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Change of Electronic Structure Associated with Hidden Order Transition in URu_2Si_2 Studied by Compton Scattering Experiment

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Directional Compton profiles have been measured on the (100) plane in URu_2Si_2 single crystal at temperatures above and below the hidden-order (HO) transition. Two-dimensional electron momentum density (2D-EMD) was reconstructed from the directional profiles through the direct Fourier-transform method. The variation of 2D-EMD with temperature clearly reflects the change in electronic structure associated with the HO transition. The change is particularly noticeable around the Γ and/or Z symmetry points. This is also verified through similar experiment on the (001) plane and reconstruction analysis.

KEYWORDS: URu_2Si_2, hidden-order transition, electron momentum density, Compton scattering, two-dimensional reconstruction

1. Introduction

The heavy fermion compound URu_2Si_2 has provided an absorbing subject of study in the field of strongly correlated electron system for more than a quarter-century. The compound undergoes a second order phase transition at T_{HO}=17.5 K, while its order parameter has not been identified clearly [1-3]. This enigmatic phenomenon is therefore called the hidden-order (HO) transition. In addition, at around 70 K, the compound shows a crossover phenomenon between itinerant and localized states of the U 5 *f* electrons. Furthermore, an unconventional superconducting state appears at 1.5 K. The evolution of electronic state in this system is considered to be involved with the plurality and the itinerancy-localization duality of 5 *f* electrons.

Particularly the electronic structure of the HO state has been investigated by proven methods such as Shubnikov–de Haas (SdH) measurement [4-7] and angle-resolved photoemission spectroscopy (ARPES) [8-15]. Although these are very effective and highly-regarded techniques, they have some disadvantages. For example, SdH measurement is of advantage to probe Fermi surface (FS) structure with the use of small

sample. It is however feasible only at very low temperatures, and is not suitable for use in high-temperature region above T_{HO}. ARPES is also a well-known method to study FS and band structures, while it is sensitive to a cleaved surface of the sample, and has a possible problem regarding the relaxation energy involved in the excitation of occupied and empty states [16,17]. Therefore, some sort of bulk-sensitive measurement is required for the study of electronic structure to cover for each other's disadvantage. Alternatively, Compton scattering experiment is given as such a method.

In fact, we have employed high-resolution Compton profile (HRCP) measurement in previous studies on URu_2Si_2 [18,19]. We investigated the electronic structures varied with temperature in association with the HO transition and the crossover phenomena from the localized to the itinerant states. In those studies, directional Compton profiles were measured in the (001) crystal plane, and two-dimensional electron momentum density (2D-EMD) and electron occupation number density (2D-EOND) were derived from the directional profiles through several analyses. As a matter of course, the electronic structures obtained in this way were projected on the view plane. In this study, we have performed HRCP measurements on the (100) crystal plane for further insight into the electronic structure related to the HO transition. In conjunction with the previous results measured on the (001) plane, we have examined the change of electronic structure in three dimensions from the viewpoint of EMD.

2. Experimental

In cases where you make Compton scattering measurement with high-energy X-ray, the impulse approximation is applicable in the analysis, and therefore, you can investigate bulk electronic structures in the initial state [20]. Concretely, the Compton profile $J(p_z)$ is described by the following formula.

$$J(p_z) = \iint \rho(p_x, p_y, p_z)\, dp_x dp_y \quad , \qquad (1)$$

where p_z means an electron momentum component along a scattering vector, and $\rho(p_x, p_y, p_z)$ is electron momentum density, that is, the square of the absolute value of the wave function in momentum representation. As shown in eq. (1), the Compton profile is the one-dimensional projection of EMD on the scattering vector. Then, in order to reconstruct the original EMD, you need to measure a number of Compton profiles in different directions of single crystal sample.

A single crystal of URu_2Si_2 used in this study was prepared by the Czochralski pulling method. In conducting a synchrotron X-ray experiment, the sample was mounted in a hermetically-sealed container. HRCP measurements were performed on the beamline BL08W at SPring-8. The energy of incident X-ray was set to 114.56 keV, which was slightly lower energy than the energy of U-K absorption edge, to circumvent the effect of fluorescent X-rays on the observed scattering intensities. The Compton scattered X-ray was energy resolved by a Si (620) bent crystal analyzer and detected by a position sensitive detector. Nine (Five) directional HRCPs were measured at even intervals between the [100] and [001] ([100] and [110]) crystal axes at 14K ($< T_{HO}$) and 20K ($> T_{HO}$), with a view to reconstructing 2D-EMD. The momentum resolution was

calculated to be 0.11 atomic unit on the basis of the energy resolution of monochromator and analyzer, and angle resolution of scattering angle.

3. Results and Discussion

Figures 1(a) and 1(b) respectively show 2D-EMDs at 14 K and 20 K reconstructed from the directional HRCPs measured on the (100) plane. Figures 1(c) and 1(d) show the same, but the HRCPs were measured on the (001) plane.

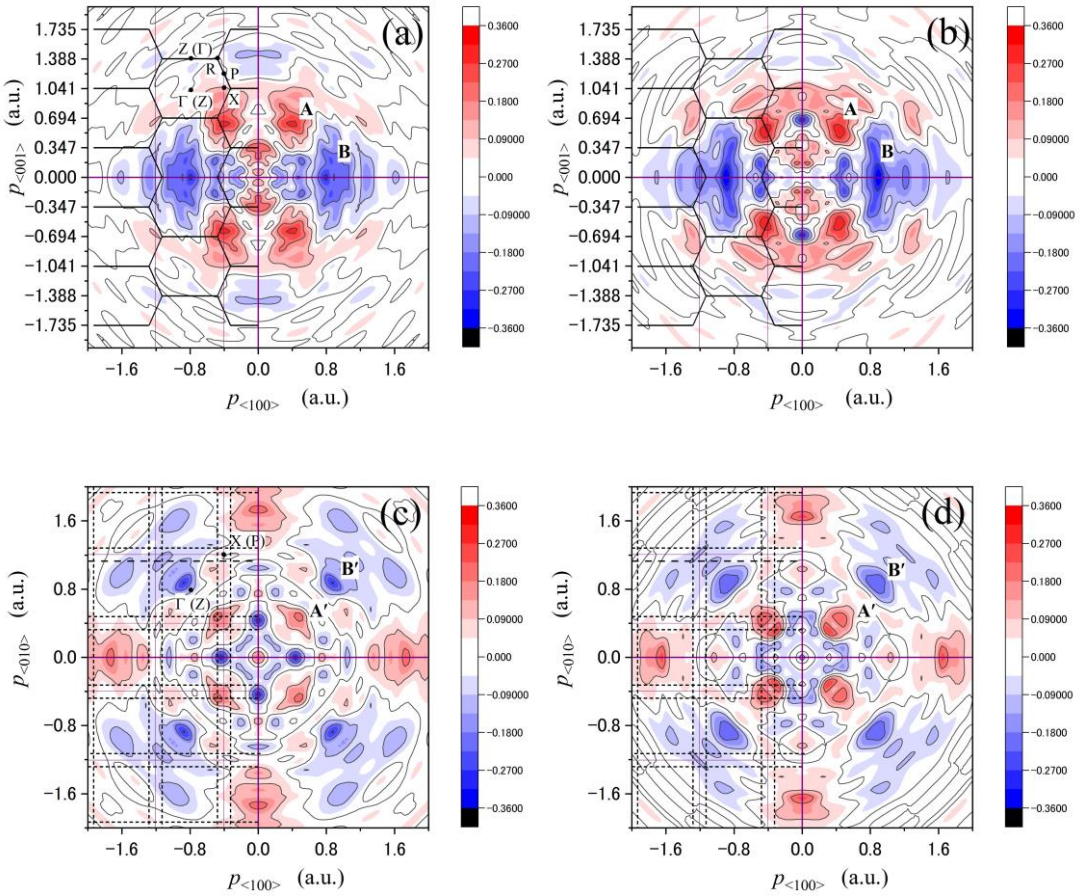

Fig. 1. 2D-EMDs measured on the (100) plane at 14K (a) and 20K (b), and likewise 2D-EMDs measured on the (001) plane at 14K (c) and 20K (d). The ordinate and abscissa axes show electron momentum in atomic unit (a.u.) along the crystal direction indicated by a subscript. These figures are close-up images around the origin in the large momentum space. Typical symmetry points are marked with a dot in Fig. 1(a) and 1(c) for reference.

Here, in order to derive the 2D-EMD, the direct Fourier-transform method was applied to the set of directional HRCPs after correcting scattering cross section, background, multiple scattering and x-ray absorption in the sample [21]. In the analysis, each HRCP is Fourier transformed to a one-dimensional $B(r)$ function which is equivalent to a reciprocal form factor along the measurement direction. Then, the 2D-$B(r)$ function is obtained by interpolating between the $B(r)$ functions in a grid pattern. The inverse Fourier transform of the 2D-B(r) derives the 2D-EMD projected on the view plane. The 2D-EMDs in Fig. 1 were expanded according to the crystal symmetry. In addition, an average 2D-EMD, which was obtained by using the average profile of directional HRCPs in the same way, was subtracted from the original 2D-EMD to emphasize the details of electronic structure. Additionally, the projected Brillouin zones (BZs) are described in Fig. 1 on the assumption that the sample has a body-centered-tetragonal structure. The densities at the Γ and Z points are projected onto the center point in every BZ. Although the densities at the X and P points are piled up around the corners of the BZs in Fig. 1(c) and 1(d), those points can be distinguished in Fig. 1(a) and 1(b). Overall, the electronic structures seem to appear notably around these symmetry points. For instance, the positive and negative structures labeled A and B are, respectively, found around the X-P-R and Z (Γ) points in Fig. 1(a) and 1(b). And if you look at Fig 1(c) and 1(d), the positive and negative structures, which are labeled A' and B', appear also around similar symmetry points in the momentum area of $p_{<100>} \approx \pm 0.4$ and ± 0.8 a.u., respectively. This means that an identical structure in the momentum space is projected onto different view planes. Figure 2, which is the difference in 2D-EMD between 14 K and 20 K, highlights the change in electronic structure associated with the HO transition. This means that the energy band structure changes at T_{HO}. As mentioned above, the U 5 *f* electrons are considered to move gradually from localize to itinerant states below the crossover temperature. The present results suggest that the U 5 *f* electrons go on to more itinerant state in the HO phase. As reported in ref.18, the energy band structure, which was calculated based on the itinerant model, indicates that the electron FS is formed

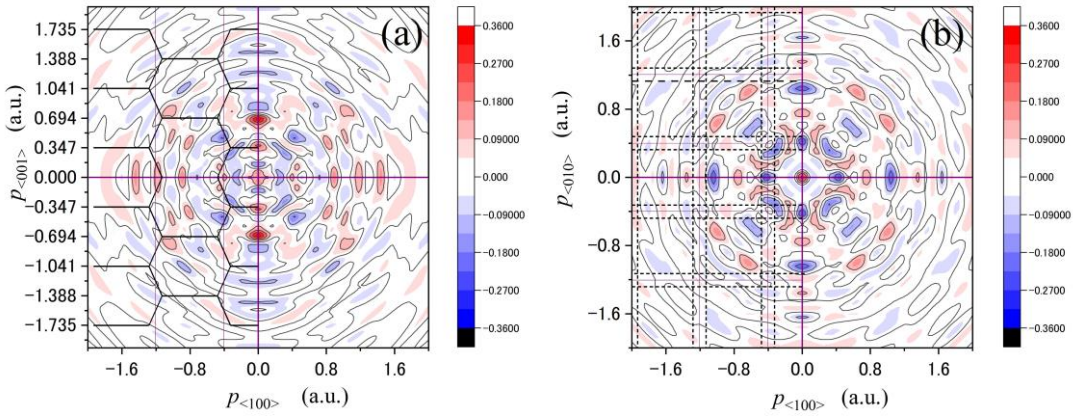

Fig. 2. Difference in 2D-EMD at between 14 K and 20K obtained on the (100) plane (a), and similar figure obtained on the (001) plane (b).

around the Γ point and the hole FS appears around the Z point. Actually, the change seen in Fig. 2 is remarkable at the Γ (Z) points along the coordinate axes. In addition, the band structure shows a high degree of the U 5 *f* electron component around these symmetry points. Taking these points into account, the U 5 *f* electrons in the paramagnetic phase above T_{HO} is described as being still in a partially-localized state and change into the fully-itinerant state through the HO transition. Although the structures around the Γ and Z points overlap each other in Fig. 1 as mentioned above, the degree of overlapping may vary considerably from place to place in the momentum space. If you want to discriminate the structures, it is effective to make a similar experiment on the (110) plane.

4. Summary

The change in electronic structure associated with the HO transition in URu2Si2 was investigated in terms of 2D-EMD. The reconstruction experiments on both the (100) and (001) planes have revealed the change in three dimensions, that is, the difference in 2D-EMD between 14K and 20K identified the positions in the momentum space where the degree of change in density is remarkable. Taking the band structure calculation into consideration, the change in 2D-EMD around the Γ (Z) points will reflect that the U 5 *f* electrons go on to the fully-itinerant state in the HO phase. In addition, if you want to clarify the change in Fermi surface structure, you can apply Lock-Crisp-West analysis, which folds a number of BZs in one BZ, to the 2D-EMD [22]. Then you will obtain 2D-EOND. In fact, we discussed the change of 2D-EOND obtained from the reconstruction experiment on the (001) plane in the previous study. In the case of deriving 2D-EOND from the experiment on the (100) plane, you need beforehand to measure directional Compton profiles at even more high momentum resolution because the BZ is thin along the Γ-Z direction.

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