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Bulk-sensitive high-energy angle-resolved photoemission study of strongly correlated electron systems

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Introduction

For understanding of properties of solids, clarification of their momentum distribution of the electron at the Fermi level (E_F) called as Fermi surface is important. Low-hν (< 100 eV) angle-resolved photoemission (ARPES) has so far been powerful to investigate the character and shape of the Fermi surface as well as the quasiparticle dispersion for layered transition metal oxides such as high-Tc cuprates, manganites, and ruthenites [1]. A quantum oscillation measurement using a de Haas-van Alphen effect is promising for detecting three-dimensional Fermi surfaces of solids, which have three-dimensional electronic structures [2,3]. However, very high-quality single crystals and low temperature are required for the de Haas-van Alphen measurement. High-energy (soft x-ray) ARPES has an advantage in detecting the bulk electronic states and bulk electronic dispersion along the normal (kz-direction) of the cleaved single crystalline surfaces by measuring hν-dependence of the spectra, which are owing to the longer photoelectron mean free path for high-hν soft x-ray ARPES compared with that for the low-hν ARPES [4,5]. Therefore, the high-energy ARPES can be a powerful tool to detect the bulk three-dimensional Fermi surfaces, which is complementary to the de Haas-van Alphen measurement. In this paper, we report hν-dependence of the bulk-sensitive high-energy ARPES spectra and an incident photon momentum effect [6].

Methods and Materials

The soft x-ray ARPES was performed at a twin-helical undulator beamline BL25SU in SPring-8 [7]. The GAMMADATA-SCIENTA SES200 analyzer was used for the measurement. The photoelectrons within polar angles of about ±6° with respect to the normal of the sample surface were simultaneously collected, thereby covering more than a whole Brillouin zone along the direction of the analyzer slit at one particular kz determined by hν as discussed later. Since the sample rotation axis was set to be parallel to the analyzer slit direction, we could perform three-dimensional Fermi surface mapping by one-axis sample rotation and measuring hν-dependence of ARPES spectra. The measuring temperature can be controlled as 20-350 K by using a closed-cycle He refrigerator and a heater.

The measured sample reported here is a typical heavy fermion system $CeRu_2Si_2$, which is a strongly correlated non-magnetic metal with the Kondo temperature of 20 K [8]. Its crystal structure is tetragonal ($ThCr_2Si_2$-type) [9] (For its Brillouin zone, see [10]). The tail of the bulk Kondo resonance has clearly been observed by the Ce 3d-4f resonance angle-integrated photoemission [11]. The clean (001) surface was obtained by cleaving *in situ* at the measuring temperature of 20 K. The absence of possible O 1s and C 1s signals was confirmed by measuring the core-level spectra. The energy resolution was set to 200 meV. The angular resolution was set to ±0.1° (±0.15°) for the perpendicular (parallel) direction to the analyzer slit, which was confirmed at BL25SU. The base pressure was 3 x 10^{-8} Pa.

Results

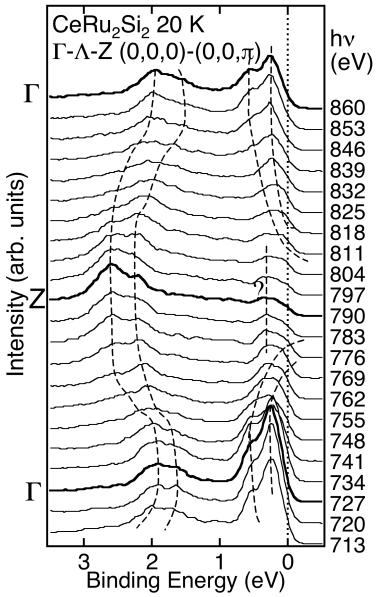

Fig. 1. Soft x-ray angle-resolved photoemission spectra of $CeRu_2Si_2$ along the Γ-Λ-Z [(0,0,0)-(0,0,π)] direction by changing hν.

We show the "normal emission" (kx = ky =0) soft x-ray ARPES spectra of $CeRu_2Si_2$ as shown in Fig. 1. We can notice that there are dispersive peaks as a function of hν in the spectra. The period of the dispersions corresponds to that of the Brillouin zone along the [001] direction, which is proportional to the square-root of the photoelectron kinetic added to the inner potential (V_0~8 eV). According to a conventional formula kz ($Å^{-1}$) = $0.5123[E_K\cos^2\theta + V_0\ (eV)]^{0.5}$ which has often been used for obtaining kz at the low-hν APRES, where E_K (~hν in the case of valence-band high-hν ARPES) and θ denote the photoelectron kinetic energy and polar angle, however, the hν corresponding to the Γ point should be ~755 eV whereas it is experimentally found to be ~727 eV. The feature of the band dispersions is qualitatively consistent with the result of the band-structure calculation [12], but a discrepancy is also seen. In the ARPES spectrum at the Z point (hν = 790 eV), there is a peak near E_F (indicated by a question character). The band-structure calculation predicts that three bands cross E_F near Z and form three small hole-pocket Fermi surfaces, and there is no band near E_F in the occupied side. We conclude that two bands actually cross E_F near Z but one band does not cross E_F, in

contrast to the prediction of the calculation. Our results are consistent with the recent result of the de Haas-van Alphen measurements [13].

Discussion

Incident photon momentum kp is very small compared with the reciprocal vector of solids at low-hν ARPES measurements. For example, kp is ~0.025 $Å^{-1}$ at hν = 50 eV. However, kp is ~0.38 $Å^{-1}$ at hν = 750 eV, which is not negligible at all at high-hν ARPES. This photon momentum is transferred to the created photoelectron in the photoemission process. Therefore, the formula estimating kz must be modified as kz ($Å^{-1}$) = $0.5123[E_K\cos^2\theta + V_0\ (eV)]^{0.5}$ + kpcosϕ, where ϕ denotes the incident angle. We show the calculated kz by using this formula in Fig. 2. The hνs corresponding to the Γ and Z points are estimated as 725 and 790 eV, being consistent with the soft x-ray ARPES result shown in Fig. 1. Our results indicate that the nearly free-electron model for photoelectrons is available also for high-energy ARPES but the incident photon momentum effect must be taken into account for analyzing the ARPES spectra.

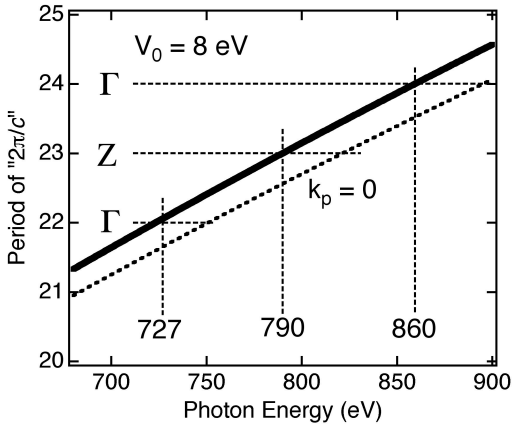

Fig. 2. Calculated kz (unit of 2π/c, where c is the lattice constant for $CeRu_2Si_2$, 9.796 Å [14]) as a function of hν with the inner potential V_0 = 8 eV. The solid (dashed) line shows the kz with considering (neglecting) incident photon momentum. Incident angle is assumed to be 45°.

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