Local moment behaviors of the valence fluctuating systems $\beta$-YbAlB$_4$ and $\alpha$-YbAlB$_4$

Y. Matsumoto, K. Kuga, N. Horie and S. Nakatsuji
Institute for Solid State Physics, University of Tokyo, Kashiwa 277-8581, Japan
E-mail: matsumoto@issp.u-tokyo.ac.jp, satoru@issp.u-tokyo.ac.jp

Abstract. $\beta$-YbAlB$_4$ is the first example of an Yb-based heavy Fermion superconductor with $T_c = 80$ mK and exhibits pronounced non-Fermi-liquid behavior above $T_c$. On the other hand, recent hard x-ray photoemission spectroscopy measurements have revealed strong intermediate valence of both $\beta$-YbAlB$_4$ and its locally isostructural polymorph $\alpha$-YbAlB$_4$. Here we present the results of the specific heat and magnetization measurements of $\beta$-YbAlB$_4$ and $\alpha$-YbAlB$_4$. The results indicate a Fermi liquid ground state for $\alpha$-YbAlB$_4$ in contrast to the quantum criticality found in $\beta$-YbAlB$_4$. Interestingly, both systems exhibit Kondo lattice behavior with large Wilson ratio below $T^* \sim 8$ K, through the renormalization of a high valence fluctuation scale $\sim 200$ K.

1. Introduction

$4f$ based heavy Fermion (HF) systems have provided prototypical systems to study interesting phenomena in the vicinity of quantum critical points, such as unconventional superconductivity and non-Fermi-liquid (NFL) phenomena. While a number of unconventional superconductors including CeCu$_2$Si$_2$, CeCoIn$_5$, and CeIrIn$_5$, have been found in Ce ($4f^1$) based intermetallic HF systems near a quantum critical point, much attention has also been paid to the unconventional quantum criticality, which does not follow the standard SDW description, observed in systems such as CeCu$_{6-x}$Au$_x$ and YbRh$_2$Si$_2$ [1, 2].

Our recent studies have found the first Yb ($4f^{13}$) based HF superconductor with the transition temperature $T_c = 80$ mK in the new compound $\beta$-YbAlB$_4$ [3, 4]. Pronounced non-Fermi-liquid behavior above $T_c$ and its magnetic field dependence indicate that the system is a rare example of a pure metal that displays quantum criticality at ambient pressure and close to zero magnetic field [3]. Furthermore, the $T/B$ scaling found in our recent high-precision magnetization measurements clarifies its unconventional zero-field quantum criticality without tuning [5]. Interestingly, hard x-ray photoemission spectroscopy (HXPES) measurements have revealed intermediate valence of Yb$^{+2.75}[6]$, providing the first unique example of quantum criticality in a mixed valent system. Whether the valence fluctuation is relevant for the mechanism of the quantum criticality and superconductivity is an interesting open question.

In this paper, we present the results of the specific heat and magnetization measurements of $\beta$-YbAlB$_4$ and $\alpha$-YbAlB$_4$. Both phases are locally isostructural with different arrangement of distorted hexagons made of Yb atoms (space group: $Cmmm(\beta$-YbAlB$_4$), $Pbam(\alpha$-YbAlB$_4$)) [7]. $\alpha$-YbAlB$_4$ also has an intermediate valence of Yb$^{+2.73}[6]$. The results indicate a Fermi liquid ground state for $\alpha$-YbAlB$_4$ in contrast to the unconventional quantum criticality in...
Figure 1. Magnetic part ($f$-electron contribution) of the specific heat $C_M$ plotted as $C_M/T$ versus $T$ for both $\bar{\beta}$- (□) and $\alpha$-YbAlB$_4$ (◇) under zero field. $C_M/T$ for the $\beta$ phase shows a $\ln T$ dependence for $0.2 \, K < T < 20 \, K$. $T_0 \sim 180 \pm 10 \, K$ and $S_0 = 3.7 \pm 0.1 \, J/molK$ are determined from the fit of the results to $C_M/T = S_0/T_0 \ln (T_0/T)$. The upturn in the lowest $T$ may contain a nuclear contribution. $C/T$ of $\alpha$-LuAlB$_4$ is also shown (◇).

$\beta$-YbAlB$_4$. The temperature dependence of the specific heat and susceptibility indicates that both systems have large valence fluctuation scale of $\sim 200 \, K$ and a small renormalized Kondo lattice scale of $\sim 8 \, K$ regardless of its ground state. Large Wilson ratios found in both systems indicate the importance of ferromagnetic intersite coupling for the emergence of the small Kondo lattice scale in the mixed valent compounds.

2. Experimental

High purity single crystals of $\beta$-YbAlB$_4$ were grown by a flux method [7]. Energy dispersive x-ray analysis found no impurity phases, no inhomogeneities and a ratio Yb:Al of 1:1. Surface impurities were carefully removed with dilute nitric acid before measurements.

The magnetization $M$ was measured by a commercial SQUID magnetometer using pure single crystals of 2.0 mg. The typical residual resistivity ratio (RRR) of these samples is 140.

The specific heat $C$ was measured by a relaxation method. For the measurements above (below) 0.4 K, high-purity single crystals of 0.8 mg (2.2 mg) with RRR > 200 (RRR > 140) were measured. The magnetic part of the specific heat $C_M$ was obtained by subtracting the specific heat of $\alpha$-LuAlB$_4$, which is the non-magnetic isostructural counterpart of $\beta$-YbAlB$_4$.

3. Results and discussion

First, we present the magnetic part of the specific heat $C_M$ devided by temperature in Fig. 1. In both $\beta$- and $\alpha$-YbAlB$_4$, $C_M/T$ values are strongly enhanced to be larger than 130 mJ/molK$^2$ in
the low $T$ limit, which is two orders magnitude larger than the band calculation estimates ($\approx 6\text{ mJ/molK}^2$) [10, 11]. While clear $\ln T$ divergent behavior is observed in $\beta$-YbAlB$_4$ in the temperature range of 0.2 K $< T < 20$ K, $C_M/T$ of $\alpha$-YbAlB$_4$ nearly saturates in the low temperatures below 1 K, indicating a Fermi liquid ground state. On the other hand, at higher temperatures above 10 K, $C_M/T$ of $\alpha$-YbAlB$_4$ merges to the one of $\beta$-YbAlB$_4$. Fitting the $\ln T$ behavior of $\beta$-YbAlB$_4$ to $C_M/T = S_0/T_0 \ln (T_0/T)$ yields $T_0 = 180 \pm 10$ K and $S_0 = 3.7 \pm 0.1$ J/molK. Here, $T_0$ sets a characteristic hybridization scale for the system and is close to the coherence temperature 250 K at the peak of the resistivity [3]. Note that $T_0$ in $\beta$-YbAlB$_4$ is much higher than other quantum critical Kondo lattice systems such as CeCu$_{12.9}$Au$_{0.1}$ ($T_0 = 6.2$ K) [8] and YbRh$_2$(Si$_1-x$Ge$_x$)$_2$ ($T_0 = 24$ K) [2, 9]. This is consistent with the intermediate valence in $\beta$-YbAlB$_4$ because mixed-valent compounds are typically characterized by a much higher value of $T_0$ than Kondo lattice systems. Interestingly however, if we plot $C_M/T$ scaled by $T_0$ vs. $T/T_0$, the $-\ln T$ dependence in these three quantum critical materials collapses on top of each other, indicating common meaning of $T_0$ as the $T$ scale below which $\sim 70$ % of the ground doublet entropy, $R \ln 2$, is released.

The temperature dependences of the d.c. magnetic susceptibility $\chi = M/H$ are shown in Fig. 3. Both systems exhibit strong Ising anisotropy with moments aligned along the $c$ axis, and almost $T$ independent $\chi$ along the $ab$ plane. Broad peaks found around 200 K in $\chi_{ab}$ for both systems are close to the $T_0$ scale obtained from $C_M$ and the coherence temperature of the resistivity. $\chi$ along the $c$ axis for these two systems shows almost the same temperature dependence for both $\alpha$ and $\beta$ phases down to $T \approx 8$ K, where it exhibits a shoulder. Below $T \lesssim 8$ K, on the other hand, these two systems show contrasting behavior i.e. while $\alpha$-YbAlB$_4$ shows saturating behavior, consistent with the Fermi liquid ground state, $\beta$-YbAlB$_4$ continues to diverge due to the quantum criticality [5]. The Curie-Weiss behavior, $\chi_c = C/(T + \Theta_W)$, is observed in the $T$ range larger than 150 K with $\Theta_W = 110 \pm 2$, 108 $\pm 5$ K for $\alpha$ and $\beta$ phase,
respectively (Fig. 4). Ising moments $I_z = 2.22, 2.24 \mu_B$ for $\alpha$ and $\beta$ phases are deduced from the Curie constant $C = N_A I_z^2/k_B$ where $N_A$ and $k_B$ are Avogadro and Boltzmann constant, respectively. Furthermore, at the lower temperatures below 20 K, another Curie-Weiss behavior is observed (Fig. 4 inset). If we fit the data in $T$ range of $6 \lesssim T \lesssim 15$ K, $\Theta_W = 29, 25$ K and $I_z = 1.4, 1.3 \mu_B$ are obtained for the $\alpha$ and $\beta$ phases, respectively.

These observations indicate an existence of local moments far below $T_0 \sim 200$ K at least down to $\sim 8$ K. This Kondo lattice behavior with a low temperature scale $T^* \sim 8$ K is striking compared with ordinary valence fluctuating materials, such as YbAl$_3$ [12], where Pauli paramagnetism is normally expected. A possible origin of this behavior may lie in the presence of ferromagnetic (FM) interactions between Yb 4$f$-electron spins. Indeed, the Wilson ratio $R_W = (\pi^2 k_B^2/\mu_0 I_z^2)(\chi/\gamma) \sim 7$ is obtained for both $\alpha$ and $\beta$ phases by using $\chi_c$ at $B = 0.1$ T, $T = 0.4$ K, $\gamma = C_M/T$ at $B = 0$, $T = 0.4$ K and $I_z$ obtained from the high temperature Curie-Weiss fit. The $R_W$ values are considerably large compared with the normal value 2 expected for the Kondo lattice. If we use $I_z$ obtained from the low temperature Curie-Weiss fit, then the Wilson ratio becomes $R_W \sim 25$ for the both systems. This significantly high value strongly suggests the existence of FM correlations. The observations of the ESR signal [13] is also consistent with the presence of FM correlations [14]. Such FM interactions are known to give rise to Kondo resonance narrowing [15] in d-electron systems where Hund's coupling causes a large downward renormalization of the Kondo temperature [15]. In our systems, however, the FM interactions driving the narrowing presumably derive from intersite RKKY coupling. Neutron scattering measurements and studies of Lu dilution effect in Yb$_1$-$_x$Lu$_x$AlB$_4$ systems are important future works to clarify the origin of the ferromagnetic intersite coupling between Yb 4$f$-electrons and the low temperature scale of $T^* \sim 8$ K in the mixed valence compounds.

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