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Soft X-ray angle-resolved photoemission study of YbCu₂Ge₂

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Abstract. Soft X-ray photoemission spectroscopies have been performed on YbCu₂Ge₂, which is thought to be a Yb divalent system, to investigate its bulk electronic structure including Yb 4*f* electrons. Small but finite Yb³⁺ multiplet peaks were found at 6-12 eV in the angle-integrated spectrum, suggesting that it is a valence fluctuation compound. An “anti-crossing” Yb 4*f* bands with conduction bands were observed in the angle-resolved spectrum. This is the evidence that the Yb 4*f* electrons form a band state through the hybridization with Ge 4*p* and Cu 3*d* electrons.

1. Introduction

Yb-based compounds have been attracting much attention due to their interesting physical properties [1, 2, 3]. As a first step toward a unified understanding of these properties, it is important to examine the electronic structure of the valence bands containing the Yb 4*f* electrons. An angle-resolved photoemission spectroscopy (ARPES) is a powerful experimental tool to directly observe the valence bands and the Fermi surfaces. Although many angle-integrated photoemission spectroscopies (AIPES) have been performed for various Yb compounds, there are only a few ARPES studies for YbRh₂Si₂ and YbIr₂Si₂ [4]. The previous ARPES studies have utilized the incident photon of 21.2 to 110 eV. However, those ARPES measurements are surface sensitive and obtained spectra have considerably large contributions of the surface Yb²⁺ state. On the other hands, in the bulk-sensitive soft X-ray ARPES (SX-ARPES) study, the surface Yb²⁺ contribution is suppressed and thus the spectra can be directly compared with the band dispersion and the Fermi surface predicted from band structure calculations. For example, Fermi surfaces obtained by the recent SX-ARPES for CeRu₂Si₂ can be explained by a band structure calculation based on a local-density-approximation (LDA) [7], while they are quite different from those obtained by the previous ARPES with the incident photon of ~ 100 eV [8].

YbCu_2Ge_2 crystallizes in the body-centered tetragonal ThCr_2Si_2 crystal structure with one formula unit per primitive cell. The NMR study [5], and the X-ray absorption spectroscopy study [6] suggested that it is a divalent compound. Dung *et al.* carried out various transport and thermodynamic measurements and a de Haas-van Alphen (dHvA) experiment on this compound [9]. They clarified that YbCu_2Ge_2 shows the Pauli paramagnetic behavior with the small electronic specific coefficient γ of 9-10 mJ/(mol K²). This small γ value indicates that 4*f* bands are located far from the Fermi energy E_F . In addition, the result of the dHvA experiment was explained by “Yb²⁺ model” which was performed by a calculation within such a situation. However, from those previous experiments, it has been not yet known the information of Yb 4*f* electrons, for example, where 4*f* bands are located and whether the 4*f* electrons hybridize with the conduction electrons (*c-f* hybridization) or not. It is important to clarify those kind of the information for various valent compounds to understand the formation of the heavy fermion state in the Yb system. Additionally, in the Yb divalent system, the Yb atom often seems to be Yb²⁺ ion. That means that the Yb 4*f* bands should be flat in the reciprocal space and be separated with the other conduction band in the language of the band picture. Nevertheless, such band structures can not be explained from the band calculation. In order to clarify the bulk valence-band structure, we have carried out the SX-AIPES and -ARPES measurements for YbCu_2Ge_2 .

2. Experiment and calculation

Single crystals of YbCu_2Ge_2 were grown by the Sn-flux method [9]. AIPES and ARPES measurements were performed at soft X-ray beamline BL23SU in SPring-8. We have used the incident photon energy of $h\nu = 450$ eV which corresponds to the high-symmetry plane including the Γ point by assuming the inner potential value of 12 eV. The energy resolution was 90 meV for both AIPES and ARPES measurements. Clean sample surfaces were obtained by *in situ* cleaving samples along [001] surface. The sample temperature was kept at 20 K because YbCu_2Ge_2 does not show the phase transition over all temperature region [9] and the phonon broadening is negligible at the temperature. The valence band structure of YbCu_2Ge_2 was calculated by using a first-principles LDA calculation with the relativistic linearized-augmented-plane-wave (RLAPW) method [10]. This calculation is based on the Dirac one electron equation, so that all the relativistic effects including the spin-orbit (SO) interaction is directly taken into account. In this calculation, we used lattice constants and each atomic position determined by the X-ray diffraction measurement [9].

3. Results and discussion

3.1. Angle-integrated spectrum

Figure 1(a) shows the AIPES spectrum of the valence band of YbCu_2Ge_2 . The Yb 4*f* states and the Cu 3*d* states are mainly observed because their photoionization cross sections (I) are larger than other states in the photon energy of $h\nu = 450$ eV ($I_{\text{Yb}} : I_{\text{Cu}} : I_{\text{other}} \approx 1 : 1/6 : 1/200$). Two sharp peaks centered at 0.12 eV and 1.42 eV are the SO split bands of the bulk Yb²⁺ 4*f* states with the splitting energy of 1.3 eV. In this SX-AIPES spectrum, the surface Yb²⁺ 4*f* states which should be observed at the higher binding energy side of the bulk Yb²⁺ 4*f* peak were hardly observed. The broad peaks centered at 4 eV are originated from the Cu 3*d* state. Contrary to our expectation, we found small but clear Yb³⁺ multiplet peaks at 6-12 eV, which are well reproduced by atomic calculations [11]. Because of no phase transition over temperatures and only one Yb site in a unit cell, that indicates that YbCu_2Ge_2 is a valence fluctuation compound. The existence of the Yb³⁺ multiplet feature in the AIPES spectrum was not predictable from the transport and thermodynamic measurements [9], the NMR one [5], and the X-ray absorption spectroscopy [6]. In these measurements, it may be difficult to separate Yb²⁺ and Yb³⁺ components in valence fluctuation compounds. In the photoemission

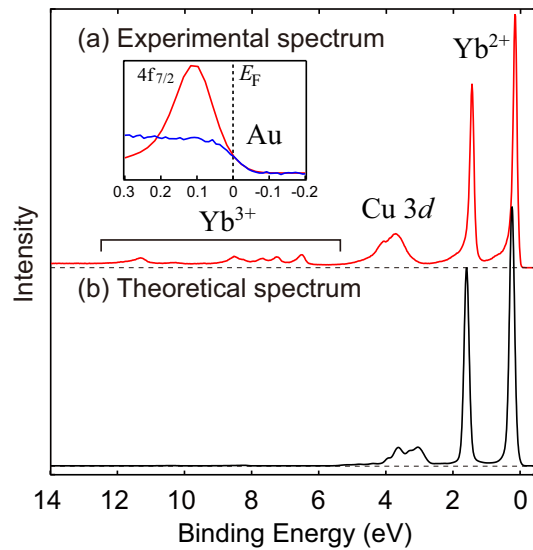


Figure 1. (a) Experimental AIPES spectrum and (b) theoretical one for YbCu_2Ge_2 . Near- E_F region together with the spectrum of Au (blue line) is shown in the inset of (a). The theoretical spectrum is obtained by the RLAPW calculation. It should be noted that the experimental spectrum (a) is the momentum-integrated one in the \mathbf{k} range covering larger than the first Brillouin zone (BZ) in the k_x - k_y plane including the Γ point because of using the single crystal in this experiment. On the other hand, the theoretical one (b) was obtained by the integration over the first BZ.

measurements, Yb^{2+} and Yb^{3+} states are well separated (~ 5 eV). It should be noted that the Yb^{3+} multiplets are not caused by the surface oxidation because O $1s$ peaks were not observed during this measurement. We exhibit the near- E_F region together with a spectrum of Au in the inset of Fig. 1(a). From the band picture, the existence of the Yb^{3+} multiplets indicates that the tail of the Yb^{2+} $4f_{7/2}$ peak should cross E_F . However the clear Fermi edge was not observed, suggesting that the Yb $4f$ states do not mainly contribute to the Fermi surfaces. That may be a reason why YbCu_2Ge_2 seems to be the divalent compound on the previous experimental studies.

In Fig. 1(b), we show a theoretical spectrum simulated from partial density-of-states obtained by the RLAPW calculation, taking into account the photoionization cross sections of each atomic orbital, the energy resolution, and Fermi-Dirac function. The theoretical spectrum is in a qualitative agreement with the experimental one, except for the presence of the Yb^{3+} multiplets. The peak position of the Yb^{2+} $4f$ doublet is almost the same value as the experimental spectrum, while the Cu $3d$ band lies at the lower binding energy side than the experimental one.

3.2. Angle-resolved spectrum

In order to know the c - f hybridization effects in the valence bands, we have performed the ARPES measurements. The ARPES spectra along the X - Σ - X line is shown in Figs. 2(a) and (b). Figure 2(a) depicts the spectra normalized with the integrated intensity of the energy-distribution-curve (EDC) at each k -point. In the EDC spectra, two flat peaks with high intensity are Yb^{2+} $4f$ bands and their peak tops denote by dashed line in Figs. 2(a) and (b). It should be noted that the $4f_{7/2}$ -band locates a little away from E_F as shown in Fig. 1(a). The Cu $3d$ band locates at 3-5 eV with large dispersion and the flat-band centered at 6.5 eV is a part of Yb^{3+} multiplet peaks. The sinusoidal structure lying on the $4f_{5/2}$ -band is conduction bands. We made a second derivative (SD) along the momentum axis to show the peak position of the conduction bands more clearly as shown in Fig. 2(b). In the SD spectra, the intensity of dispersive bands are enhanced, while that of flat bands are weakened. An anti-crossing feature of the Yb $4f_{5/2}$ bands with the conduction bands is clearly observed at the middle point of X - Σ . This is the evidence for the hybridization between the Yb $4f$ electrons and the conduction electrons in the bulk state. From the calculated result, the intersection part of the sinusoidal conduction bands with Yb $4f_{5/2}$ bands is the hybridized band mainly due to the Ge $4p$ and Cu $3d$ electrons.

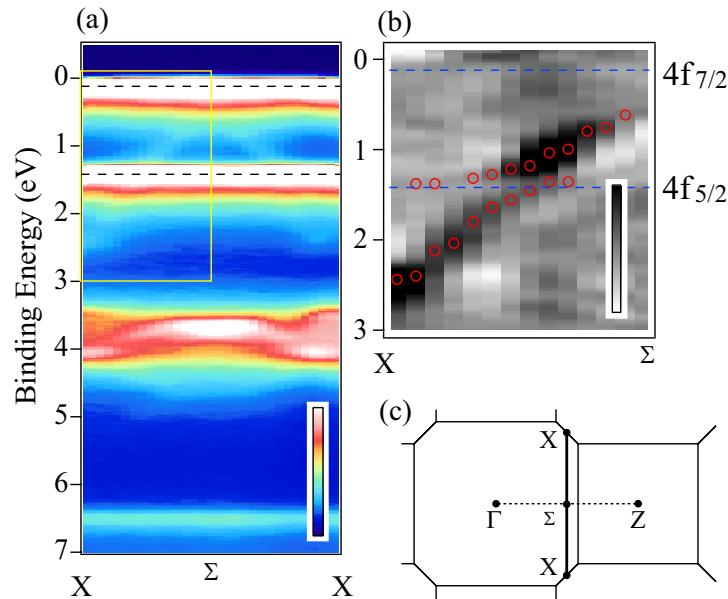


Figure 2. (a,b) ARPES spectra along the X-Σ-X line. The EDC spectra are shown in (a), while (b) shows the negative second derivatives spectra of MDC spectra in the yellow rectangular area of (a). The broken lines indicate peak positions of Yb 4f bands. The open circles represent peak positions of hybridized bands between Yb 4f_{5/2} bands and conduction bands in (b). (c) Brillouin zone and measured k range (bold line) in (a) and (b).

4. Conclusion

We have performed the soft X-ray photoemission spectroscopy to investigate the bulk band structure of YbCu₂Ge₂. YbCu₂Ge₂ has finite Yb³⁺ components, indicating that it is a valence fluctuation compound. That is not expected from the previous experimental studies. The anti-crossing feature of Yb 4f bands with conduction bands shows that the Yb 4f electrons form the band states through the hybridization with the conduction electron. The 4f bands should cross E_F somewhere and make Fermi surfaces because the Yb³⁺ multiplets exist in our spectra. In order to find the evidence of Yb 4f bands crossing E_F , we have carried out ARPES measurements on the high symmetry plane and analyzed the experimental data.

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References

- [1] Stewart G R 1984 *Rev. Mod. Phys.* **56** 755
- [2] Stewart G R 2001 *Rev. Mod. Phys.* **73** 797
- [3] Stewart G R 2006 *Rev. Mod. Phys.* **78** 743
- [4] For example, Danzenbächer S, Kucherenko Yu, Laubschat C, Vyalikh D V, Hossain Z, Geibel C, Zhou X J, Yang W L, Mannella N, Hussain Z, Shen Z-X and Molodtsov S L 2006 *Phys. Rev. Lett.* **96** 106402
- [5] Sampathkumaran E V, Gupta L C and Vijayaraghavan R 1979 *J. Phys. C* **12** 4323
- [6] Padalia B D, Hatwar T K and Ghatikar M N 1983 *J. Phys. C* **16** 1537
- [7] Okane T, Ohkochi T, Takeda Y, Fujimori S -I, Yasui A, Saitoh Y, Yamagami H, Fujimori A, Matsumoto Y, Sugi M, Kimura N, Komatsubara T and Aoki H 2009 *Phys. Rev. Lett.* **102** 216401
- [8] Denlinger J D, Gweon G -H, Allen J W, Olson C G, Maple M B, Sarrao J L, Armstrong P E, Fisk Z and Yamagami H 2001 *J. Electron Spectrosc. Relat. Phenom.* **117** 347
- [9] Dung N D, Matsuda T D, Haga Y, Ikeda S, Yamamoto E, Ishikura T, Endo T, Tatsuoka S, Aoki Y, Sato H, Takeuchi T, Settai R, Harima H and Ōnuki Y 2009 *J. Phys. Soc. Jpn.* **78** 084711
- [10] Yamagami H 1998 *J. Phys. Soc. Jpn.* **67** 3176
- [11] Gerken F 1983 *J. Phys. F* **13** 703