Fermi Surface Variation Near Quantum Critical Point of $CeRu_2(Si_{1-x}Ge_x)_2$ Studied by Resonant ARPES

$CeRu_2Si_2$ is a representative heavy-fermion (HF) compound with a paramagnetic ground state. The large specific heat coefficient γ~350 mJ/mol K^2 indicates the itinerancy of the Ce 4*f* electrons in the ground state, and the Kondo crossover temperature T_0 is estimated to be ~20-25 K. A magnetic ground state appears upon substitution of Ge atoms for Si atoms, i.e., $CeRu_2(Si_{1-x}Ge_x)_2$, corresponding to the application of a negative chemical pressure: The system is antiferromagnetic for x = 0.07-0.57 and ferromagnetic for x = 0.57-1.0 [1]. The boundary of the emergence of the antiferromagnetic ground state, namely, the critical composition x_c = 0.07, at a very low temperature is considered to be the quantum critical point (QCP), where the Ce 4*f* electrons may be transformed from itinerant electrons to localized electrons, possibly accompanied by variations in the shape and volume of Fermi surfaces (FSs). The de Haas van Alphen (dHvA) experiment indicated that the 4*f*-itinerant FSs are observed in paramagnetic $CeRu_2Si_2$, while the FSs of ferromagnetic $CeRu_2Ge_2$ are almost the same as those of $LaRu_2Ge_2$, indicating that the 4*f* electrons are localized in $CeRu_2Ge_2$ [2,3]. The important issue is in the point where the variation from the 4*f*-itinerant FSs to the 4*f*-localized FSs occurs upon the substitution of Ge atoms for Si atoms, and one of the most possible candidates is the point near QCP, i.e., around x_c. Therefore, in order to verify the existence of the FS variation across x_c, we have investigated the Fermi surfaces $CeRu_2Si_2$ and $CeRu_2(Si_{0.82}Ge_{0.18})_2$ in paramagnetic state at around T_0 by soft X-ray angle-resolved photoelectron spectroscopy (ARPES) [4]. In order to enhance the photoemission signals of the Ce 4*f* electrons, we have performed an ARPES experiment in the Ce $3d \rightarrow 4f$ resonance energy region. ARPES was also performed for $LaRu_2Si_2$ as a reference material where the 4*f* electrons do not participate in the FS formation. The experiments were performed at beamline **BL23SU**.

Figures 1(b) and 1(c), 1(d) and 1(e), and 1(f) and 1(g) show ARPES spectra of the valence bands of $LaRu_2Si_2$, $CeRu_2Si_2$, and $CeRu_2(Si_{0.82}Ge_{0.18})_2$, respectively, along the *X*-*Z* and Γ-*X* directions of the Brillouin zone (BZ) illustrated in Fig. 1(a). The angle-scanning measurements were performed at photon energies $h\nu$ corresponding to the Γ-*X*-*Z* plane, i.e., $h\nu$ = 765, 860, and 855 eV (*off-resonance* hereafter) for $LaRu_2Si_2$, $CeRu_2Si_2$, and $CeRu_2(Si_{0.82}Ge_{0.18})_2$, respectively. As indicated by the red broken lines, the observed dispersions of energy bands are similar between $CeRu_2Si_2$ and $CeRu_2(Si_{0.82}Ge_{0.18})_2$, while the energy dispersions of $LaRu_2Si_2$ are remarkably different from those of $CeRu_2Si_2$.

Figures 2(a)-2(c) show off-resonance FS images of $LaRu_2Si_2$, $CeRu_2Si_2$, and $CeRu_2(Si_{0.82}Ge_{0.18})_2$, respectively, represented by the intensities of the ARPES spectra integrated near Fermi energy (E_F)

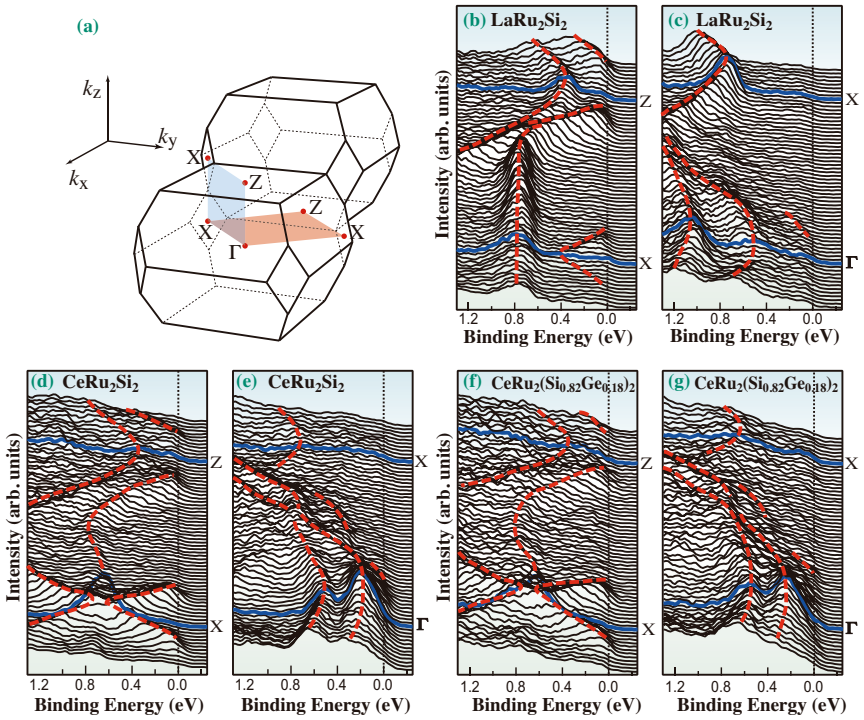

Fig. 1. **(a)** Brillouin zone for $CeRu_2(Si_{1-x}Ge_x)_2$. Off-resonance ARPES spectra of $LaRu_2Si_2$ along **(b)** *X*-*Z* and **(c)** Γ-*X* directions, those of $CeRu_2Si_2$ along **(d)** *X*-*Z* and **(e)** Γ-*X* directions, and those of $CeRu_2(Si_{0.82}Ge_{0.18})_2$ along **(f)** *X*-*Z* and **(g)** Γ-*X* directions.

as a function of momenta (k_x, k_y). For comparison, the calculated FSs of $LaRu_2Si_2$ and $CeRu_2Si_2$ in the k_x–k_y plane [5] are shown in Figs. 2(d) and 2(e), respectively. The numbers 1-4 and 5 designate the hole FSs and the electron FS, respectively. Figures 2(f) and 2(g) show the calculated three-dimensional (3D) images of the large hole FS of band 4 of $CeRu_2Si_2$ and $LaRu_2Si_2$, respectively, indicating a significant volume variation due to the participation of the Ce 4*f* electrons in the FS formation [5]. A large FS surrounding the *Z* point was experimentally detected in every compound. While the experimental large FS of $LaRu_2Si_2$ has a square shape and agrees well with the calculated hole FS of band 4, the experimental large FS of $CeRu_2Si_2$ has a circular shape and agrees well with the calculated electron FS of band 5. The large FS of $CeRu_2(Si_{0.82}Ge_{0.18})_2$ is almost identical to that of $CeRu_2Si_2$. A residual drawback in this assignment is that the hole FS of band 4 in Fig. 2(e), which is considered to be the heaviest FS branch including the largest contribution of the Ce 4*f* electrons [5], is not observed in both $CeRu_2Si_2$ and $CeRu_2(Si_{0.82}Ge_{0.18})_2$.

Therefore, angle-scanning measurements were performed at the Ce 3*d*→4*f* resonant energy of $h\nu$ = 881 eV (*on-resonance*) for $CeRu_2Si_2$ and $CeRu_2(Si_{0.82}Ge_{0.18})_2$ to enhance the signals of the 4*f*

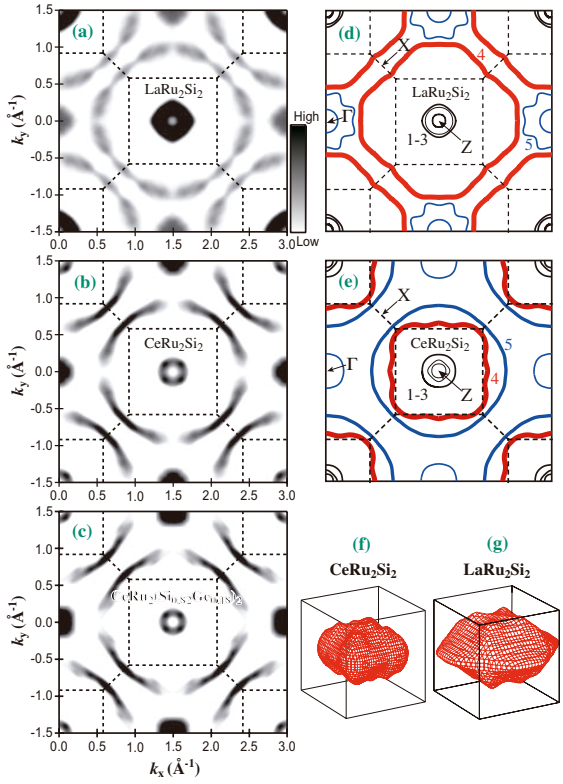

Fig. 2. Off-resonance FS images of $LaRu_2Si_2$ **(a)**, $CeRu_2Si_2$ **(b)**, and $CeRu_2(Si_{0.82}Ge_{0.18})_2$ **(c)** in the k_x-k_y plane, compared with calculated FS images of $LaRu_2Si_2$ **(d)** and $CeRu_2Si_2$ **(e)** [5]. BZ is illustrated by broken lines. Calculated 3D images of hole FS of band 4 of $CeRu_2Si_2$ **(f)** and $LaRu_2Si_2$ **(g)** around Z point (body center) [5].

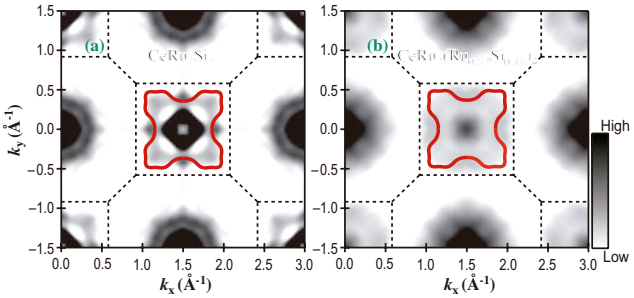

Fig. 3. On-resonance FS images in k_x-k_y planes of **(a)** $CeRu_2Si_2$ and **(b)** $CeRu_2(Si_{0.82}Ge_{0.18})_2$, compared with calculated hole FS of band 4 at $k_z = 0.3\ \pi/c$ away from *Γ*-*Z*-*X* plane, represented by red lines [5].

electrons. The resonance energy corresponds to k_z = 0.29 π/c and 0.37 π/c for $CeRu_2Si_2$ and $CeRu_2(Si_{0.82}Ge_{0.18})_2$, respectively. Figures 3(a) and 3(b) show on-resonance FS images of $CeRu_2Si_2$ and $CeRu_2(Si_{0.82}Ge_{0.18})_2$, respectively. Surprisingly, the large circular FS surrounding the *Z* point in the off-resonance images becomes invisible in the on-resonance images. On the other hand, another FS appears only inside the square BZ boundary surrounding the Z point in both $CeRu_2Si_2$ and $CeRu_2(Si_{0.82}Ge_{0.18})_2$. The size and shape of this FS are comparable to those of the calculated hole FS of band 4 at k_z = 0.3 π/c away from the *Γ*-*Z*-*X* plane, shown by red lines in Figs. 3(a) and 3(b), and thus the observed FS is assigned to be the FS of band 4, which includes the largest contribution of the Ce 4*f* electrons. Thus, the assignment of the FSs in Figs. 2 and 3 is totally consistent.

The observation of the FS including a strong 4*f* contribution in both $CeRu_2Si_2$ and $CeRu_2(Si_{0.82}Ge_{0.18})_2$ indicates that the 4*f*-delocalized regime is extended beyond the critical composition x_c. The absence of the FS change near x_c is strongly implied in the paramagnetic state below T_0, and it can be considered that the spin-density-wave model is suitable for the quantum criticality of $CeRu_2(Si_{1-x}Ge_x)_2$.

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