eV} \)), and the Fermi surface of ThPd_2Al_3 mainly consists of the Al and Th states. This situation is significantly different from the case of ThRu_2Si_2 where the transition metal d band is close to the Fermi energy, and the d bands are hybridized with the U 5f states in URu_2Si_2 [20][22]. We further discuss the orbital character of the Fermi surface of ThPd_2Al_3 and UPd_2Al_3 in the later of this section.

## Figure 1.

Valence-band spectrum of ThPd_2Al_3 measured at \( h\nu = 800 \text{eV} \). Also, the bottom part of this figure provides a comparison of the valence band spectra of ThPd_2Al_3 and UPd_2Al_3 measured at \( h\nu = 800 \text{eV} \).
Figure 2(c) shows the experimental Fermi surface map of ThPd₂Al₃, which was obtained by integrating the photoemission intensity over 100 meV across $E_F$. There is a hexagonal-shaped feature with enhanced intensity centered at the $\Gamma$ point. In addition, the intensity at the $K$ point is also enhanced and these points are connected to each other and form a very complex shape. In Fig. 2(d), we illustrate the calculated Fermi surface using solid curves, and we also demonstrate the simulation the Fermi surface map based on the band-structure calculation. The three dimensional shape of the calculated Fermi surfaces of ThPd₂Al₃ is also shown in Fig. 2(e). In the band-structure calculation, the bands 16-18 form Fermi surfaces. Bands 16 and 18 form a hole-type Fermi surface around the $M$ point and an electron-type Fermi surface around the $\Gamma$ point, respectively. In contrast, band 17 forms a very complicated Fermi surface with a three-dimensional shape, but forms the electron-type Fermi surface with a star-like shape around the $\Gamma$ point. Experimental and calculated Fermi surfaces agree very well although the features are broader in the experimental map compared to the calculated map. Despite the fact that the details of the experimental Fermi surface are not very clear, the topology of the Fermi surface agrees with the result of the band-structure calculation. Accordingly, the band structure and Fermi surface of ThPd₂Al₃ were well-explained by the band-structure calculation.

Next, we performed a comparison between the ARPES spectra of ThPd₂Al₃ and UPd₂Al₃, as shown in figure 3. Figures 3(a) and (b) represent the experimental ARPES spectra of ThPd₂Al₃ and the corresponding calculated band structure, respectively. The color coding of the calculated bands corresponds to the contributions from the Th 6$d$ and the Al 3$p$ states. Note that the contribution from the Al 3$s$ is almost negligible in these binding energies. The experimental ARPES spectra are well-explained by the band-structure calculation, as discussed in Figs. 2 and thus the orbital character of each band should also agree with the result of the band-structure calculation. The calculation suggests that the Fermi surface of ThPd₂Al₃ mainly consists of the Al 3$p$ and Th 6$d$ states, but it has an enhanced Al 3$p$ character. In particular, bands 17 and 18, which form the Fermi surfaces, have a dominant contribution from the Al 3$p$ states, and the overall good agreement between the experimental data and the respective calculations suggests that the Fermi surfaces should have an enhanced contribution from the Al 3$p$ states.

Figures 3(c) and (d) show the experimental ARPES spectra of UPd₂Al₃ and the corresponding calculated band structure. The ARPES spectra of UPd₂Al₃ were taken from Ref. 7. The U 5$f$ difference spectrum obtained by resonant photoemission measurement 9 and the calculated U 5$f$ DOS are shown in the right panel of Fig. 3(c). The ARPES spectra were recorded at $h\nu = 600$ eV, and the sample temperature was 20 K. The experimental energy dispersions of ThPd₂Al₃ and UPd₂Al₃ are very similar to each other, but the intensity of the energy dispersions distributed at $E_B = E_F - 1.2$ eV is enhanced in the spectra of UPd₂Al₃. Moreover, there exist very flat features in the vicinity of $E_F$, which represent the
contributions from the U 5f states since the photoionization cross section of the U 5f states is dominant at the used photon energy. The overall structure of the experimental spectra of UPd2Al3 are also essentially explained by the band-structure calculation although the detail of each dispersion was not resolved experimentally. In particular, the very flat features at $E_F$ originate from the renormalized U 5f bands due to the strong electron correlation effect.

The comparison between the ARPES spectra of ThPd2Al3 and UPd2Al3 indicates that the U 5f states are strongly hybridized with the non-f dispersive bands in ThPd2Al3 which correspond to the calculated bands 15-18 of ThPd2Al3. These calculated bands have the enhanced contribution from the Al 3p states, suggesting that the U 5f states are strongly hybridized with the Al 3p states in UPd2Al3. The crystal structure of UPd2Al3 consists of alternating stacks of U–Pd and Al layers along the c axis, and the presence of the enhanced U 5f–Al 3p hybridization suggests that the U 5f states have a strong three-dimensional nature similar to the case of heavy fermion compound URu2Si2 [22]. As a result, the Fermi surface of UPd2Al3 should also have a strong three dimensional nature due to the enhanced U 5f–Al 3p hybridization. Previous ARPES studies of UPd2Al3 [4, 5] have shown a cylindrical Fermi surface along the Γ–A line. The Fermi surface has an enhanced contribution from the U 5f states, but the present result suggests that it also has the enhanced contribution from the Al 3p states.

Note that the U 5f states have a strong electron correlation effect, and the U 5f states in UPd2Al3 have incoherent peak distributed at approximately $E_B = 0.2 - 1$ eV [9]. Thus, the experimental bands around this binding energy should have the contribution from the incoherent component of the U 5f states. This is consistent with the dual nature of the U 5f states in UPd2Al3, as proposed theoretically, where the heavy quasi-particle band is described by the effective renormalized theory while the high-energy structure is explained by the multiplet side bands arising from the Hund’s rule [23, 24]. Experimentally, there are not non-dispersive but dispersive bands in this binding energy region, and thus the energy dispersions in $E_B = 0.2 - 1$ eV are hybridized with the incoherent U 5f states. It is theoretically proposed that the localized multiplet bands are hybridized with dispersive non-f bands [24], and the experimental spectra are consistent with the theory.

**IV. CONCLUSION**

In the present study, we investigated the electronic structure of ThPd2Al3 using photoelectron spectroscopy, and the results were compared with the band-structure calculation and the spectra of the isostructural heavy fermion superconductor UPd2Al3. The Pd 4d states in the valence-band spectrum of ThPd2Al3 were found to be shallower than those in the valence-band spectrum of UPd2Al3 by approximately 200 meV, suggesting that the U 5f states are involved in the valence-band structure of UPd2Al3, and are not impurity-like contributions. The Fermi surface and the band structure of ThPd2Al3 obtained by ARPES were well explained by the band-structure calculation. The electronic structure in the very vicinity of $E_F$ is dominated by contributions from the Al 3p and Th 6d states with minor contributions from the Pd 4d states. The comparison between the ARPES spectra of ThPd2Al3 and UPd2Al3 suggests that the electronic structure of UPd2Al3 in the very vicinity of $E_F$ is dominated by...
the enhanced U 5f–Al 3p hybridization. This indicates that the electronic structure of UPd2Al3 has a three-dimensional nature.

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