Electronic structure of URu$_2$Si$_2$ in paramagnetic phase studied by soft x-ray photoemission spectroscopy

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Abstract. We have performed soft x-ray photoemission spectroscopy study on the heavy-fermion compound URu$_2$Si$_2$. We have found that the U 5f states are distributed in the binding energy range between 0.6 eV and the Fermi level. In angle-resolved photoemission spectra, we have observed the U 5f-Ru 4d hybridized band. Our results strongly suggest that the U 5f states form itinerant energy bands in this system.

1. Introduction
The heavy-fermion compound URu$_2$Si$_2$ has attracted much attention over the past two decades. It undergoes a puzzling phase transition into the “hidden order” (HO) state at $T_0 = 17.5$ K. Although intensive experimental and theoretical studies have been performed so far [1], the true order parameter of the HO phase has not been directly detected experimentally, and none of the proposed theories can fully account for all the experimental observations. According to various experiments such as specific heat [2], thermal conductivity [3, 4] and Hall effect [5], an energy gap opens in a large portion of Fermi surface below $T_0$, leading to a large reduction of carrier density. This implies a drastic reconstruction of the electronic state at $T_0$. Therefore it is essential to elucidate the electronic structure in order to clarify the nature of the HO transition. Photoemission spectroscopy (PES) provides a direct probe of the band structure of solids. To investigate the electronic structure of URu$_2$Si$_2$, angle-resolved photoemission spectroscopy (ARPES) studies have been performed in photon energy region of $h\nu = 21$-156 eV [6, 7, 8]. According to these ARPES studies, the U 5f states are distributed in a narrow energy range within 0.2 eV below the Fermi level ($E_F$) and form less dispersive bands. The bandwidth of the observed U 5f bands is much narrower than that of the band structure calculation based on the local density approximation, suggesting an existence of a strong renormalization effect. However, these studies were performed in the lower photon energy range, and it was difficult to relate the observed band structure to the bulk properties because of the high surface sensitivity of those experiments.
Recently, PES experiments in soft x-ray region (SX-PES) have been recognized as a useful experimental technique in probing bulk electronic structures of strongly correlated materials [9, 10]. In the present study, we have performed the soft x-ray angle-integrated and angle-resolved PES experiments to elucidate the electronic structure of URu$_2$Si$_2$.

2. Experimental methods

Single crystal samples of URu$_2$Si$_2$ were prepared by the Czochralski pulling method in a tetra-arc furnace. The samples were cleaved in situ by the top post method with the surface parallel to (001) plane just before the measurements. SX-PES measurements were performed at BL23SU in SPring-8 using a Scienta SES 2002 electron energy analyzer. The base pressure was typically $4 \times 10^{-8}$ Pa. The energy resolutions were about 80 and 180 meV for $h\nu = 403$ and 760 eV, respectively. The sample temperature was kept at 20 K above $T_0$ in order to avoid a reconstruction of Brillouin zone.

3. Results and discussion

We first show the angle-integrated photoemission (AIPES) spectra. Figure 1 shows the AIPES spectra of URu$_2$Si$_2$ measured at $h\nu = 403$ and 760 eV. These photon energies were chosen to trace the cuts including the Γ point in momentum space. The solid angle determined by the angular acceptance of the electron analyzer corresponds to the two dimensional plane which covers the first and part of the neighboring Brillouin zones (BZs). Both spectra were normalized to match the tail of valence band states below 2 eV. The photoionization cross-section calculations for atomic orbitals [11] suggest that the contributions from the U 5f and the Ru 4d states are about one order of magnitude larger than those of other states such as Si 3p and U 6d in this photon energy range. The relative photoionization cross section of the U 5f states with respect to the Ru 4d states increases by a factor of about 2 by increasing photon energy $h\nu$ from 403 to 760 eV. As shown in Fig. 1, the intensity in the energy region from 0.6 eV to $E_F$ enhances by increasing photon energy $h\nu$, suggesting the existence of the U 5f states in this energy region.

To investigate the U 5f states in more detail, we have measured ARPES spectra at $h\nu = 760$ eV. This photon energy corresponds to the scan along the Γ-X-Z plane in the body-centered tetragonal BZ (Fig. 2(a)) by assuming the inner potential of photoelectron final states to be 12 eV [6]. Figure 2(b) shows the ARPES spectra of URu$_2$Si$_2$ along the Z-Y-X line. As seen
in Fig. 2(b), a strongly dispersive hole-like band with a band maximum at around $E_F$ was observed around the Z point. In addition, we have observed another dispersive band along the Y-X. These bands are considered to be contributions mainly from the Ru 4d states since these bands disperse to higher binding energies. In contrast, in the vicinity of $E_F$, we have observed a less dispersive band between the Z point and the Y point, suggesting a strong U 5f character. This band strongly hybridized with the rapidly dispersing band in the Y-X, suggesting that the U 5f states are strongly hybridized with the Ru 4d states. The existence of the f-d hybridized band is in contrast to the case of U 5f localized compound UPd$_3$ [12] where the hybridization between the U 5f states and the valence bands is weak and the U 5f states form non-dispersive bands in higher binding energy. Thus this result suggests that the U 5f states contribute to the energy band formation and have an itinerant character in this compound.

We compare the present results with previous ARPES studies. Denlinger et al. [6] and Ito et al. [8] have also revealed that the U 5f states are distributed in the vicinity of $E_F$. However, the energy distributions of the U 5f states in these studies are within 0.2 eV from $E_F$, which is more than three times narrower. In addition, Ito et al. [8] have observed a U 5f derived band in the vicinity of $E_F$ along Z-Y-X line. The energy dispersion of the reported U 5f band is quite different from that of present study and is much narrower. Since the escape depths of photoelectrons in the present experiment are much larger than those of previous studies, those differences are considered to be originated from U 5f electronic states between the surface and the bulk electronic structures.

4. Conclusion
We have studied the electronic structure of URu$_2$Si$_2$ by means of SX-PES. In the AIPES spectra, we have revealed that the U 5f states are distributed over the binding energy region between 0.6 eV and $E_F$. In the ARPES spectra, we have found that the U 5f states are strongly hybridized with the rapidly dispersing Ru 4d band, suggesting that the U 5f states contribute to the energy-band formation in this compound and have an itinerant character.
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