ELSEVIER

Journal of Physics and Chemistry of Solids 63 (2002) 1223–1226

JOURNAL OF PHYSICS AND CHEMISTRY OF SOLIDS

www.elsevier.com/locate/jpcs

Ultrahigh-resolution and angle-resolved photoemission study of SmB_6

S. Nozawa[a,*], T. Tsukamoto[a], K. Kanai[b], T. Haruna[c], S. Shin[b,c], S. Kunii[d]

[a]*Department of Applied Physics, Tokyo University of Science, 1–3 Kagurazaka, Shinjuku-ku, Tokyo 162-8601, Japan*
[b]*The Institute of Physical and Chemical Research (RIKEN), Sayo-gun, Hyogo 679-5148, Japan*
[c]*Institute for Solid State Physics, University of Tokyo, 5-1-5 Kashiwa, Chiba 277-8581, Japan*
[d]*Department of Physics, Tohoku University, Sendai, Miyagi 980, Japan*

Abstract

The photoemission spectra with the ultrahigh-energy resolution reveal the small gap formation at lower temperatures than the characteristic coherent temperature T_{coh}. In addition, the temperature dependence of the density of states at Fermi level clearly shows a simple scaling relationship with T_{coh}. The narrow 4f-band with a very small dispersion just below E_F are observed by angle-resolved spectra at the first time.

Keywords: A. Magnetic materials; C. Photoelectron spectroscopy; D. Electronic structure

1. Introduction

SmB_6 is considered as a typical example of a Kondo insulator [1,2]. The origin of the semiconducting gap at low temperatures in SmB_6 is a long-standing problem and the hybridization-gap model may be a groundwork for the description of its energy gap [3]. The magnetic susceptibility of SmB_6 is well described as that of intermediate valence system [4]. The hump in the susceptibility at about 100 K is concerned with the characteristic spin-fluctuation energy k_BT^*; the system with local moments obeys Curie–Weiss law above T^*. The long-range coherence among the compensated magnetic moments is gradually developed below T^*. One of the key predictions of the hybridization-gap model is a simple scaling relationship between T^* and the magnitude of the direct energy gap E_g [5]. Because the gap-opening is due to the involvement of the conduction electron into the singlet formation, the excitation energy of the compensated magnetic moment, k_BT^*, should be related with the size of the E_g. In fact, the loss of the low-frequency intensities in the optical conductivity of SmB_6 at low temperatures were reported [6].

It is meaningful to investigate the gap formation in detail to get basic information about the semiconducting state of SmB_6. The electronic structure can be directly probed by photoemission spectroscopy. Recently, the improvement in the energy resolution of photoemission spectroscopy enable us to observe the fine structure around Fermi level (E_F) [7]. The photoemission technique is the powerful tool to go into the details of the electronic structure with the small energy gap, such as Kondo insulator.

In this work, we performed the temperature dependent ultrahigh-resolution photoemission (UHRPE) and angle-resolved UHRPE (AR-UHRPE) spectroscopy on SmB_6. The UHRPE spectra reveals not only the properties of the gap but also a clear relationship between T^* and Δ. The AR-UHRPE spectrum at 5.7 K shows the quasi-particle band at E_F with a small dispersion in the Δ-axis direction.

2. Experimental

The single crystal of SmB_6 were grown by a floating zone method. Measurements were carried out in an ultrahigh-vacuum chamber where the base pressure is under 5×10^{-11} Torr. Samples are cooled by using the ^{4}He flow-type cryostat. Clean sample surfaces were obtained by scraping or fracturing these surfaces with a diamond file in the measurement chamber every ~40 min in temperature equilibrium. The UHRPE measurement system consists of a SCIENTA SES-2002 analyzer and a GAMMADATA discharging lamp. The position of E_F and an energy

* Corresponding author. Tel.: +81-471-36-3381; fax: +81-471-36-3383.
E-mail address: shun@issp.u-tokyo.ac.jp (S. Nozawa).

0022-3697/02/$ - see front matter
PII: S0022-3697(02)00020-3

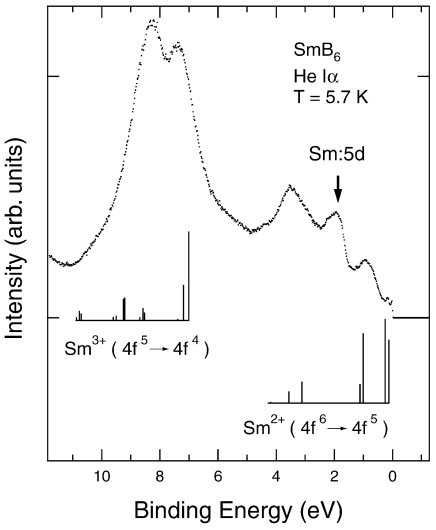

Fig. 1. Valence-band photoemission spectrum of SmB_6 measured by the He Iα resonance line ($h\nu = 21.2$ eV) at 5.7 K. Vertical bars are multiplet structures calculated by Gerken [8].

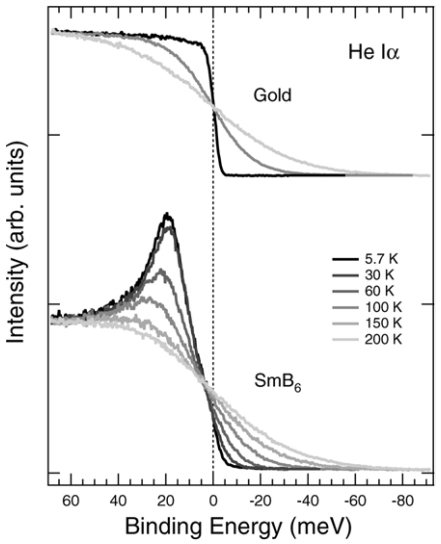

Fig. 2. Upper panel: photoemission spectra of Gold, for comparison. Lower panel: temperature dependence of high-resolution photoemission spectra of SmB_6 near E_F measured by He Iα resonance line ($h\nu = 21.2$ eV). The spectral intensity are normalized by the integrated intensity of the valence band spectra, assuming that the 4f-electron number is conserved within wide energy range ($\leq$1.8 eV).

resolution of the system were determined by referring to the Fermi edge in the UHRPE spectra of Au, which was evaporated on the sample holder. The energy resolution of the system was estimated to be ~2.8 meV. In the AR-UHRPE measurements, the single crystal of SmB_6 was prepared from cleaving in vacuum exposing the (100) surface at $T = 5.7$ K. The AR-UHRPE measurements were operated with energy and angular resolutions of $\Delta E \sim 7.8$ meV, $\Delta\theta \sim \pm 0.25°$.

3. Valence-band spectra

The valence-band spectrum of SmB_6 measured by using He Iα resonance line is shown in Fig. 1. Vertical bars stand for the final-state multiplet structure calculated by Gerken [8]. In this spectrum, we can see two sets of structures, which are caused from divalent ($4f^6 \rightarrow 4f^5$) and trivalent ($4f^5 \rightarrow 4f^4$) excitations. This provides an evidence for the intermediate-valence state as the ground state in SmB_6. Moreover, in the light of a band calculation for EuB_6 [9], the peak at ~2 eV is Sm 5d band. The observed valence-band spectrum well reproduces the spectrum measured by X-ray photoelectron spectroscopy [10] which has a high bulk sensitivity.

4. Temperature dependant UHRPE spectra

In an attempt to study the temperature dependence of gap formation, we measured the photoemission spectra near the Fermi edge from 5.7 to 200 K. The upper panel of Fig. 2 shows the photoemission spectra of Gold for a comparison. The lower panel of the figure shows the photoemission spectra of SmB_6. The Fermi edge of the spectra broadens by raising temperature due to the thermal-excitations of electron. But the spectra of gold at any temperature have midpoints of the leading-edge with the same intensity at Fermi level, E_F. With decreasing temperature, we observe a sharp peak grows at ~20 meV in spectra of SmB_6 and the opening of the small energy gap, which leads to a shift in the leading edge of the spectra.

In order to isolate the temperature dependence of the spectral density of states (DOS), we have divided the spectra by the Fermi–Dirac (FD) distribution function which is convoluted with the experimental energy resolution as shown in Fig. 3. The spectra directly reflect the DOS as a function of temperature. The DOS below ~20 meV is nearly constant at temperature above 150 K. The DOS below ~10 meV gradually decreases and a quasi-particle peak grows at around 20 meV by lowering temperature. The spectral intensity transfers from the neighborhood of E_F to the higher binding-energy region ($\geq$10 meV). The transfer of the intensity notably appears below 100 K. The energy difference between this quasi-particle peak (indicated by arrows) and the bottom of the conduction band is corresponding to the direct energy-gap size of E_g. The quasi-particle peak in 5.7 K is observed in higher binding energy side than that in 30 K. This indicates that the gap strongly depends on temperature and it is caused by many-body effect. In spectra measured above 60 K, the energy position of a quasi-particle peak cannot be estimated much correctly from the spectra due to the thermal-excitation broadening effect.

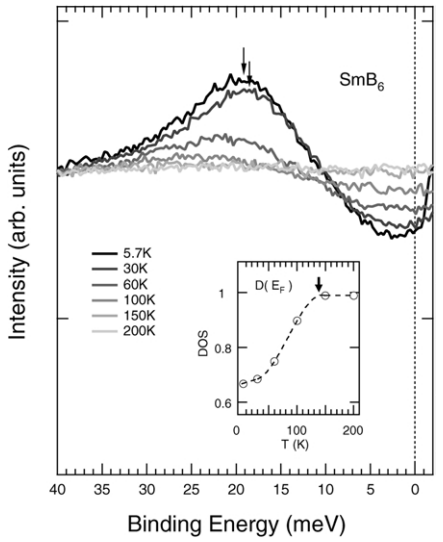

Fig. 3. Temperature dependence of DOS of SmB_6 deduced by dividing the spectra by the FD function. The inset shows a plot of the intensity at E_F versus reduced temperature.

In the Kondo-lattice system, the coherence temperature T_{coh}, where the coherent Fermi liquid starts to be developed with decreasing temperature, is estimated by the characteristic hump in the electrical resistivity. However, in a Kondo insulator system, it is rather difficult to estimate the T_{coh} by the experiment of resistance, because the resistance rises without forming a clear hump due to opening a gap accompanied with the coherent hybridization of Kondo compensated magnetic moments [11]. We examined, therefore, the coherence temperature T_{coh} from behavior of the intensity of the spectra at E_F, $D(E_F)$. The $D(E_F)$ of the spectra in Fig. 3 are plotted against temperature in the inset. We can see that $D(E_F)$ is gradually suppressed from ~140 K shown by the arrow. It indicates the opening of the gap at E_F due to the hybridization between 4f and conduction states. Consequently, our result suggests T_{coh} is about 140 K. The value of T_{coh} estimated from the electron spin resonance spectrum is also resorted to be about 150 K [12]. This supports our results of the UHRPE spectra.

Recently, there is interesting report using optical measurements that an intra-gap state lying ~3 meV below the bottom of the conduction band is observed [6,13]. Then, in order to acquire the knowledge of the electronic state in a gap, we observed electron DOS in the range in the vicinity of E_F from 4.9 to 15 K using UHRPE spectroscopy. The lower panel of the Fig. 4 shows the UHRPE spectra of SmB_6. The structure in the high binding energy correspond to the quasi-particle peak in Fig. 3. We observe the small structures around 4 meV (indicated by dotted line). The upper panel of Fig. 4 shows the spectra divided by FD distribution function convoluted by experimental energy resolution. We can see the small structures around 4 meV also in these spectra and these are almost independent of temperature. Therefore, we suggest that this structure reflects the localized narrow band, which exists at slightly below E_F, and the energy difference between this narrow band and the bottom of conduction band is few meV and it is considered to be the gap detected in the optical conductivity.

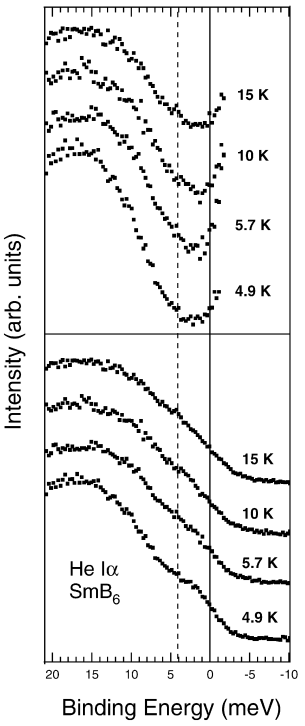

Fig. 4. Temperature-dependent photoemission spectra in the vicinity of E_F of SmB_6 (lower panel) and those divided by FD function (upper panel).

5. Angle-resolved UHRPE spectra

The intensity plot of the AR-UHRPE spectra at 5.7 K of SmB_6 along the Γ–X line (Δ-axis) measured by using He Iα resonance line are shown in Fig. 5. The dotted line is a guide for eyes. It is difficult to identify the hybridization gap character in the reciprocal lattice space from the ARPE spectrum reported before [14]. On the other hand, we observe the quasi-particle band around 20 meV with a small dispersion in the Γ–X line. Accordingly, it is confirmed that the strong hybridization arises in Δ-axis. The result is qualitatively consistent with the band calculation result reported before [15], although the calculation showed the larger dispersion than our result in the 4f band. Consequently, compared with the band calculation, our result suggests the more localized character of the 4f band.

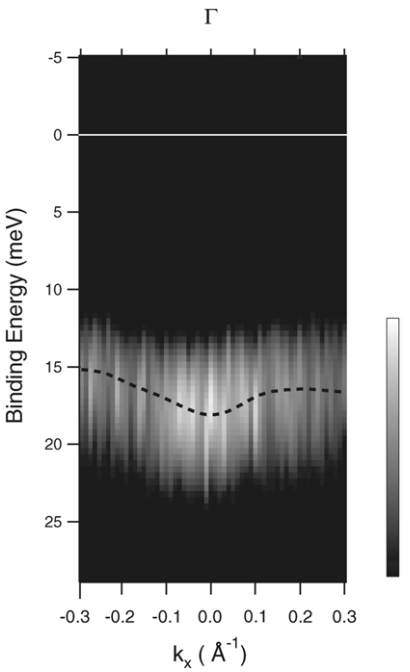

Fig. 5. Intensity plot of spectra along the Γ–X line near the Fermi energy measured by the He Iα resonance line ($h\nu = 21.2$ eV). The dotted line is a guide for eyes.

6. Conclusions

In this work, the UHRPE spectroscopy on SmB_6 is reported. The valence band spectra show the characteristic multiplet structures of the valence fluctuating Sm system. On the other hand, from the results of the UHRPE spectra near E_F, we found a the small gap formation at lower temperatures than coherent temperature T_{coh}. In addition, the temperature dependence of the $D(E_F)$ clearly shows a simple scaling relationship with T_{coh}. The narrow 4f-band with a very small dispersion just below E_F are observed by AR-UHRPE spectra at the first time.

Acknowledgements

One of us (K.K.) acknowledges the support of a fellowship from Special Postdoctoral Researches Program at RIKEN.

References

[1] T. Kasuya, Europhys. Lett. 26 (1994) 277.
[2] G. Aeppli, Z. Fisk, Comments, Condens. Matter Phys. 16 (1992) 155.
[3] P. Wachter, in: K.A. Gschneider Jr., L. Eyring (Eds.), Handbook on the Physics and Chemistry of Rare Earths, vol. 19, North-Holland, Amsterdam, 1994, p. 177.
[4] J. Roman, V. Pavlik, K. Flachbart, Th. Herrmannsdörfer, S. Rehmann, E.S. Konovalova, Yu.B. Paderno, Physica B 230–232 (1997) 715.
[5] M. Jarrell, Phys. Rev. B 51 (1995) 7429.
[6] B. Gorshunov, N. Sluchanko, A. Volkov, M. Dressel, G. Knebel, A. Loidl, S. Kunii, Phys. Rev. B 59 (1999) 1808.
[7] T. Kiss, T. Yokoya, A. Chainani, S. Shin, J. Electron Spectrosc. Relat. Phenom. 635 (2001) 114.
[8] F. Gerken, J. Phys. F 13 (1983) 703.
[9] S. Massidda, A. Continenza, T.M. De Pascale, R. Monnier, Z. Phys. B 102 (1997) 83.
[10] J.W. Allen, L.I. Johansson, I. Lindau, S.B. Hagstrom, Phys. Rev. B 21 (1980) 1335.
[11] J.C. Nickerson, R.M. White, K.N. Lee, R. Bachmann, T.H. Geballe, G.W. Hull Jr., Phys. Rev. B 3 (1971) 2030.
[12] T.S. Al'tshuler, V.N. Mironov, G.G. Khaliullin, D.I. Khomskii, Pis'ma Zh Eksp. Teor. Fiz. 40 (1984) 28 (JETP Lett. 40 (1984) 754).
[13] N.E. Sluchanko, V.V. Glushkov, B.P. Gorshunov, S.V. Demishev, M.V. Kondrin, A.A. Pronin, A.A. Volkov, A.K. Savchenko, G. Grüner, Y. Bruynseraede, V.V. Moshchalkov, S. Kunii, Phys. Rev. B 61 (2000) 9906.
[14] J.D. Denlinger, G.-H. Gweon, J.W. Allen, C.G. Olson, Y. Dalichaouch, B.-W. Lee, M.B. Maple, Z. Fisk, P.C. Canfield, P.E. Armstrong, Physica B 281 and 282 (2000) 716.
[15] A. Yanase, H. Harima, Prog. Theor. Phys. Supp. 108 (1992) 19.