Change of electronic structure from itinerant to localized state in heavy-fermion Ce compound: A Compton scattering study

The heavy fermion system shows a variety of interesting properties since a strongly correlated *f* electron changes its aspects depending on conditions such as temperature, magnetic field, pressure, and chemical substitution. Studies of the *f* electronic state have so far ranged over many heavy fermion materials. In particular, $CeRu_2Si_2$ is still an intriguing substance as a typical heavy fermion compound. The Ce-4*f* electron is considered to hybridize with a conduction electron below the Kondo temperature, T_K, with the result that it acquires the itinerant character as a heavy quasiparticle. In the meantime, above T_K, the Ce-4*f* electron is thought to localize in the paramagnetic phase. So far, the electronic structure of $CeRu_2Si_2$ has been frequently investigated by several experimental techniques. For instance, the de Haas–van Alphen effect measurement has been used to observe the Fermi surface (FS) topology of the itinerant state [1]; however, since this method requires a very low temperature, it is difficult to apply this method to the localized state at high temperature. The angle-resolved photoelectron spectroscopy (ARPES) experiment is also an effective technique for investigating the electronic structure. Recent ARPES experiments revealed the detailed band structure and FS of $CeRu_2Si_2$. In those studies, however, measurements were performed in the vicinity of T_K and the temperature dependence was not measured, hence the electronic structure of $CeRu_2Si_2$ was discussed in comparison with that of related compounds such as $LaRu_2Si_2$ and $CeRu_2(Si_{1-x}Ge_x)_2$ [2-4]. In this study, we employed high resolution Compton profile (HRCP) measurement as another approach for investigating the change in the Ce-4*f* electronic state in $CeRu_2Si_2$ [5].

The sample measured was a single crystal of $CeRu_2Si_2$ grown by the Czokralski pulling method in a triarc furnace. The T_K of the sample was evaluated to be 20 K from the result of magnetic susceptibility measurement. The HRCP measurements were made at beamline **BL08W**. The energy of incident X-ray was 115 keV, and the scattered X-ray was energy resolved with a Ge(620) analyzer crystal and detected by a position sensitive detector. The momentum resolution was 0.11 atomic units (a.u.). For the purpose of reconstructing a momentum density (MD), five directional HRCPs were measured at even intervals between the [100] and [110] crystal axes at 5 K and room temperature (RT). The two-dimensional MD projected onto the (001) plane in momentum space was reconstructed from the HRCPs by the direct Fourier transform method. Then the LCW analysis, which folds the MD back into the projected first Brillouin zone (BZ), was applied to the 2D MD to obtain the 2D electron occupation number density (EOND), where the EOND is the number density of electrons (holes) at each wave number, *k*, in the BZ. The EOND thus obtained has the following advantage in investigating the change in the *f*-electronic state in this system. The MDs of core electrons and fully occupied bands, in principle, make no contribution to the structure of the EOND. Therefore, the structure found in the EOND reflects the electronic state of the itinerant electron, that is, we have a chance of detecting the variation of *f* electron from the itinerant to the localized states.

Figures 1(a) and 1(b), respectively, show 2D EONDs of itinerant electrons at 5 K and RT. In this

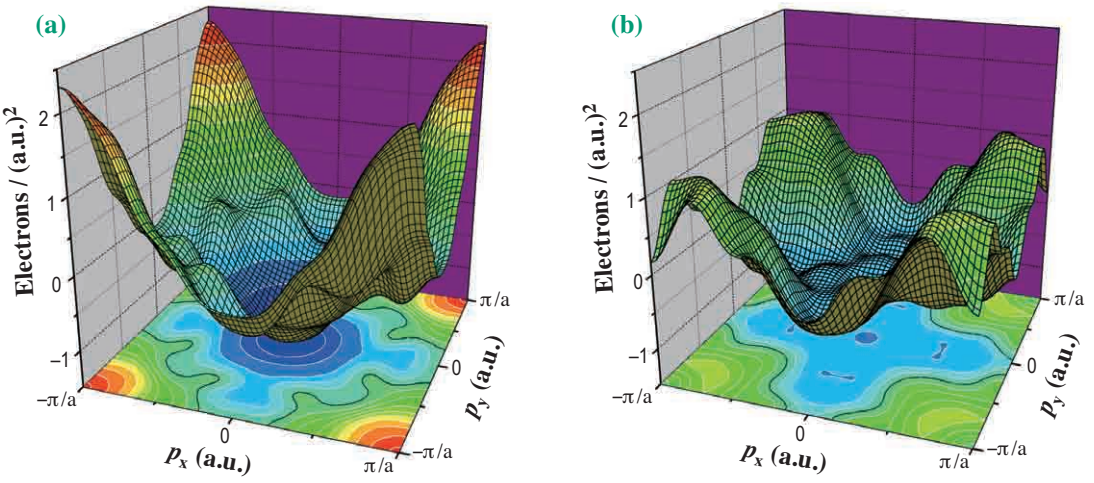

Fig. 1. Experimental 2D EONDs obtained at **(a)** 5K and **(b)** RT. [5]

regard, however, in order to emphasize the structure arising from itinerant electrons, the mean 2D EOND, which was derived from the average of the directional profiles, was subtracted from the original 2D EOND. Here, note that in the case of the 3D EOND, the boundaries between occupied and unoccupied areas correspond to the FS. In the 2D case, the densities perpendicular to a view plane are projected onto the view plane. Even so, the change in the electronic state involved with f electrons can be detected in the 2D EOND. In fact, the most striking change is observed around the corner of the projected BZ; that is, a columnar structure observed at 5 K is reduced in size at RT. To identify the cause of the change, we have also carried out a band calculation on this system by the local-density approximation based on the full-potential linearized augmented plane wave method, where the Ce-4*f* electron is treated as an itinerant one. Figure 2 shows 2D EONDs of (a) the 14th, (b) the 15th, and (c) the sum total of relevant bands derived from the calculation result. These bands across the Fermi energy level and hence are involved in the formation of the FS including f electron contribution. The depressed area in Fig. 2(a) reflects the contribution of holes in the 14th band to the FS, and the pillar section at the corner in Fig. 2(b) is the contribution of electrons in the 15th band. The theoretical 2D EOND in Fig. 2(c) well reproduces the general appearance of the experimental one in Fig. 1(a), and the pillar section in Fig. 2(b) well describes the difference in density between Figs. 1(a) and 1(b). In the present case, the densities at the X, W, and P symmetric points in the BZ are projected on the corner of the 2D EOND. The band calculation reveals that these symmetric points are highly occupied by the Ce-*f* (ℓ=3) component in the 15th band. Therefore, the columnar structure in Fig. 1(a) is attributed to the Ce-4*f* electron in the 15th band, and the shrinkage of the columnar structure in Fig. 1(b) clearly specifies the localization of the Ce-4*f* electron.

As described above, in the present study, the Ce-4*f* electron, which changes its nature from the itinerant to the localized states, was well detected in $CeRu_2Si_2$. The Compton experiment is feasible without constraint on the experimental conditions such as temperature, magnetic field, pressure, and chemical substitution. Therefore, this technique will also be effective in systematically investigating the change in the electronic structure along with the metamagnetic transition in $CeRu_2Si_2$, the hidden order in URu_2Si_2, and other interesting phenomena of the heavy fermion system.

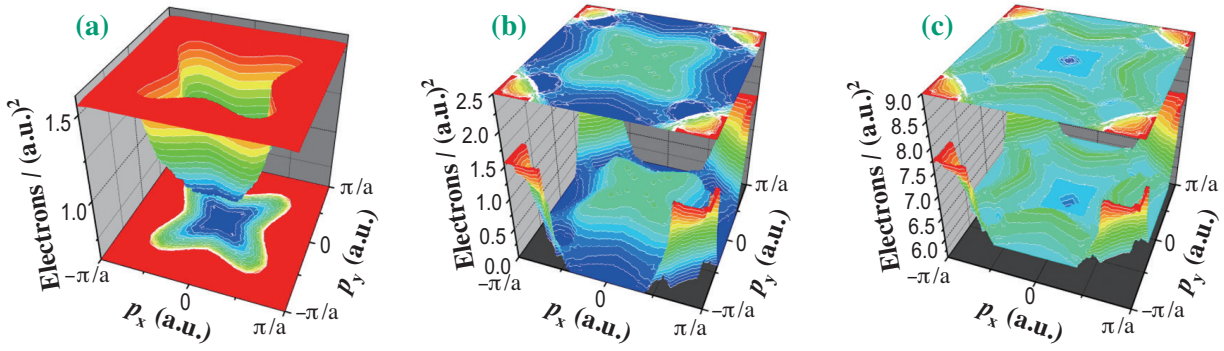

Fig. 2. Theoretical 2D EONDs of **(a)** the 14th, **(b)** the 15th, and **(c)** the sum total of relevant bands. [5]

Akihisa Koizumi

Graduate School of Material Science,
University of Hyogo

E-mail: akihisa@sci.u-hyogo.ac.jp

References

[1] M. Sugi *et al.*: Phys. Rev. Lett. **101** (2008) 056401.
[2] J.D. Denlinger *et al.*: J. Electron Spectrosc. Relat. Phenom. **117-118** (2001) 347.
[3] M. Yano *et al.*: Phys. Rev. B **77** (2008) 035118.
[4] T. Okane *et al.*: Phys. Rev. Lett. **102** (2009) 216401.
[5] *A. Koizumi, G. Motoyama, Y. Kubo, T. Tanaka, M. Itou and Y. Sakurai: Phys. Rev. Lett.* ***106*** *(2011) 136401.*