Itinerant to Localized Transition of *f*-Electrons in Heavy Fermion Superconductor UPd_2Al_3

Electrons in solids are conventionally classified as either band-like itinerant ones or atomic-like localized ones. However, *f* electrons in heavy Fermion (HF) compounds show both itinerant and localized behaviors depending on the temperature. Above the characteristic temperature T^*, which is typically few K to a few tens K, their magnetic properties are well described by the ionic *f*-electron model, suggesting that they behave as 'localized' *f*-electrons. On the other hand, at temperatures well below T^*, their Fermi surfaces (FSs) are generally well explained by the 'itinerant' *f*-electron model. The temperature dependence of the electronic structure of HF compounds has never been explored, and their precise behaviors and their origin have remained unknown since their discovery 30 years ago. We have experimentally studied the band structure of the HF superconductor UPd_2Al_3 at temperatures well below and above T^* by angle-resolved photoelectron spectroscopy (ARPES) to clarify how the *f*-electrons change their characteristics around T^*. ARPES is one of the most powerful experimental methods for exploring the electronic structure of solids. By performing ARPES experiments in the soft X-ray region (SX-ARPES), bulk-sensitive and 5*f*-dominant band structures and FS's can be obtained [1]. Figure 1 shows the crystal structure of UPd_2Al_3 which exhibits an antiferromagnetic transition at T_N = 14 K and undergoes a transition into the superconducting state at T_C = 2 K. Its FSs at low temperatures were investigated by de Haas-van Alphen experiments [2], and the result was well explained by the itinerant electron model [3]. On the other hand, the magnetic susceptibility follows the Curie-Weiss law above 50–70 K, suggesting the existence of localized magnetic moments at high temperatures [4]. Therefore, the T^* of UPd_2Al_3 is considered to be about 50–70 K. Photoemission experiments were performed at beamline **BL23SU**. The photon energy used was 595 eV, and the total energy resolution was 120 meV.

Figure 2(a) shows the temperature dependence of the SX-ARPES spectra of UPd_2Al_3 measured at 20 K and 100 K, which were considerably lower and higher than T^*. The position of the measurement in the Brillouin zone is also indicated. Although the essential structures of the spectra were very similar at 20 K and 100 K, noticeable differences between them were clearly observed. Between 20 K to 100 K, the peak structure located around the Fermi level (E_F) shows strong temperature dependence. Changes in the spectral functions were also observed at the higher-binding-energy sides. However, the changes is E_B>1.5 eV are mainly changes in the peak intensity, and their positions are not changed. This suggests that they are not intrinsic changes of the electronic structures. We also measured the temperature dependence of the ARPES spectra of UNi_2Al_3, whose characteristic temperature is higher than 300 K. Figure 2(b) shows the comparison of the APRES spectra of UNi_2Al_3 measured at 20 K and 100 K. It is clear that the spectra are not different at these two temperatures. Therefore, the temperature dependence of the ARPES spectra of UPd_2Al_3 at around $E_B = E_F$ - 1.5 eV originates from the changes in the electronic structure of UPd_2Al_3 below and above T^*.

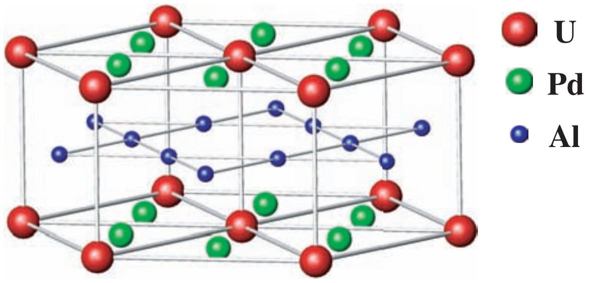

Fig. 1. Crystal structure of UPd_2Al_3.

We further analyzed the temperature dependence of these ARPES spectra of UPd_2Al_3. Figure 3 shows the second derivatives of the ARPES spectra of UPd_2Al_3 measured at 20 K and 100 K. Before taking the second derivative, the ARPES spectra were divided by the Fermi-Dirac function convoluted with the Gaussian function to reveal the behaviors of the quasi-particle bands in the vicinity of E_F. In these figures, the bright parts correspond to the peaks in the ARPES spectra. The temperature dependence of quasi-particle bands is well recognized in these figures. At 20 K, the quasi-particle bands below E_F disperse to above E_F, suggesting that they are itinerant electrons. On the other hand, at 100 K, they move toward the higher-binding-energy side, and form less dispersive bands around E_B = 0.1 eV, suggesting that they are localized electrons. These changes imply that the quasi-particle bands of U 5*f* origin at low temperatures, which form the itinerant HF states, are excluded from the FSs at high temperatures. On the other hand, the temperature-induced changes in the deeper binding energy region (E_B < 1 eV) were also

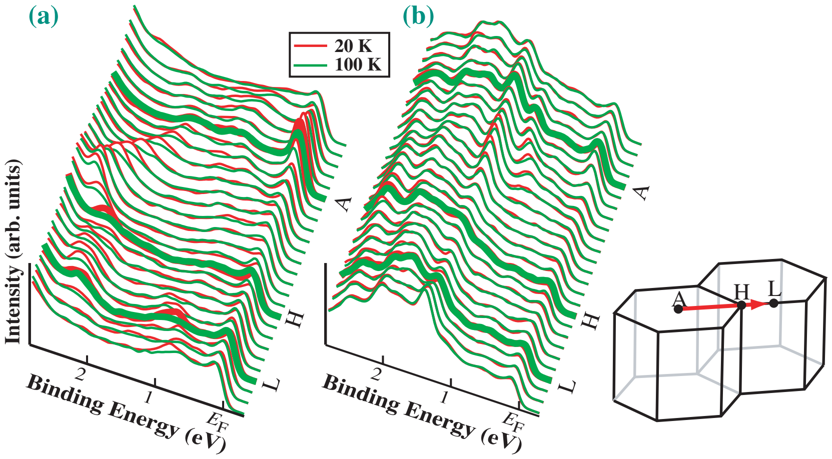

Fig. 2. Temperature dependence of ARPES spectra of **(a)** UPd_2Al_3 and **(b)** UNi_2Al_3.

discerned in these figures. For example, in the circles labeled 1 in Fig. 3, the structure located at E_B = 1 eV moves toward E_B = 1.1 eV as temperature increases from 20 K to 100 K. In addition, the complicated band structures indicated in the circles labeled 2 also have significant temperature dependences. Thus, the transition is accompanied with changes in the band structure with an energy order of 1 eV (~10^4 K), which has never been predicted previously. The result demonstrated for the first time the behaviors and the details of the itinerant and localized states and the transition between them in energy-momentum space. The present result will give new, deeper insight into not only heavy-electron superconductivity but also various types of unconventional superconductivity in solids [5].

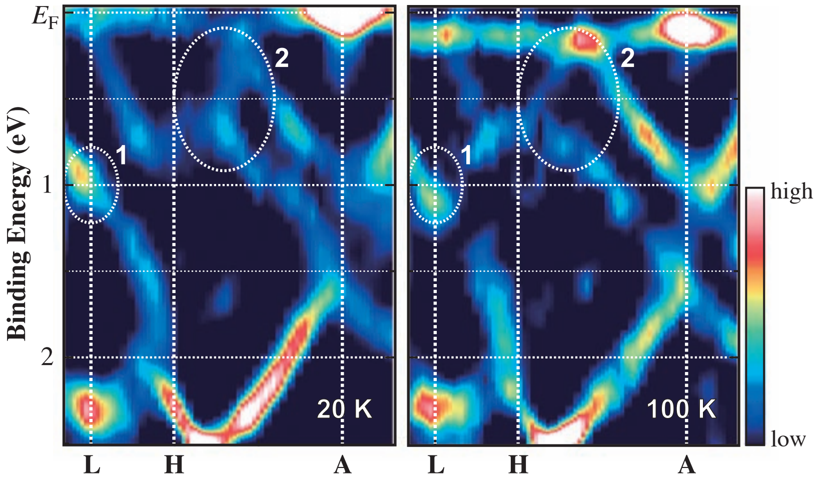

Fig. 3. Temperature dependence of band structure of UPd_2Al_3.

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