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High resolution photoemission study of $YbAl_3$ at low temperature

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The valence v and the $4f^{14} \rightarrow 4f^{13}_{7/4}$ transition energy T_K of $YbAl_3$ at low temperature were determined by the high resolution valence band photoemission spectroscopy. The obtained values $v = 2.65 \pm 0.03$ and $T_K = 30 \pm 15$ meV are consistent with the zero-temperature magnetic susceptibility $\chi_m(0)$, supporting the Anderson Hamiltonian description for its electronic structure.

1. Introduction

The Anderson Hamiltonian description of the electronic structures of Ce compounds has been so successful [1] that it is natural to ask whether a similar approach can be applied to other rare earth compounds. In particular, Yb compounds have received most attention in this respect because of the electron-hole symmetry between Yb and Ce compounds [2]. That is, the results obtained for Ce compounds can be applied to Yb compounds by simply interchanging the role of the 4f electron with that of the 4f hole. For example, the 4f spectral weights calculated for Ce compounds can be used for interpreting photoemission and inverse photoemission spectra of Yb compounds by reversing the direction of the electron energy axis. Recent photoemission spectroscopy (PES) and Bremsstrahlung isochromat spectroscopy (BIS) studies [3,4] on $YbAl_3$ have been interpreted in this framework, and it was proposed that the hybridization between the 4f electron and the conduction electron of this Yb compound is smaller than that of most Ce mixed-valent compounds but still substantial.

Although $YbAl_3$ has been studied by many different experimental techniques, the PES/BIS study is unique in that it can determine the parameters linking the so-called 'high-energy' and 'low-energy' experiments within the Anderson Hamiltonian framework. For example, the inelastic neutron scattering experiment can give information on the Kondo temperature T_K, but the valence of the Yb 4f ion v cannot be determined. On the other hand, the Mössbauer experiment can determine the valence, but cannot give information on the Kondo temperature. But in the photoemission experiment, both v and T_K can be determined simultaneously, which in turn can give information on the hybridization strength Δ between the 4f level and conduction electrons and the bare 4f energy level ε_f^0 through the relations derived from the Anderson Hamiltonian [2,5,6]. In this way, both low and high energy parameters can be checked for consistency, and the applicability of the Anderson Hamiltonian to describe the electronic structure of this compound can be studied.

Unfortunately, the two previously published results on PES/BIS studies of $YbAl_3$ do not agree with each other [3,4], and even the consistency of the Anderson Hamiltonian parameters deduced from the PES/BIS measurements with a static property such as the zero-temperature magnetic susceptibility of $YbAl_3$ is in doubt [7]. In this paper, we report the result of a high resolution, low temperature ($T \simeq 15$ K) photoemission spectroscopy study of $YbAl_3$ to settle the controversy, and to determine the applicability of the Anderson Hamiltonian description for this compound.

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2. Experiment

The experiment was carried out at the Ames/Montana beamline in the Synchrotron Radiation Centre (SRC) of the University of Wisconsin, USA. The photon energy monochromator was the extended range grasshopper, and the experiment was done with the angle resolved chamber (base pressure $\leq 1 \times 10^{-10}$ Torr) equipped with a 50 mm radius hemispherical analyzer made by VSW. The $YbAl_3$ sample was a polycrystalline ingot and a clean surface was obtained by fracturing in situ. This sample was from the same ingot that had been used for previous PES and BIS studies by our group [3]. The temperature of the $YbAl_3$ sample was lowered to about 15 K using a closed-cycle helium refrigerator in order to minimize the thermal broadening. The Fermi level reference was determined by measuring the valence band spectra of the sputtered Pt foil at the same time as the $YbAl_3$ spectra.

3. Data analysis

We first found the valence v of the Yb 4f ion for $YbAl_3$ using the intensity ratio between the divalent and the trivalent components in the valence band PES spectrum. For this purpose, we obtained the wide range spectrum with the 100 eV photon energy as represented by dots in fig. 1. The peaks between E_F and 3 eV binding energy are from the $4f^{14} \rightarrow 4f^{13}$ (divalent) transition and the peaks between 5 eV and 11 eV are due to the $4f^{13} \rightarrow 4f^{12}$ (trivalent) transition. The results of the curve fitting with the spin-orbit structure of the $4f^{13}$ final state and the multiplet structure of the $4f^{12}$ final state are shown by the solid lines in the same figure. We considered the surface and bulk effects which are already known for Yb compounds in the $4f^{13}$ final state [3]. We obtained the intensity ratio between the bulk divalent peak $I(4f^{13})$ and the bulk trivalent peak $I(4f^{12})$ from the result of this curve fitting, and found that $I(4f^{13})/I(4f^{12}) = 0.576 \pm 0.050$. Neglecting the hybridization effect which may transfer weights between two final states (the hydridization effect for Yb compounds is expected to be smaller than that for Ce compounds because of the lanthanide contraction of the 4f wave function), we can obtain valence $v = 2 + n_f$ and the number of the 4f hole n_f from the relation

$$n_f = \frac{1}{1 + \frac{13}{14} \times \frac{I(4f^{13})}{I(4f^{12})}} \quad (1)$$

The value of n_f obtained in this way is 0.65 ± 0.03. This valence is smaller than that obtained in [3], $v = 2.78 \pm 0.03$, using the PES spectra taken at room temperature. In fact, the change in valence with temperature of this order of magnitude is expected theoretically due to the thermal occupation of low-lying magnetic states [2].

Since the $4f^{14} \rightarrow 4f^{13}$ electron removal peak position in the PES spectra of Yb compounds ($4f^0 \rightarrow 4f^1$ electron addition peak position in the BIS spectra of Ce compounds) is located at T_K from the Fermi level according to the result of calculations for the 4f spectral weight of the Anderson impurity Hamiltonian [2,5,6], the value of T_K can be obtained by measuring the binding energy of the $4f^{14} \rightarrow 4f^{13}$ peak precisely in PES. Since the value of T_K for $YbAl_3$ is a few hundred degrees Kelvin, it requires a high resolution experimental set up. It is also necessary to know the Fermi level position accurately (work function of the instrument) and the resolution of the experimental instrument. For this purpose, we measured the spectrum of the Pt metal with 50 eV photon energy with exactly the same setting used for the $YbAl_3$ spectra, the raw data of which are shown in fig. 2 by dots. Considering the DOS of the Pt metal as flat (which is not a bad approximation for this narrow range) and the resolution of the experimental instrument given by the Gaussian function, we fitted this spectra by the convolution method, the result of which is represented by the solid line in fig. 2. By this method, we were able to determine the Fermi level position with an accuracy of ± 15 meV and the total instrumental resolution was found to be Γ (full width at half maximum: FWHM) $= 84 \pm 10$ meV. We have negelcted thermal Fermi function broadening at this low temperature.

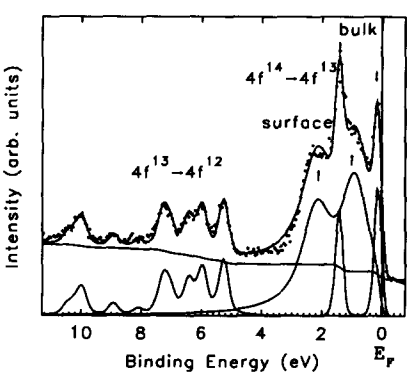

Fig. 1. Wide range valence band spectrum of $YbAl_3$ by using the photon energy 100 eV. Dots represent the raw data and the solid line represents the fit.

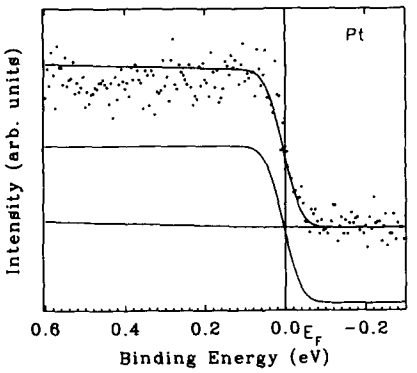

Fig. 2. Spectrum of Pt metal near the Fermi level with high instrumental resolution Γ (FWHM) = 84 ± 10 meV by using the photon energy 50 eV. Dots represent the raw data and the solid line represents the fit.

We now analyze the high resolution data of $YbAl_3$ taken with the 50 eV photon energy shown by dots in fig. 3. Using the position of E_F and the instrumental resolution obtained as mentioned above, we tried to fit the spectrum with the least-squares curve fitting procedure. We assumed Lorentizian peak shapes for two broad surface peaks and the bulk $4f^{13}_{5/2}$ peak at ~1.3 eV from the Fermi level. However, when we assumed the Lorentzian shape for the bulk $4f^{13}_{7/2}$ peak close to the Fermi level, we found that the rising edge at the Fermi level could not be fitted with the known instrumental resolution of Γ (FWHM) = 84 ± 10 meV. In fact, this is expected since the theory shows that the Kondo resonance has non-Lorentzian shape [2], and its width is larger at the higher binding energy side for Yb compounds. We tried to mimic this shape by

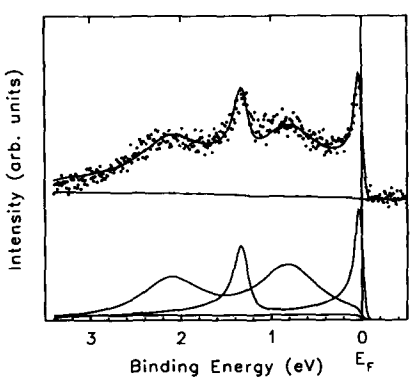

Fig. 3. Spectrum of $YbAl_3$ near the Fermi level with high instrumental resolution Γ (FWHM) = 84 ± 10 meV by using the photon energy 50 eV. Dots represent the raw data and the solid line represents the fit.

assuming the quasiparticle peak line shape in the Fermi liquid theory [8], where the lifetime width γ is proportional to the square of the distance from the Fermi level. That is, we use the following intrinsic line shape function for the bulk $4f^{13}_{7/2}$ peak:

$$f(\varepsilon) = \frac{A(\varepsilon - E_F)^2}{(\varepsilon - T_K)^2 + A^2(\varepsilon - E_F)^4}, \tag{2}$$

where T_K is the peak position and E_F is the Fermi level. Also, since the $YbAl_3$ compound is metallic, we must consider the many body effect resulting from the Coulomb interaction between the valence electron and the core hole. Hence we convoluted with the asymmetry function of the $\exp(-\varepsilon/\xi)/\varepsilon^{1-\alpha}$ form, where ξ is the Mahan cutoff energy (we use ξ to be 6.0 eV) and α represents the asymmetry parameter [9]. The result obtained by the convolution of these various functions is represented by the solid line in fig. 3. We found from this curve fitting that the T_K value for the bulk $4f^{13}_{7/2}$ peak is 30 ± 15 meV, and the asymmetry parameter α is 0.2, which is the same as the value used to fit the wide range valence band spectrum of fig. 1. The coefficient A used in the fitting was 44.5 eV^{-1}, and the Lorentzian width (FWHM) for the bulk $4f^{13}_{5/2}$ peak was 120 meV.

4. Discussion and summary

Now that n_f and T_K have been obtained from experimental PES data, we can predict the value of the zero-temperature magnetic susceptibility $\chi_m(0)$ using the following relation obtained in the lowest order solution of the Anderson impurity Hamiltonian [2,5,6]:

$$\chi_m(0) = \frac{\mu^2_{eff}}{3} \times \frac{n_f}{T_K}, \tag{3}$$

where $\mu_{eff} = \sqrt{j(j+1)}g\mu_B = 4.54\mu_B$ for the Yb $4f^{13}_{7/2}$ ion. Using our values of $T_K = 30 \pm 15$ meV and $n_f = 0.65 \pm 0.03$ obtained above, we find $\chi_m(0) = (4.98 \pm 1.70) \times 10^{-3}$ emu/mol, which is close to the experimental value of 4.62×10^{-3} emu/mol for $YbAl_3$ [10]. This shows that the Anderson Hamiltonian parameters obtained in the 'high energy' PES experiment can also explain the 'low energy' magnetic susceptibitily measurement, and the Anderson Hamiltonian framework is a valid starting point for discussing electronic structures of Yb compounds as in the case of Ce compounds.

By using the n_f and T_K values obtained above, we can also estimate the hybridization parameter Δ and

the bare 4f electron energy level $|\varepsilon_f^0|$ from the following lowest order relation of the Anderson impurity Hamiltonian [2,5,6]:

$$\frac{n_f}{1-n_f} = \frac{N_f}{\pi} \times \frac{\Delta}{T_K}, \qquad \varepsilon_f^0 = T_K + \frac{N_f \Delta}{\pi} \times \ln\left(\frac{T_K}{B}\right), \tag{4}$$

where N_f is the degeneracy of the f level, which is taken to be 8 here since the spin–orbit splitting energy for the Yb 4f level is about 1.30 eV, and B is the conduction band width (assumed to be 2.0 eV here). We find Δ and $|\varepsilon_f^0|$ to be 21.9 ± 10.0 meV and 390 ± 90 meV, respectively. These values should not be taken too literally because we have neglected all higher order corrections and the finite U effect. However, we see that the hybridization parameter Δ is smaller than that of typical Ce mixed-valence compounds as expected from the lanthanide contraction of the 4f wave function, but it is still big enough to be significant in determining the physical properties. Also we can say that the spin fluctuation is more important than the charge fluctuation for $YbAl_3$, since the value of $|\varepsilon_f^0|$ is much larger than $N_f\Delta$ and T_K.

In summary, we find the Anderson Hamiltonian parameters of $YbAl_3$ from the high-resolution, low-temperature photoemission spectra. These parameters describe both the low and high energy properties of $YbAl_3$ consistently, and the spectral features are consistent with the prediction based on the Anderson impurity Hamiltonian. We conclude that the Anderson impurity Hamiltonian is a good starting point to describe the electronic structures of Yb compounds.

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