SYSTEMATIC EVOLUTION OF THE KONDO PEAK IN $YbCu_{5-x}Ag_x$*

T. Okane[a], S.-I. Fujimori[a], K. Mamiya[a], J. Okamoto[a]
A. Fujimori[a,b], N. Tsujii[c] and K. Yoshimura[d]

[a]Synchrotron Radiation Research Center
Japan Atomic Energy Research Institute, Mikazuki, Hyogo 679-5148, Japan
[b]Department of Complexity Science and Engineering, University of Tokyo
Bunkyo-ku, Tokyo 113-0033, Japan
[c]National Institute for Materials Science, Tsukuba, Ibaraki 305-0047, Japan
[d]Department of Chemistry, Kyoto University, Kyoto 606-8502, Japan

(Received July 10, 2002)

The electronic structure of the $YbCu_{5-x}Ag_x$ system, which belongs to a series of dense Kondo compounds, is studied by high-resolution photoemission spectroscopy. A sharp Yb 4*f*-related peak, which is regarded as the Kondo peak, was observed just below the Fermi level for each compounds. As x decreases, the intensity of the Kondo peak decreases and its energy position is shifted towards the Fermi level.

PACS numbers: 71.27.+a, 75.30.Mb, 79.60.–i

A Series of $YbCu_4M$ compounds (M = Ag, Au, Pd, In, *etc.*) crystallize in the cubic $AuBe_5$-type structure and show rich variety of low-temperature properties associated with the valence instability of Yb [1]. $YbCu_4Ag$ shows a typical dense Kondo behavior and has a moderately large electronic specific heat coefficient of $\gamma \cong 245$ mJ/mol K^2 [1, 2]. Its solid solution system $YbCu_{5-x}Ag_x$ crystallizes in the cubic $AuBe_5$-type structure in the range of $0.125 \leq x \leq 1$ and belongs to a series of dense Kondo compounds [3]. It was experimentally confirmed that the characteristic temperature, which is proportional to the Kondo temperature and was determined with the magnetic susceptibility measurement, varies as 181.0, 121.3, 102.0, 86.2 K for $x = 1.0$, 0.7, 0.5, 0.25, respectively [3]. The specific heat coefficient γ increases from 210 to 440 mJ/mol K^2 in going from $x = 1.0$ to 0.25 [3].

* Presented at the International Conference on Strongly Correlated Electron Systems, (SCES 02), Cracow, Poland, July 10–13, 2002.

The electronic structures of Yb Kondo systems have been extensively investigated by photoemission spectroscopy (PES) [4]. The behavior of the observed $4f$ signal has been successfully explained with the single impurity Anderson model (SIAM) using non-crossing approximation (NCA), which predicts the existence of a sharp many-body resonance, so-called Kondo peak, located at $\sim k_{\mathrm{B}}T_{\mathrm{K}}$ (T_{K} Kondo temperature) below the Fermi level E_{F} with the width of $\sim k_{\mathrm{B}}T_{\mathrm{K}}$, which loses its spectral weight as the temperature rises [5]. A PES experiment on $YbCu_4Ag$ was performed to study the temperature dependence of the Kondo peak by changing the sample temperature and observed the behavior consistent with the NCA calculation [6]. However, another group objected the Kondo scaling for Yb-based Kondo systems based on the quantitative inconsistency in the Kondo temperature dependence of $YbCu_4Ag$ and $YbCu_4Au$, pointing out the importance of thermal broadening of the Fermi edge and the sharp peak to interpret experimental data [7].

In this study, we have investigated the alloy system $YbCu_{5-x}Ag_x$ ($x = 1.0, 0.7, 0.5, 0.25$) by high-resolution PES. The variation of the Kondo peak corresponding to the difference of the Kondo temperature has been observed. Since the measurements were made under a fixed temperature of 14 K, the observed variation was not disturbed by the thermal broadening of the Fermi edge and the peak signal.

Polycrystalline samples of $YbCu_{5-x}Ag_x$ were prepared from 99.9% pure Yb, 99.999% pure Ag and Cu metals by argon arc melting. Details of the sample preparation are described in Ref. [3]. The PES spectra were measured with two excitation energies: He I radiation ($h\nu = 21.2$ eV) produced by a He discharge lamp and synchrotron radiation ($h\nu = 700$ eV) produced at the beam line BL23SU of SPring-8. The energy resolution for the He I and synchrotron radiation measurements were 5 meV and 200 meV, respectively. Clean surfaces were obtained by scraping *in situ* with a diamond file. Details of the PES measurements are the same as described in Ref. [8].

Fig. 1 shows the valence band spectra of $YbCu_{5-x}Ag_x$ for $h\nu = 700$ eV. The sample temperature was fixed at $T = 14$ K. The two peaks at binding energies E_{B} of about 0.1 and 1.4 eV are the spin-orbit-doublet of the $4f^{13}_{7/2}$ and $4f^{13}_{5/2}$ final states of Yb^{2+}. The complicated peak structures in the range from 5 to 11 eV are the $4f^{12}$ multiplet structure of Yb^{3+}, and are well described by the calculated atomic multiplet lines [9]. The features in the range from 2 to 5 eV are dominated by the Cu $3d$ band. In the range from 5 to 7eV, the Ag $4d$ band structure, which is not clearly observed in the figure, overlaps the $Yb^{3+}4f$ signal. Small humps around 1.0 and 2.3 eV are the surface component of the spin-orbit-doublet of the Yb^{2+} $4f$ signal. The spectra in Fig. 1 have been normalized to the height of the Cu $3d$

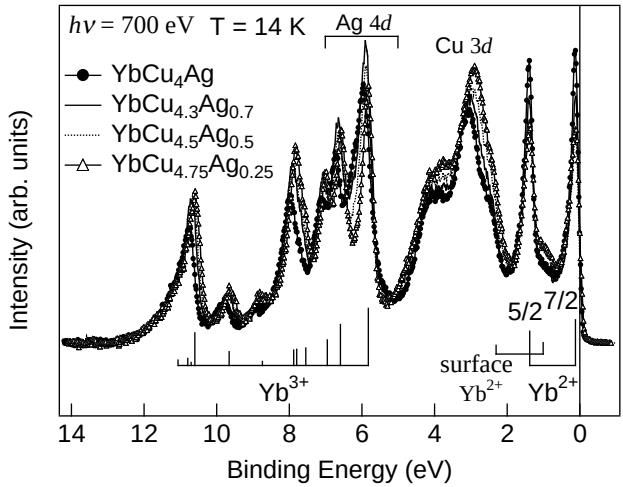

Fig. 1. Valence-band spectra of $YbCu_{5-x}Ag_x$ ($x = 1.0$, 0.7, 0.5, 0.25). The photon energy is 700 eV. Background due to inelastically scattered electrons has been subtracted from each spectrum. The bar diagrams are from the multiplet calculation in Ref. [9].

structure around 3 eV, multiplied by a factor of $5 - x$. As x decreases, the intensity of the Yb^{2+} signal decreases and the intensity of the Yb^{3+} signal especially at around 7.9 and 10.6 eV increases, while the intensities of the peak structures in the range from 5 to 7 eV show a complicated behavior due to the mixture of an increase in the Yb^{3+} signal and a decrease in the Ag $4d$ signal. Thus, the intensity ratio Yb^{3+}/Yb^{2+} increases with decreasing x, meaning an increase in the Yb $4f$ hole number and thus an increase in the Yb valence. This tendency is consistent with the increase in the γ value with decreasing x. One can also notice that the peak positions of the Yb^{3+} $4f$ multiplet structure shift towards E_F as x decreases. This indicates a rise of the $4f$ energy level with decreasing x, which leads to the stabilization of the Yb^{3+} configuration compared to Yb^{2+}.

The near-E_F region measured with a high energy resolution is shown in Fig 2. The observed peak structure in each spectrum is regarded as the Kondo peak whose energy position, intensity, and width can be related to the Kondo temperature of the compounds. As x decreases, the intensity of the Kondo peak decreases and its energy position shifts from 20 meV to 14 meV in going from $x = 1.0$ to 0.25. This behavior can be interpreted as reflecting the drop of the Kondo temperature with decreasing x, based on the Kondo scaling scenario of SIAM with NCA [5]. To make a quantitative discussion, the data should be analyzed with NCA calculations, which will be presented elsewhere.

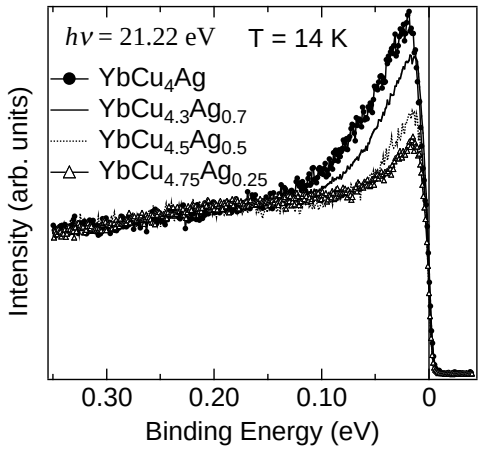

Fig. 2. High-resolution spectra of $YbCu_{5-x}Ag_x$ in the near-E_F region recorded with He I radiation. The spectra have been normalized to the area from 0.15 to 0.40 eV.

In conclusion, the PES experiment for $YbCu_{5-x}Ag_x$ has revealed an increase in the valence, a rise of the Yb $4f$ energy level, an energy shift and intensity variation of the Kondo peak with decreasing x, *i.e.*, decreasing Kondo temperature.

We thank M. Shimizu for useful advice.

REFERENCES

[1] C. Rossel, K.N. Yang, M.B. Maple, Z. Fisk, E. Zirngiebl, J.D. Thompson, *Phys. Rev.* **B35**, 1914 (1987).

[2] M.J. Besnus, P. Haen, K. Yoshimura, K. Kosuge, H. Michor, K. Kreiner, G. Hilshner, *Physica B* **163**, 571 (1990).

[3] N. Tsujii, J. He, N. Hamdaoui, A. Herr, A. Meyer, *Phys. Rev.* **B55**, 1032 (1997).

[4] D. Malterre, M. Grioni, Y. Baer, *Adv. Phys.* **45**, 299 (1996).

[5] N.E. Bickers, D.L. Cox, J.W. Wilkins, *Phys. Rev.* **B36**, 2036 (1987).

[6] P. Weibel, M. Grioni, D. Malterre, B. Dardel, Y. Baer, M.L. Besnus, *Z. Phys.* **B91**, 337 (1993).

[7] J.M. Lawrence, A.J. Arko, J.J. Joyce, P.C. Canfield, Z. Fisk, J.D. Thompson, R.J. Barlett, *J. Magn. Magn. Mater.* **108**, 215 (1992).

[8] T. Okane, S.-I. Fujimori, A. Ino, A. Fujimori, S.K. Dhar, C. Mitra, P. Manfrinetti, A. Palenzona, O. Sakai, *Phys. Rev.* **B65**, 125102 (2002).

[9] F. Gerken, *J. Phys. F* **13**, 703 (1983).