

Impact of the U 5*f* states in the electronic structure of heavy-fermion superconductor UPd₂Al₃: Electronic structure study of ThPd₂Al₃

Actinides exhibit distinct physical properties. For example, a number of unconventional superconductors have been discovered among them and have attracted much attention. These characteristic physical properties originate from strongly correlated 5*f* states, which exhibit both localized and itinerant properties. UPd₂Al₃ is a typical uranium compound that exhibits an antiferromagnetic transition at $T_N = 14$ K and undergoes a transition into a superconducting state at $T_c = 2$ K [1]. To understand the 5*f* state, it is essential to observe its band structure and Fermi surface. Angle-resolved photoemission spectroscopy (ARPES) is a powerful experimental technique that allows experimental observation of the band structure and Fermi surfaces of materials [2]. In this study, the electronic structure of ThPd₂Al₃, a non-5*f* reference compound for UPd₂Al₃, was investigated by ARPES using soft X-rays from SPRING-8 BL23SU to clarify the contribution of the U 5*f* state to UPd₂Al₃ [3]. The crystal structures of ThPd₂Al₃ and UPd₂Al₃ are shown in Fig. 1. Both compounds share a common hexagonal structure, with Th(U)–Pd and Al layers stacked alternately along the *c*-axis.

Figure 2 summarizes the band structure and Fermi surfaces of ThPd₂Al₃ obtained by ARPES: the ARPES spectrum of ThPd₂Al₃ measured along the Γ -K-M high symmetry line at $h\nu = 660$ eV and the calculated band structure with the simulation of the ARPES spectra based on the band-structure calculation are shown in Figs. 2(a) and 2(b), respectively. The prominent dispersions at $E_B \geq 2.5$ eV are the contributions from the Pd 4*d* states, while the dispersive bands at $E_F \leq E_B \leq 2.5$ eV reflect the contributions mainly from other Al and Th states. The experimental band structure was well explained by band structure

calculations. The Fermi surface map obtained by the integration of the ARPES spectra over 100 meV across E_F , the calculated Fermi surface with the simulation of the experimental Fermi surface map, and the three-dimensional shapes of the calculated Fermi surfaces are shown in Figs. 2(c,d) and 2(e), respectively. The Fermi surface of ThPd₂Al₃ was also well explained by band structure calculations.

Next, we compared the ARPES spectra of ThPd₂Al₃ and UPd₂Al₃ to reveal the contribution of the U 5*f* states to the electronic structure of UPd₂Al₃. Figures 3(a) and 3(b) show the experimental ARPES spectra of ThPd₂Al₃ and the corresponding calculated band structures, respectively. The color coding of the calculated bands represents the contributions from the Th 6*d* and Al 3*p* states. The experimental ARPES spectra are well explained by the band structure calculations, which suggest that the Fermi surface of ThPd₂Al₃ exhibits an enhanced Al 3*p* character. Figures 3(c) and 3(d) show the experimental ARPES spectra of UPd₂Al₃ recorded at $h\nu = 600$ eV and the corresponding calculated band structure. The U 5*f* difference spectrum obtained by the resonant photoemission measurements and the calculated U 5*f* DOS are shown in the right panel of Fig. 3(c). 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, which is the contribution from the U 5*f* states. The overall structure of the experimental spectra of UPd₂Al₃ was also explained by band structure calculations, although the details of each dispersion were not resolved experimentally. A comparison between the ARPES spectra of ThPd₂Al₃ and UPd₂Al₃ indicated that the U 5*f* states were strongly hybridized with the non-*f* dispersive bands, which corresponded to the calculated bands 15–18 for ThPd₂Al₃. These calculated bands have an enhanced contribution from the Al 3*p* states, suggesting that the U 5*f* states are strongly hybridized with the Al 3*p* states of UPd₂Al₃. This implies that the U 5*f* states form heavy quasi-particle bands with a strong three-dimensional character because the U–Pd and Al layers are stacked along the *c*-axis, as shown in Fig. 1. By contrast, the U 5*f* states have a strong electron correlation effect, and the U 5*f* states in UPd₂Al₃ have an incoherent peak distributed at approximately $E_B = 0.2 - 1$ eV as shown in the right panel of Fig. 3(c). Thus, the experimental bands near this binding energy can be attributed to

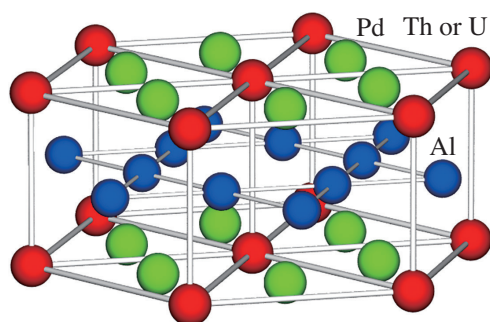


Fig. 1. Crystal structures of ThPd₂Al₃ and UPd₂Al₃.

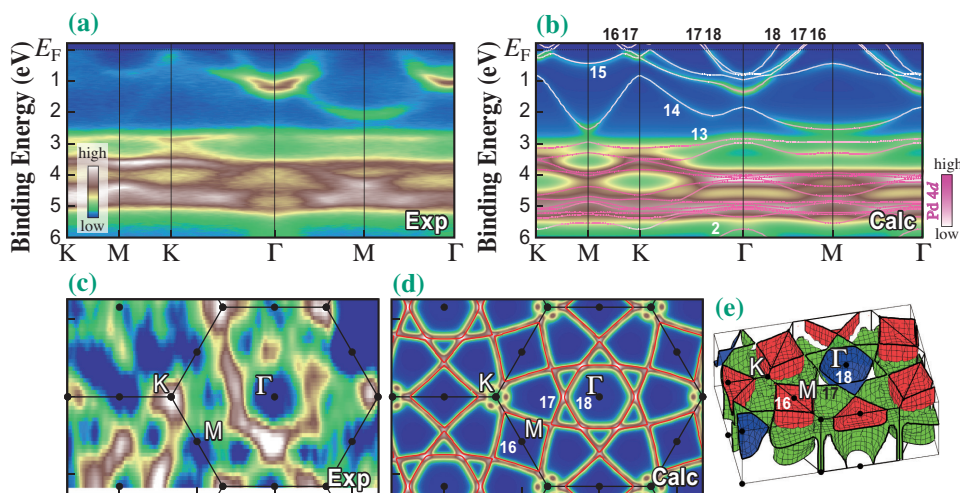


Fig. 2. Band structure and Fermi surface of ThPd₂Al₃ obtained by angle-resolved photoemission spectroscopy (ARPES).

an incoherent component. This is consistent with the dual nature of the U 5f states in UPd₂Al₃ as proposed theoretically, where superconductivity appears as a result of strong coupling between the heavy quasi-particle bands and the localized component of the U 5f states [4].

Accordingly, the Fermi surface and band structure of ThPd₂Al₃ obtained by ARPES were well explained

by band structure calculations. A comparison between the ARPES spectra of ThPd₂Al₃ and UPd₂Al₃ suggests that the electronic structure of UPd₂Al₃ in the vicinity of E_F is dominated by enhanced U 5f - Al 3p hybridization, and the U 5f states form heavy quasi-particle bands with a strong three-dimensional character, which is responsible for the superconductivity of this compound.

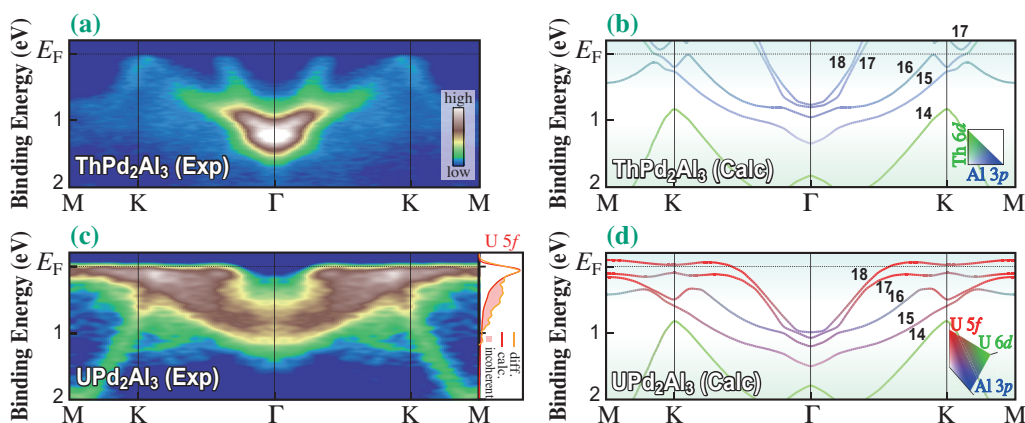


Fig. 3. Comparison between the ARPES spectra of ThPd₂Al₃ and UPd₂Al₃ and the result of the band structure calculations.

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