Anisotropic heavy Fermi-liquid formation in the valence fluctuating $\alpha$-YbAlB$_4$

Yosuke Matsumoto,$^{1,\star}$ K. Kuga,$^1$ T. Tomita,$^1$ Y. Karaki,$^{1,2}$ and S. Nakatsuji$^{3,\dagger}$

$^1$Institute for Solid State Physics, University of Tokyo, Kashiwa, Chiba 277-8581, Japan
$^2$Faculty of Education, University of the Ryukyus, Nishihara, Okinawa 903-0213, Japan
(Dated: January 20, 2013)

$\alpha$-YbAlB$_4$ is the locally isostuctural polymorph of $\beta$-YbAlB$_4$, the first example of an Yb-based heavy fermion superconductor which exhibits pronounced non-Fermi-liquid behavior above $T_c$. Interestingly, both $\alpha$-YbAlB$_4$ and $\beta$-YbAlB$_4$ have strongly intermediate valence. Our single crystal study of the specific heat, magnetization and resistivity has confirmed the Fermi liquid ground state of $\alpha$-YbAlB$_4$ in contrast with the quantum criticality observed in $\beta$-YbAlB$_4$. Both systems exhibit Kondo lattice behavior with the characteristic temperature scale $T^* \sim 8$ K in addition to a valence fluctuation scale $\sim 200$ K. Below $T^*$, $\alpha$-YbAlB$_4$ forms a heavy Fermi liquid state with an electronic specific heat coefficient $\gamma \sim 130$ mJ/mol K$^2$ and a large Wilson ratio more than 7, which indicates ferromagnetic correlation between Yb moments. A large anisotropy in the resistivity suggests that the hybridization between 4f and conduction electrons is much stronger than along the c-axis. The strongly anisotropic hybridization as well as the large Wilson ratio is the key to understand the unusual Kondo lattice behavior and heavy fermion formation in mixed valent compounds.

PACS numbers: 71.27.+a, 71.28.+d, 75.20.Hr, 75.30.Mb

I. INTRODUCTION

4f based heavy fermion (HF) systems have attracted much attention with interesting phenomena such as unconventional superconductivity and non-Fermi-liquid (NFL) behavior found in the vicinity of quantum critical points.$^{1-2}$ Our recent studies have found the first Yb (4f$^{13}$) based HF superconductivity with the transition temperature $T_c = 80$ mK in the new compound $\beta$-YbAlB$_4$.$^7$ Pronounced NFL 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.$^2$ Furthermore, the $T/B$ scaling found in our recent high-precision magnetization measurements clarifies its unconventional zero-field quantum criticality without tuning, which can not be explained by the standard theory based on spin-density-wave fluctuations.$^{10-12}$ In contrast to the canonical quantum critical materials, hard X-ray photoemission spectroscopy (HXPES) measurements have revealed strongly intermediate valence of Yb$^{2.75}$, providing the only example of quantum criticality in a mixed valent system.$^{13}$ 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, magnetization and resistivity measurements of $\alpha$-YbAlB$_4$, the locally isostuctural polymorph of $\beta$-YbAlB$_4$ with different arrangement of distorted hexagons made of Yb atoms (space group: $Pbam(\alpha$-YbAlB$_4$), $Cmmm(\beta$-YbAlB$_4$))$^{14-15}$. According to the HX-PES measurement,$^{13}$ $\alpha$-YbAlB$_4$ also has an intermediate valence of Yb$^{2.73}$. The results indicate a Fermi liquid (FL) ground state for $\alpha$-YbAlB$_4$ in contrast to the unconventional quantum criticality observed in $\beta$-YbAlB$_4$. Interestingly, both systems exhibit Kondo lattice behavior with a small renormalized temperature scale of $T^* \sim 8$ K although both of them have a large valence fluctuation scale of $\sim 200$ K. Below $T^*$, $\alpha$-YbAlB$_4$ forms a heavy Fermi liquid state with an electronic specific heat coefficient $\gamma \sim 130$ mJ/mol K$^2$ and a large Wilson ratio more than 7, which indicates ferromagnetic correlation between Yb moments. Kadowaki-Woods ratio is found similar to those found in the normal Kondo lattice systems and considerably larger than mixed valent systems. Furthermore, a large anisotropy observed in the resistivities of $\alpha$-YbAlB$_4$ suggests strongly anisotropic hybridization between 4f and conduction electrons. This strong anisotropy in the hybridization is the key to understand the mechanism of the heavy fermion formation as well as the Kondo lattice behavior found in the intermediate valence system. Partial information has already been discussed in Ref.$^{15}$

II. EXPERIMENTAL

High purity single crystals of $\alpha$-YbAlB$_4$ were grown by a flux method.$^{12}$ Energy dispersive X-ray (EDX) and inductive coupled plasma (ICP) analyses 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. We succeeded in growing pure crystals with residual resistivity ratio (RRR) up to 110. The magnetization $M$ at $T > 2$ K was measured by a commercial SQUID magnetometer using pure single crystals (RRR $\sim 50$) of 2.4 mg. The magnetization data at $T < 4$ K and $B < 0.05$ T were obtained by using a high precision SQUID magnetometer installed in a $^3$He-$^4$He dilution refrigerator.$^2$ The specific heat $C$ of pure single crystals (1.1 mg, RRR $\sim 50$) was measured at temperature range $0.4 < T < 200$ K by a relaxation method.
Four-terminal resistivity measurements were made by using a DC method (300 K ⩾ T ⩾ 0.5 K) and an AC method (1.4 K ⩽ T ⩽ 35 mK).

III. RESULTS AND DISCUSSION

First, we present the magnetic part of the specific heat $C_m$ divided by temperature in Fig. 1 (a). $C_m$ was obtained by subtracting the specific heat of $\alpha$-LuAlB$_4$ shown in the same figure. Here, $\alpha$-LuAlB$_4$ is the nonmagnetic isostructural counterpart of $\alpha$-YbAlB$_4$. The Debye temperature of $\alpha$-LuAlB$_4$ is estimated to be 380 K from the $T^3$ dependence of $C$ below 10 K. In both $\alpha$- and $\beta$-YbAlB$_4$, $C_m/T$ is strongly enhanced to be ⩾ 130 mJ/molK$^2$ in the low $T$ limit, which is large compared to ordinary valence fluctuating materials, such as CeSn$_2$ and YbAl$_2$ (see Fig. 1 (a)) and is two orders magnitude larger than the band calculation estimates ($\sim$ 6 mJ/molK$^2$).\textsuperscript{19,20} While clear lnT divergent behavior is observed in $\beta$-YbAlB$_4$ in the temperature range of 0.2 K < T < 20 K, $C_m/T$ in $\alpha$-YbAlB$_4$ nearly saturates at $T < 1$ K, indicating a Fermi liquid ground state. On the other hand, at higher temperatures above 10 K, $C_m/T$ in $\alpha$-YbAlB$_4$ merges to the lnT behavior of $\beta$-YbAlB$_4$. Fitting the lnT 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 for $\beta$-YbAlB$_4$. Here, $T_0$ provides a characteristic hybridization scale for the system and is close to the coherence temperature of 250 K set by the resistivity peaks.\textsuperscript{7} Another rough estimate of $T_0$ can be made using the temperature where the magnetic part of the entropy $S_m$ reaches $R \ln 2$ (the entropy of a ground state doublet). In this way, $T_0$ for $\alpha$-YbAlB$_4$ can be estimated to be $T_0 \sim 160 \pm 20$ K, as shown in Fig. 1 (b). In order to obtain $S_m$, we assume a constant value of $C_m/T$ (127 mJ/molK$^2$) below the lowest temperature of the measurements 0.4 K. These large values of $T_0$ are consistent with the intermediate valence of these systems because mixed-valent compounds are typically characterized by a much higher value of $T_0$ than Kondo lattice systems.\textsuperscript{21,22} A proposed crystalline electric field (CEF) level scheme, which reproduces the magnetic susceptibility, suggests a CEF level splitting of $\Delta = 80$ K.\textsuperscript{19} However, a Schottky peak of this level splitting which would appear at $\sim 25$ K with a height of 130 mJ/K$^2$mol is not seen here. This is probably because the CEF levels are smeared out by the valence fluctuations.

The temperature dependence of the d.c. magnetic susceptibility $\chi = M/B$ are shown in Fig. 1(c). Both systems exhibit strong Ising anisotropy with the strongly $T$ dependent $c$-axis $\chi$ and almost $T$ independent $\chi$ along the $ab$-plane.\textsuperscript{15,18} Broad peaks found around 200 K in $\chi_{ab}$ for both systems (Fig. 2) are close to the $T_0$ scale obtained from $C_m$ and the coherence temperature of the resistivity which we will discuss later. The $c$-axis component for both systems shows almost the same temperature dependence down to $T \sim 8$ K. Below $T \lesssim 8$ K, on the other hand, these two systems show contrasting behavior: while $\beta$-YbAlB$_4$ continues to diverge due to the quantum criticality,\textsuperscript{9} $\alpha$-YbAlB$_4$ shows saturating behavior, indicating the Fermi liquid formation. The Curie-Weiss behavior, $\chi_c = C/(T+\Theta_W)$, is observed at $T > 150$
K with $\Theta_W = 110 \pm 2$, 108 $\pm$ 5 K for $\alpha$ and $\beta$ phases, respectively (Fig. 2). 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 constants, respectively. Furthermore, at $T < 20$ K, another Curie-Weiss behavior is observed (Fig. 2 inset).

If we fit the data to the Curie-Weiss law at 6 $\leq T \leq$ 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 suggest existence of local moments far below $T_D \sim 200$ K possibly 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 where Pauli paramagnetism is normally expected, such as CeSn$_2^{23}$ and YbAlB$_4^{18}$ (see Fig. 1 (c)). One of the possible origins of this behavior may lie in Kondo resonance narrowing due to the presence of ferromagnetic (FM) interactions between Yb 4f-electron spins where FM interactions cause a large downward renormalization of the Kondo temperature from $T_0 \sim 200$ K to $T^* \sim$ 8 K.$^{22}$ 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 Kondo lattice systems. If we use $I_z$ obtained from the low temperature Curie-Weiss fit, the Wilson ratio becomes $R_W \sim 25$ for both systems, these significantly high values can be regarded as a consequence of the FM correlations.

Alternatively, the large $R_W$ values might also be explained by the possible proximity to a valence quantum criticality as it have been recently pointed out by Watanabe and Miyake$^{25}$. In this case, the low temperature scale $T^* \sim$ 8 K might arise from the characteristic energy scale for the valence fluctuations and not from the Kondo resonance narrowing. So far, we do not have experimental evidence to uniquely specify the mechanism among the possible scenarios. Further studies are required to solve this issue.

The temperature dependence of the in-plane resistivity with current along [-110] direction, which we denote $\rho_{ab}$, and c-axis resistivity $\rho_c$ are shown in Fig. 3. We have also measured the $\alpha$-axis resistivity $\rho_{\alpha}$ and have found no significant difference from $\rho_{ab}$. This is consistent with the the recent band calculation$^{22}$, which predicts a nearly isotropic transport within the plane. Further investigation of the in-plane anisotropy including the $b$-axis resistivity $\rho_b$ is now underway. Note that $\rho_c$ in $\beta$-YbAlB$_4$ is not available so far due to the tiny thickness of $\sim$ 10 $\mu$m along the c-axis of single crystals. The magnetic part of the resistivity $\rho_m$ is obtained by subtracting the non-magnetic contribution estimated by $\rho_{ab}$ of $\alpha$-, $\beta$-LuAlB$_4$ (solid and dash dotted lines, respectively) or $\rho_c$ of $\alpha$-LuAlB$_4$ (dashed line). Inset shows low T part of $\rho_{ab}$ and $\rho_c$. 

![FIG. 2: Temperature dependence of the inverse susceptibility $\chi^{-1} = B/M$ under the field along the the ab-plane and c-axis. Solid and broken lines are Curie-Weiss fits above 150 K for $\alpha$- (open circles) and $\beta$-YbAlB$_4$ (open squares), respectively. Inset shows the low temperature part of $\chi^{-1}$ under the field along the the c-axis.](image1)

![FIG. 3: Temperature dependence of the in-plane and c-axis resistivity $\rho_{ab}$ and $\rho_c$ of $\alpha$-YbAlB$_4$ and $\rho_{ab}$ of $\beta$-YbAlB$_4$. The magnetic part of the resistivity $\rho_m$ is obtained by subtracting the non-magnetic contribution estimated by $\rho_{ab}$ of $\alpha$-, $\beta$-LuAlB$_4$ (solid and dash dotted lines, respectively) or $\rho_c$ of $\alpha$-LuAlB$_4$ (dashed line). Inset shows low T part of $\rho_{ab}$ and $\rho_c$. Alternatively, the large $R_W$ values might also be explained by the possible proximity to a valence quantum criticality as it have been recently pointed out by Watanabe and Miyake$^{25}$. In this case, the low temperature scale $T^* \sim$ 8 K might arise from the characteristic energy scale for the valence fluctuations and not from the Kondo resonance narrowing. So far, we do not have experimental evidence to uniquely specify the mechanism among the possible scenarios. Further studies are required to solve this issue.](image2)
be considered as the coherence peak providing the characteristic hybridization temperature scale. On the other hand, $\rho_{c}^{(c)}$ in $\alpha$-YbAlB$_4$ decreases monotonously on cooling below 300 K.

Interestingly, $\rho_{c}$ is much smaller than $\rho_{ab}$ in $\alpha$-YbAlB$_4$, i.e. the conductivity of the system exhibits quasi-1D anisotropy. The ratio, $\rho_{ab}/\rho_{c}$, increases at low temperatures making a peak at $T \sim 6$ K (Fig. 4 (a), solid black line). At the peak, $\rho_{ab}/\rho_{c}$ reaches $\sim 13$ and approaches a constant value of $\sim 11$ in lowest temperatures. This low temperature anisotropy is one order of magnitude larger than typical anisotropic heavy fermion systems such as CeCoIn$_5$.$^{27}$, CeCu$_2$Si$_2$.$^{28}$, CeNiIn$_2$.YbAgGe.$^{22}$ where the ratio is almost $T$ independent and $\lesssim 3$ below 300 K. On the other hand, $\rho_{ab}/\rho_{c}$ in $\alpha$-LuAlB$_4$ is nearly temperature independent with a slight increase from 3.5 at 300 K to 4.8 at lowest temperatures (Fig. 4 (a), dotted broken gray line). This temperature independent anisotropy in $\alpha$-LuAlB$_4$ should come from the anisotropy of the Fermi surface. Interestingly, at $T \sim 300$ K, $\rho_{ab}/\rho_{c}$ in $\alpha$-YbAlB$_4$ approaches to a similar value to the one in $\alpha$-LuAlB$_4$ although $f$ electron contribution is dominant in $\alpha$-YbAlB$_4$. This suggests that the topology of the Fermi surfaces of these systems are similar to each other at high $T > T_0$.

The peak found in $\rho_{ab}/\rho_{c}$ arises mainly from a rapid decrease in $\rho_{ab}$ below $T \sim 10$ K (Fig. 3 inset). This can be clearly seen in the temperature derivative of $\rho$, $d\rho/dT$ shown in Fig. 4 (b). While $d\rho_{c}/dT$ is small and shows a weak $T$ dependence, $d\rho_{ab}/dT$ exhibits a rapid increase below $T \sim 10$ K close to the low temperature scale of Kondo lattice behavior, $T^* \sim 8$ K. This suggests that further coherence develops around $f$ electrons due to the formation of heavy quasi particles below this temperature. The absence of similar increase in $d\rho_{c}/dT$ and large $\rho_{ab}/\rho_{c}$ suggest that the associated heavy fermions are only mobile within the $ab$-plane, but not along the $c$-axis. This is consistent with the recent band calculation which found that the dispersion along the $ab$-plane is narrow due to the $4f$ electron contribution in comparison with the one along the $c$-axis for many of the bands mostly coming from conduction electrons.$^{26}$ We find the anomalies in $d\rho/dT$ at 40-50 K around the same temperature scale as for the reflection points in $\chi$ (Fig. 4 (b)) where $\chi$ starts to show further increase on cooling. This temperature scale can be regarded as the onset temperature of the Kondo lattice behavior.

A possible explanation for the large anisotropy would be the anisotropic hybridization between the conduction and $f$ electrons i.e. the smaller hybridization along the $c$-axis. In this case, while $T_0 \sim 200$ K has its origin in the in-plane hybridization, the hybridization scale along the $c$-axis should be smaller. This may also explain why the coherence peak is observed only in $\rho_{ab}$. Indeed, the recent band calculation suggests the smaller hybridization along the $c$-axis in $\beta$-YbAlB$_4$.$^{26}$ Although the lower symmetry in $\alpha$-YbAlB$_4$ makes its band structure more complex, the general features such as anisotropic hybridization are expected to be similar to each other.

In addition, according to a recent theory on the electronic structure, a hybridization node is expected along the $c$-axis.$^{19,31}$ The evolution of $\rho_{ab}$-$\rho_{c}$ such as the $\sim 7$ K data of $\rho_{ab}$-$\rho_{c}$ shows a similar trend for the $f$ electron contribution in $\alpha$-YbAlB$_4$. This suggests that the topology of the Fermi surfaces of these systems are similar to each other at high $T > T_0$.

![Figure 4](image_url)

**FIG. 4:** (a) Temperature dependence of the ratio $\rho_{ab}/\rho_{c}$ and $\Delta \rho_{ab}/\Delta \rho_{c}$. Here $\Delta \rho$ is defined by $\Delta \rho \equiv \rho - \rho_0$ (see text). (b) Temperature derivative of the resistivity $d\rho/dT$. The ratio, $\rho_{ab}/\rho_{c}$, increases at low temperatures making a peak at $T \sim 6$ K (Fig. 4 (a), solid black line). At the peak, $\rho_{ab}/\rho_{c}$ reaches $\sim 13$ and approaches a constant value of $\sim 11$ in the lowest temperatures. This low temperature anisotropy is one order of magnitude larger than typical anisotropic heavy fermion systems such as CeCoIn$_5$,$^{27}$ CeCu$_2$Si$_2$,$^{28}$ CeNiIn$_2$.YbAgGe,$^{22}$ where the ratio is almost $T$ independent and $\lesssim 3$ below 300 K. On the other hand, $\rho_{ab}/\rho_{c}$ in $\alpha$-LuAlB$_4$ is nearly temperature independent with a slight increase from 3.5 at 300 K to 4.8 at lowest temperatures (Fig. 4 (a), dotted broken gray line). This temperature independent anisotropy in $\alpha$-LuAlB$_4$ should come from the anisotropy of the Fermi surface. Interestingly, at $T \sim 300$ K, $\rho_{ab}/\rho_{c}$ in $\alpha$-YbAlB$_4$ approaches to a similar value to the one in $\alpha$-LuAlB$_4$ although $f$ electron contribution is dominant in $\alpha$-YbAlB$_4$. This suggests that the topology of the Fermi surfaces of these systems are similar to each other at high $T > T_0$.
ρ shows ∆T

FIG. 5: (a) Temperature dependence of ∆ρversus T². Note that ∆ρc for α-YbAlB₄ is multiplied by a factor of 13 for clarity. The arrow indicates T_F = 240 mK estimated for ρc of α-YbAlB₄ by using the resistivity exponent α(see text). (b) The resistivity exponent α defined by ∆ρ = AT²(see text).

The temperature dependent parts of the resistivity ∆ρ ≡ ρ − ρ₀ at T < 1 K are shown in Fig. 5(a). Here ρ₀ is the zero temperature limit of the resistivity, which was estimated by a power law fit of the low T data down to 35 mK (the detail is discussed later). ρ₀ are 9.4, 0.82 µΩcm for ρab, ρc of α-YbAlB₄, respectively (RRR ~ 20 and 70) and 0.49 µΩcm for ρab of β-YbAlB₄ (RRR ~ 250). The anisotropy in ρ₀, which corresponds to ρab/ρc ~ 11 in the lowest T, is almost the same as that of ∆ρ (∆ρab/∆ρc), which is as large as 13 in the low T limit (Fig. 5(a)). ∆ρab of β-YbAlB₄ takes a value between ∆ρab and ∆ρc of the α-phase. On the other hand, if we compare ∆ρ/ρ₀, β-YbAlB₄ exhibits much larger value than those in α-YbAlB₄. For instance, ∆ρab/ρ₀ = 0.85 at T = 1 K in β-YbAlB₄ is ~ 10 times larger than the respective ∆ρ/ρ₀ = 0.11(ρab) and 0.08(ρc) for α-YbAlB₄. This cannot be explained only by the better sample quality in β-YbAlB₄, and thus the quantum criticality in β-YbAlB₄ should also be responsible for the enhancement. Indeed, the application of the magnetic field, suppressing the criticality, decreases ∆ρ/ρ₀ of β-YbAlB₄ to the same order as that in α-YbAlB₄. Note that even a α-YbAlB₄ sample with the highest RRR ~ 110 (estimated by ρc) does not exhibit superconductivity down to 35 mK (not shown).

To demonstrate the difference in the ground state of α- and β-YbAlB₄, we show the temperature dependence of the power law exponent α defined by ∆ρ = ρ₀ + A’T² (Fig. 5(b)). α is obtained by using the equation α = d log ∆ρ/d log T. ρ₀ was determined using the best fitting result to the above equation that indicates the corresponding power law behavior in the widest temperature range from the lowest temperature. α is strongly dependent on ρ₀, and its error due to 0.01% change in ρ₀ are shown in Fig. 5(b). While the exponent α in β-YbAlB₄ is small < 1.5 at the low temperatures, those in α-YbAlB₄ are much larger and approaches the normal value of 2 expected for FL on cooling. This can be also confirmed in the plot against T² (inset of Fig. 5(a)), where ρc(T) of α-YbAlB₄ shows a linear dependence on T² below T_FL ~240 mK. The observation of α ~ 2 in the lowest temperatures in addition to almost saturating c and C_m/T below T* ~ 8 K indicates that the ground state of α-YbAlB₄ is a Fermi liquid.

The T²-coefficient A defined by ∆ρ = ρ₀ + AT² was estimated by the linear fit in the inset of Fig. 5(a) below 240 mK. The obtained A values are 0.094 and 1.27 µΩ cm / K² for ρc and ρab, respectively. Kadowaki-Woods ratio A/γ² estimated by using these anisotropic A values are 5.8 × 10⁻⁶ and 7.8 × 10⁻⁵ µΩ cm(K mol/mJ)² for ρc and ρab, respectively. Here γ is a low temperature limit of C/T, and in the present case, the value at 0.4 K (127 mJ/mol K²) was used. It is known that, the ratio A/γ² is close to 1.0 × 10⁻⁵ µΩ cm(K mol/mJ)² in many heavy fermion compounds of Kondo lattice systems. On the other hand, Tsujii et al. have suggested that the ratio is considerably smaller in intermediate valence systems, or equivalently, the systems with large orbital degeneracy N i.e. the system with a large hybridization scale T₀ compared to CEF splitting ΔCEF. In this case, the expected ratio is close to the typical value known for transition metals A/γ² = 0.4 × 10⁻⁶ µΩ cm(K mol/mJ)², which is 25 times smaller than the above ratio for heavy fermions. To illustrate this, we show in Fig. 6 a full logarithmic plot of A versus γ (Kadowaki-Woods plot) for representative Ce and Yb based 4f electron systems. A/γ² for most of the mixed valent materials or materials with large N in Ce systems: N = 6 and in Yb systems: N = 8) is much smaller than the original Kadowaki-Woods ratio and has value of the order of 10⁻⁷ µΩ cm(K mol/mJ)². Compared to these small values observed in mixed valence materials, the ratio obtained for ρc and ρab of α-YbAlB₄ is much larger and close to the typical value for heavy fermions. In β-YbAlB₄, the ratio for ρab also takes a similar value of 4.4 × 10⁻⁵ µΩ cm(K mol/mJ)² in magnetic field of 2 T along the c-axis. The large A/γ² in α-YbAlB₄ and β-YbAlB₄ (B = 2 T ||
c-axis) indicate that the system behaves like Kondo lattices at low temperatures rather than mixed valent materials. Interestingly, the ratio obtained for $\rho_{ab}$ in both $\alpha$- and $\beta$-YbAlB$_4$ is several times larger than the typical value for Kondo lattice systems. This deviation may come from material dependent properties such as dimensionality and carrier concentration$^{35}$. Further analyses based on fermiology is required to clarify the origin of the enhancement in the Kadowaki-Woods ratio.

IV. CONCLUSION

Our detailed measurements have confirmed that both $\alpha$-YbAlB$_4$ and $\beta$-YbAlB$_4$ exhibit Kondo lattice behavior with a small renormalized temperature scale of $T^* \sim 8$ K in addition to a large valence fluctuation scale of $\sim 200$ K. Below $T^* \sim 8$ K, $\alpha$-YbAlB$_4$ forms a heavy Fermi liquid state with $\gamma \sim 130$ mJ/mol K$^2$ in contrast to the unconventional quantum criticality observed in $\beta$-YbAlB$_4$. The Kadowaki-Woods ratio takes a typical value for Kondo lattice systems and considerably larger than those for mixed valent systems. This is consistent with the Kondo lattice behavior found in the temperature dependence of the susceptibility and specific heat. The large Wilson ratio more than 7 suggests that a ferromagnetic intersite coupling between Yb 4f-electrons and/or proximity to a valence quantum criticality, may be the origin of the Kondo lattice behavior. Furthermore, the large anisotropy observed in the resistivities suggests that hybridization between 4f and conduction electrons is much stronger within the ab-plane than along the c-axis. This strongly anisotropic hybridization and the large Wilson ratio are the keys to understand the unusual Kondo lattice behavior and heavy fermion formation in these mixed valence compounds. The future works including neutron scattering measurements and studies of Lu dilution effect in Yb$_{1-x}$Lu$_x$AlB$_4$ systems are necessary to clarify the origin of these unusual behaviors.

Acknowledgments

We thank N. Horie, E. C. T. O’Farrell, C. Petrovic, P. Coleman, A. H. Nevidomskyy, H. Harima, S. Watanabe, C. Broholm, K. Ueda and T. Sakakibara for supports and useful discussions. This work is partially supported by Grants-in-Aid (No. 21684019) from JSPS, by Grants-in-Aids for Scientific Research on Innovative Areas (No. 20102007, No. 21102507) from MEXT, Japan, by Global COE Program “the Physical Sciences Frontier”, MEXT, Japan, by Toray Science and Technology Grant.

1 Present address: Department of Physics, College of Humanities and Sciences, Nihon University, Sakurajosui, Setagaya-ku, Tokyo 156-8550, Japan

2 G. R. Stewart, Rev. Mod. Phys., 73, 797 (2001).

3 H. Q. Yuan, F. M. Grosche, M. Deppe, C. Geibel, G. Sparn, and F. Steglich, Science, 302, 2104 (2003).

4 H. v. Löhneysen, A. Rosch, M. Voit, and P. Wölfle, Rev. Mod. Phys., 79, 1015 (2007).

5 P. Monthoux, D. Pines, and G. G. Lonzarich, Nature, 450, 1177 (2007).

6 P. Gegenwart, Q. Si, and F. Steglich, Nature Phys., 4, 186 (2008).

7 S. Nakatsuji, K. Kuga, Y. Machida, T. Tayama, T. Sakak-
ibara, Y. Karaki, H. Ishimoto, S. Yonezawa, Y. Maeno, E. Pearson, G. G. Lonzarich, L. Balicas, H. Lee, and Z. Fisk, Nature Phys., 4, 603 (2008).
8 K. Kuga, Y. Karaki, Y. Matsumoto, Y. Machida, and S. Nakatsuji, Phys. Rev. Lett., 101, 137004 (2008).
9 Y. Matsumoto, S. Nakatsuji, K. Kuga, Y. Karaki, N. Horie, Y. Shimura, T. Sakakibara, A. H. Nevidomskyy, and P. Coleman, Science, 331, 316 (2011).
10 J. A. Hertz, Phys. Rev. B, 14, 1165 (1976).
11 T. Moriya, Spin Fluctuations in ItinerantElectron Magnetism (Springer, Berlin, 1985).
12 A. J. Millis, Phys. Rev. B, 48, 7183 (1993).
13 M. Okawa et al., Phys. Rev. Lett., 104, 247201 (2010).
14 Z. Fisk, K. N. Yang, M. B. Maple, and H. R. Ott, Valence Fluctuations in Solids (North-Holland, New York, 1981) pp. 345–347.
15 R. T. Macaluso, S. Nakatsuji, K. Kuga, E. L. Thomas, Y. Machida, Y. Maeno, Z. Fisk, and J. Y. Chan, Chem. Mater., 19, 1918 (2007).
16 Y. Matsumoto, K. Kuga, N. Horie, and S. Nakatsuji, J. Phys.: Conf. Ser., 273, 012006 (2011).
17 S. H. Liu, C. Stassis, and J. K. A. Gschneidner, Valence Fluctuations in Solids, edited by L. M. Falicov, W. Hanke, and M. B. Maple (North-Holland, Amsterdam, 1981) p. 99.
18 A. L. Cornelius et al., Phys. Rev. Lett., 88, 117201 (2002).
19 A. H. Nevidomskyy and P. Coleman, Phys. Rev. Lett., 102, 077202 (2009).
20 E. C. T. O’Farrell, D. A. Tompsett, S. E. Sebastian, N. Harrison, C. Capan, L. Balicas, K. Kuga, A. Matsuo, K. Kindo, M. Tokunaga, S. Nakatsuji, G. Csányi, Z. Fisk, and M. L. Sutherland, Phys. Rev. Lett., 102, 216402 (2009).
21 H. v. Löhneysen, J. Phys.: Condens. Matter, 8, 9689 (1996).
22 J. Custers et al., Nature, 424, 524 (2003).
23 T. Tsuchida and W. E. Wallace, J. Chem. Phys., 43, 3811 (1965).
24 A. H. Nevidomskyy and P. Coleman, Phys. Rev. Lett., 103, 147205 (2009).
25 S. Watanabe and K. Miyake, Phys. Rev. Lett., 105, 186403 (2010).
26 D. A. Tompsett, Z. P. Yin, G. G. Lonzarich, and W. E. Pickett, Phys. Rev. B, 82, 235101 (2010).
27 A. Malinowski, M. F. Hundley, C. Capan, F. Ronning, R. Movshovich, N. O. Moreno, J. L. Sarrao, and J. D. Thompson, Phys. Rev. B, 72, 184506 (2005).
28 Y. Ônuki, Y. Furukawa, and T. Komatsubara, J. Phys. Soc. Jpn., 53, 2197 (1984).
29 H. Fujii, T. Takabatake, and Y. Andoh, J. Alloys Compd., 181, 111 (1992).
30 P. G. Niklowitz, G. Knebel, J. Flouquet, S. L. Bud’ko, and P. C. Canfield, Phys. Rev. B, 73, 125101 (2006).
31 A. Ramires, P. Coleman, A. H. Nevidomskyy and A. M. Tsvelik, private communication.
32 N. Tsujii, H. Kontani, and K. Yoshimura, Phys. Rev. Lett., 94, 057201 (2005).
33 M. S. Torikachvili, S. Jia, E. D. Mun, S. T. Hannahs, R. C. Black, W. K. Neils, D. Martien, S. L. Bud’ko, and P. C. Canfield, Proc. Natl. Acad. Sci. U.S.A., 104, 9960 (2007).
34 T. D. Matsuda, N. D. Dung, Y. Haga, S. Ikeda, E. Yamamoto, T. Ishikura, T. Endo, T. T. R. Settai, and Y. Onuki, Phys. Status Solidi B, 247, 757 (2010).
35 M. J. Rice, Phys. Rev. Lett., 20, 1439 (1968).
36 K. Kadowaki and S. B. Woods, Solid State Commun., 58, 507 (1986).
37 N. Tsujii, K. Yoshimura, and K. Kosuge, J. Phys. Condens. Matter, 15, 1993 (2003).
38 A. C. Jacko, J. O. Fjarrestad, and B. J. Powell, Nature Phys., 5, 422 (2009).