Probing Bulk Three-dimensional Fermi Surfaces of a Strongly Correlated Material by Soft X-ray Angle-resolved Photoemission

The topology of Fermi surfaces (FSs), the momentum distribution of the electrons with the highest energy on the occupied side (Fermi level, E_F), dominates the macroscopic properties of solids such as electric resistivity, specific heat and magnetic susceptibility. Quantum oscillation measurements based on the de Haas-van Alphen (dHvA) or the Shubnikov-de Haas effect are useful techniques for detecting bulk FSs. The dHvA measurement has so far been applied to many strongly correlated rare-earth materials [1]. However, their electron- or hole-like character and their shape cannot be experimentally revealed by these measurements alone. In addition, these techniques require low temperatures and almost defect-free single crystals. FSs may change their shapes in accord with possible phase transitions at higher temperatures of the order of a few tens of K, where the dHvA measurement is inapplicable. Angle-resolved photoemission (ARPES) is a powerful tool for simultaneously detecting electronic dispersions and FSs. Nowadays it is recognized that high-resolution high-energy photoemission spectroscopy can probe bulk electronic states. In addition, high-energy soft X-ray ARPES has an advantage that it clearly resolves the momentum perpendicular to the sample surface, k_z , owing to a long photoelectron mean free path. Therefore, the soft X-ray ARPES with controlling excitation energy $h\nu$ in addition to polar and azimuthal angles has the potential to detect three-dimensional bulk FSs at high and low temperatures. We have succeeded in probing the three-dimensional FSs of a strongly correlated Ce-based material $CeRu_2Ge_2$ in the paramagnetic phase at 20 K [2], at which it is difficult to experimentally examine the FSs by the dHvA measurement.

The soft X-ray ARPES measurements were performed at beamline **BL25SU**. A clean surface of $CeRu_2Ge_2$ was obtained by cleaving *in situ* at the measuring temperature of 20 K providing a [001]

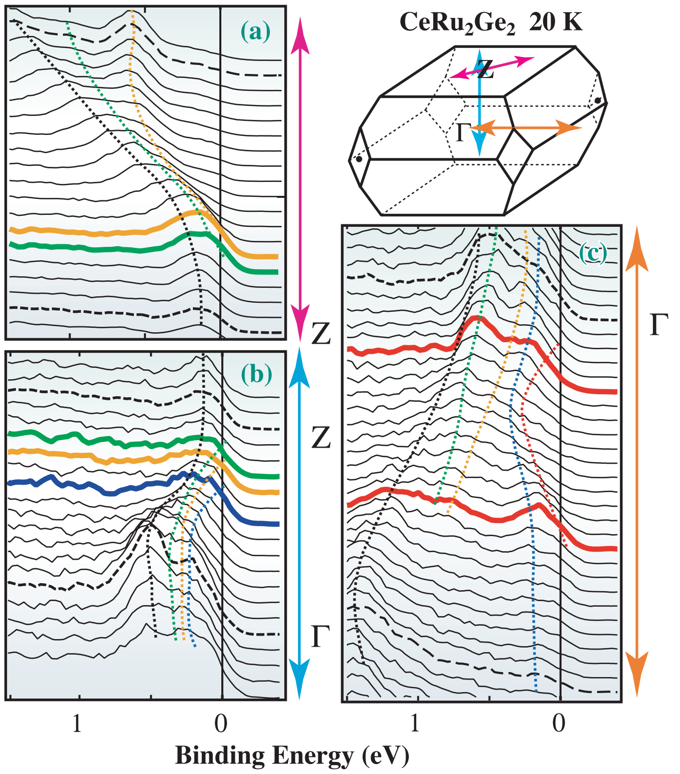

Fig. 1. High-energy soft X-ray ARPES spectra of $CeRu_2Ge_2$ measured at $h\nu$ = 735-840 eV in the paramagnetic phase at 20 K. **(a)** Spectra from point Z in the (110) direction at a fixed $h\nu$ of 755 eV. **(b)** Spectra along line from Z in the $\Gamma(k_z)$ direction obtained by changing $h\nu$. **(c)** Spectra from point Γ in the (100) direction at a fixed $h\nu$ of 820 eV. The spectra with solid color bold lines indicate those on the Fermi surfaces. The dotted lines represent the electronic dispersions as a guide for the eyes. The Brillouin zone for $CeRu_2Ge_2$ is also shown.

plane. A Gammadata-Scienta SES200 analyzer was used, which covers more than a whole Brillouin zone along the direction of the analyzer slit. The energy resolution was set to 200 meV for FS mappings. The angular resolutions were ±0.1° and ±0.15° in the perpendicular and parallel directions to the analyzer slit, respectively. These values correspond to the momentum resolutions of 0.024 Å^{-1} and 0.035Å^{-1} at $h\nu$ = 700 eV.

Figure 1 shows the high-energy soft X-ray ARPES spectra of CeRu$_2$Ge$_2$ in the paramagnetic phase. The energy positions of many peaks and shoulders in the spectra are changed depending on not only the detection angle (Figs. 1(a) and 1(c)) but also the excitation energy (Fig. 1(b)). Namely, these structures disperse three-dimensionally in the momentum space, which is an indication of the dispersions originated from the bulk electronic. From the ARPES measurements, we obtained intensity maps in the vicinity of E$_F$, which are equivalent to the cross sections of the FSs, for CeRu$_2$Ge$_2$ at 20 K along the [110] and [001] planes, as shown in Fig. 2. From these cross sections, small and large ellipsoidal Fermi surfaces centered at Z, and small cylinder-like Fermi surfaces centered at X, are expected.

Based on these results, we can obtain the qualitative shapes of the three-dimensional Fermi surfaces for CeRu$_2$Ge$_2$ in the paramagnetic phase, as shown in Fig. 3. Although the ellipsoidal Fermi surfaces are similar to those in the magnetic phase below T_c ~ 8 K [3] and those in a theoretical result based on a localized 4f model [4], it was found that the cylindrical Fermi surfaces are qualitatively different from those in the magnetic phase and in those the theoretical result.

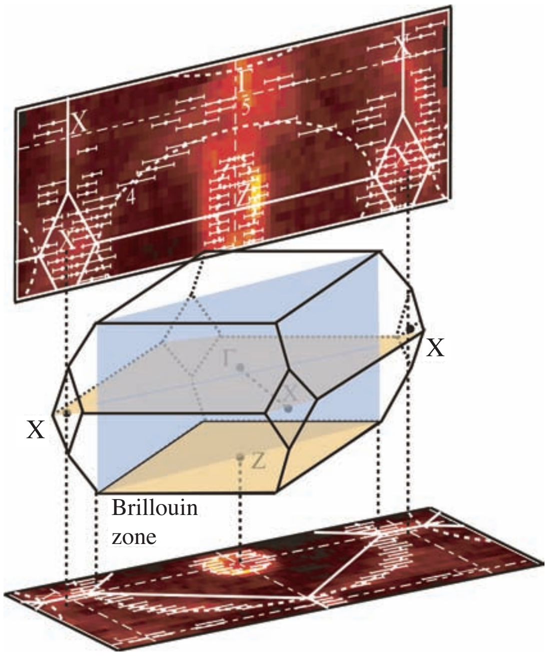

Fig. 2. ARPES intensity maps at E$_F$ ± 100 meV and estimated Fermi wave vectors corresponding to the cross sections of the Fermi surfaces of CeRu$_2$Ge$_2$. The upper panel shows the cross-sectional Fermi surfaces cut by the [110] plane in the momentum space. The middle panel represents the Brillouin zone. The lower panel shows the cross sections of the same Fermi surfaces, but cut by the [001] plane including point Z.

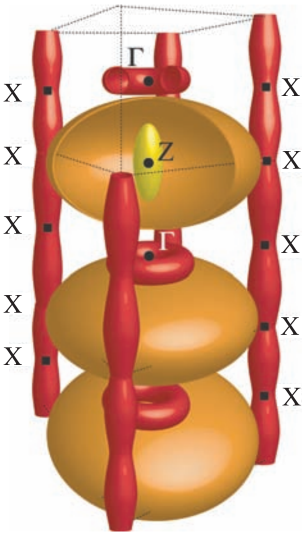

Fig. 3. Schematically drawn and qualitative image of the three-dimensional Fermi surfaces of CeRu$_2$Ge$_2$ based on our ARPES results.

In summary, we have directly examined the FS topology of CeRu$_2$Ge$_2$ at a "high" temperature in the paramagnetic phase. We are convinced that the use of high-energy soft X-ray ARPES demonstrated here will become another complementary and powerful technique for probing the bulk FSs in solids in near future.

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