Anisotropic heavy-Fermi-liquid formation in valence-fluctuating α-$YbAlB_4$

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α-$YbAlB_4$ is the locally isostructural polymorph of β-$YbAlB_4$, the first example of a Yb-based heavy fermion superconductor that exhibits pronounced non-Fermi-liquid behavior above T_c. Interestingly, both α-$YbAlB_4$ and β-$YbAlB_4$ have strongly intermediate valence. Our single-crystal study of the specific heat, magnetization, and resistivity has confirmed the Fermi-liquid ground state of α-$YbAlB_4$ in contrast to the quantum criticality observed in β-$YbAlB_4$. Both systems exhibit Kondo lattice behavior with the characteristic temperature scale $T^* \sim 8$ K in addition to a valence-fluctuation scale $\sim$200 K. Below T^*, α-$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 greater than 7, which indicates ferromagnetic correlation between Yb moments. A large anisotropy in the resistivity, which is one of the largest in heavy fermions, suggests that the hybridization between $4f$ and conduction electrons is much stronger in the ab plane than along the c axis, indicating this is an excellent system to study for revealing the anisotropic hybridization effects. The strongly anisotropic hybridization as well as the large Wilson ratio is the key to understanding the unusual Kondo lattice behavior and heavy-fermion formation in mixed-valence compounds.

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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.[1–6] Our recent studies have found the first Yb- ($4f^{13}$-) based HF superconductivity with the transition temperature $T_c = 80$ mK in the compound β-$YbAlB_4$.[7,8] 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.[7] Furthermore, the T/B scaling found in our recent high-precision magnetization measurements clarifies its unconventional zero-field quantum criticality without tuning,[9] which cannot 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 a strongly intermediate valence of $Yb^{+2.75}$, providing an example of quantum criticality in a mixed-valence system.[13] Whether the valence fluctuation is relevant for the mechanism of 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 α-$YbAlB_4$, the locally isostructural polymorph of β-$YbAlB_4$ with a different arrangement of distorted hexagons made of Yb atoms [space groups $Pbam$(α-$YbAlB_4$) and $Cmmm$(β-$YbAlB_4$)].[14,15] According to the HXPES measurement,[13] α-$YbAlB_4$ also has an intermediate valence of $Yb^{+2.73}$. The results indicate a Fermi-liquid (FL) ground state for α-$YbAlB_4$ in contrast to the unconventional quantum criticality observed in β-$YbAlB_4$. 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$200 K. Below T^*, α-$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 greater than 7, which indicates a ferromagnetic correlation between Yb moments. A Kadowaki-Woods ratio is found that is similar to those found in the normal Kondo lattice systems and considerably larger than mixed-valence systems. Furthermore, the resistivity of α-$YbAlB_4$ exhibits one of the strongest anisotropies in heavy fermions. This strongly suggests anisotropic hybridization between $4f$ and conduction electrons, which is the key to understanding the mechanism of heavy-fermion formation as well as the Kondo lattice behavior found in the intermediate-valence system. Thus the system should be one of the best systems to study for elucidating the effects of anisotropic hybridization. Partial information has already been discussed in Ref. 16.

II. EXPERIMENT

High-purity single crystals of α-$YbAlB_4$ were grown by a flux method.[15] Energy dispersive x-ray and induction coupled plasma analyses found no impurity phases, no inhomogeneities, and a Yb:Al ratio 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 superconducting quantum interference device (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.[9] The specific heat C of pure single crystals (1.1 mg, RRR $\sim$ 50) was measured in the temperature range $0.4 < T < 200$ K by a relaxation method using a physical property measurement system. Four-terminal resistivity measurements were made by using a dc method (300 K $\gtrsim T \gtrsim$ 0.5 K) and an ac method (1.4 K $\gtrsim T \gtrsim$ 35 mK).

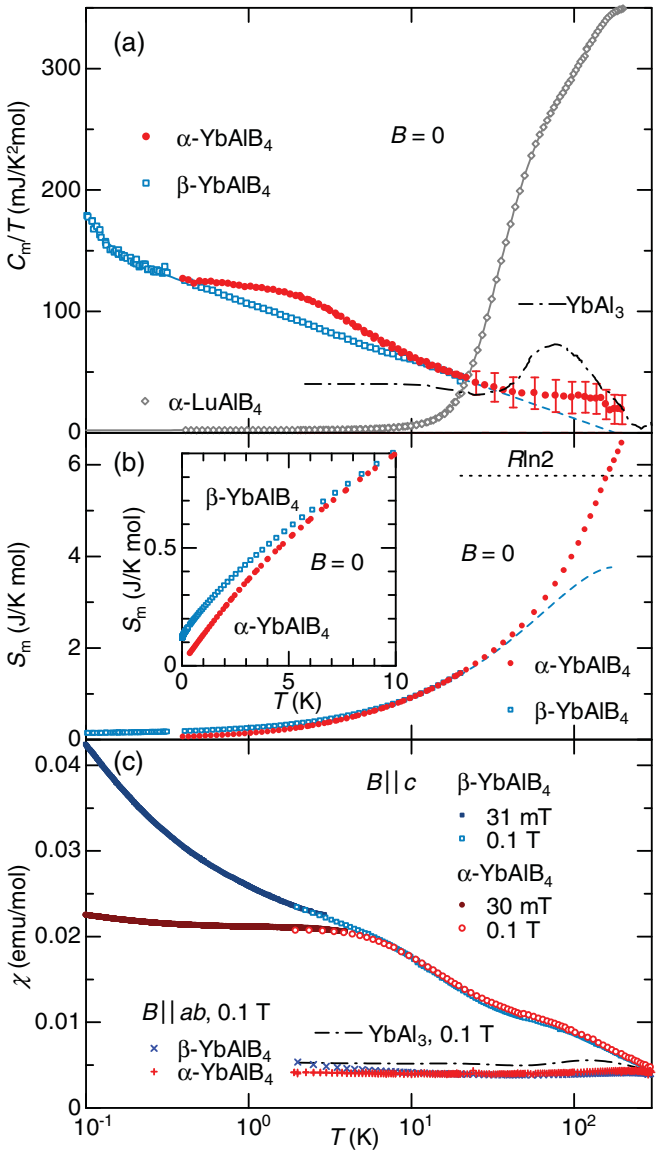

FIG. 1. (Color online) (a) Magnetic part (f-electron contribution) of the specific heat C_m plotted as C_m/T versus T for both β- and α-$YbAlB_4$ under zero field. For the β phase C_m/T shows a $\ln T$ dependence for 0.2 K $< T <$ 20 K.[9] The dash-dotted line is the fit of the results to $C_m/T = S_0/T_0 \ln(T_0/T)$ (see the text). Also shown are C/T of α-$LuAlB_4$ and C_m/T of the intermediate-valence cubic system $YbAl_3$.[18] (b) Magnetic part of the entropy S_m, which was obtained by integrating C_m/T. In α-$YbAlB_4$, a constant value of 127 mJ/K^2 mol is assumed below 0.4 K. Above 10 K, C_m/T in α-$YbAlB_4$ merges with the $\ln T$ behavior of β-$YbAlB_4$. Thus the data for β-$YbAlB_4$ shifted to take the same value as that for α-$YbAlB_4$ at 20 K. The dashed line is obtained from the $\ln T$ fitting used in (a). The inset shows the low T part. (c) Temperature dependence of the dc susceptibility $\chi = M/B$ measured in a field along the ab plane and c axis for both β- and α-$YbAlB_4$. Also shown is χ of the intermediate-valence cubic system $YbAl_3$.[18]

III. RESULTS AND DISCUSSION

We present first the magnetic part of the specific heat C_m divided by temperature in Fig. 1(a). The specific heat C_m was obtained by subtracting the specific heat of α-$LuAlB_4$ shown in the same figure. Here α-$LuAlB_4$ is the nonmagnetic isostructural counterpart of α-$YbAlB_4$. The Debye temperature of α-$LuAlB_4$ is estimated to be 380 K from the T^3 dependence of C below 10 K. In both α- and β-$YbAlB_4$, C_m/T is strongly enhanced to be $\gtrsim$ 130 mJ/mol K^2 in the low T limit, which is large compared to ordinary valence-fluctuating materials, such as $CeSn_3$ (Ref. 17) and $YbAl_3$ (Ref. 18) [see Fig. 1(a)], and is two orders magnitude larger than the band calculation estimates ($\sim$6 mJ/mol K^2).[19,20] While clear $\ln T$ divergent behavior is observed in β-$YbAlB_4$ in the temperature range 0.2 K $< T <$ 20 K, C_m/T in α-$YbAlB_4$nearly saturates at $T <$ 1 K, indicating a Fermi-liquid ground state. In contrast, at higher temperatures above 10 K, C_m/T in α-$YbAlB_4$ merges with the $\ln T$ behavior of β-$YbAlB_4$. Fitting the $\ln T$ behavior of β-$YbAlB_4$ 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/mol K for β-$YbAlB_4$.[9] 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 peak.[7] Another rough estimate of T_0 can be made by 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 α-$YbAlB_4$ 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/mol K^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-valence compounds are typically characterized by a much higher value of T_0 than Kondo lattice systems.[6,21,22] A proposed crystalline electric field (CEF) level scheme, which reproduces the magnetic susceptibility, suggests a CEF level splitting of $\Delta = 80$ K.[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 by the valence fluctuations.

The temperature dependence of the dc magnetic susceptibility $\chi = M/B$ is shown in Fig. 1(c). Both systems exhibit a strong Ising anisotropy with the strongly T-dependent c axis χ and almost-T-independent χ along the ab plane.[15] Broad peaks found around 200 K in χ_{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 β-$YbAlB_4$ continues to diverge due to the quantum criticality,[9] α-$YbAlB_4$ shows saturating behavior, indicating Fermi-liquid formation. The Curie-Weiss behavior $\chi_c = C/(T + \Theta_W)$ is observed at $T >$ 150 K with $\Theta_W = 110 \pm 2$ and 108 ± 5 K for the α and β phases, respectively (Fig. 2). Ising moments $I_z = 2.22\,\mu_B$ and $2.24\,\mu_B$ for the α and β phases are deduced from the Curie constant $C = N_A I_z^2/k_B$ where N_A and k_B are the 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 $\lesssim T \lesssim$ 15 K, $\Theta_W = 29$ and 25 K and $I_z = 1.4\,\mu_B$ and $1.3\,\mu_B$ are obtained for the α and β phases, respectively.

These observations suggest the existence of local moments far below $T_0 \sim 200$ K, possibly down to $\sim$8 K. This Kondo

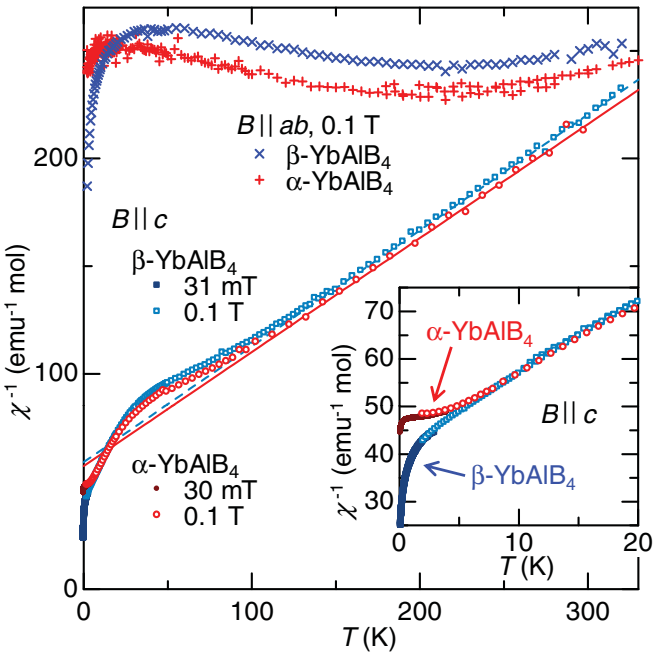

FIG. 2. (Color online) Temperature dependence of the inverse susceptibility $\chi^{-1} = B/M$ under the field along the ab plane and c axis. Solid and dashed lines are Curie-Weiss fits above 150 K for α- (open circles) and β-$YbAlB_4$ (open squares), respectively. The inset shows the low-temperature part of χ^{-1} under the field along the c axis.

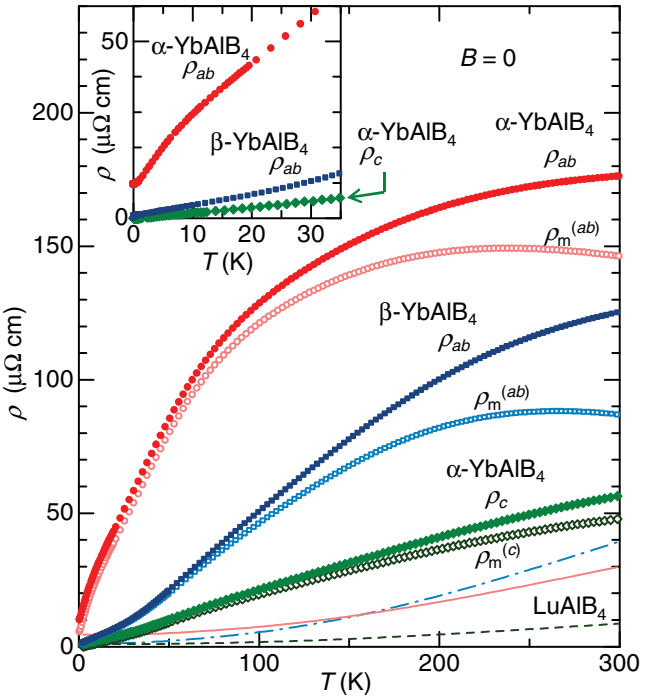

FIG. 3. (Color online) Temperature dependence of the in-plane and c-axis resistivity ρ_{ab} and ρ_c of α-$YbAlB_4$ and ρ_{ab} of β-$YbAlB_4$. The magnetic part of the resistivity ρ_m is obtained by subtracting the nonmagnetic contribution estimated by ρ_{ab} of α- and β-$LuAlB_4$ (solid and dash-dotted lines, respectively) or ρ_c of α-$LuAlB_4$ (dashed line). The inset shows the low T part of ρ_{ab} and ρ_c.

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 $CeSn_3$ (Ref. 23) and $YbAl_3$ (Ref. 18) [see Fig. 1(c)]. A possible origin of this behavior may lie in Kondo resonance narrowing[24] 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.[9] Indeed, the Wilson ratio $R_W = (\pi^2 k_B^2/\mu_0 I_z^2)(\chi/\gamma) \sim 7$ is obtained for both α and β phases by using χ_c at $B = 0.1$ T and $T = 0.4$ K, $\gamma = C_m/T$ at $B = 0$ and $T = 0.4$ 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 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 the [−110] direction, which we denote ρ_{ab}, and c-axis resistivity ρ_c are shown in Fig. 3. We have also measured the a-axis resistivity ρ_a and have found no significant difference from ρ_{ab}. This is consistent with the recent band calculation,[26] which predicts a nearly isotropic transport within the plane. Further investigation of the in-plane anisotropy including the b-axis resistivity ρ_b is now under way. Note that ρ_c in β-$YbAlB_4$ is not yet available due to the tiny thickness of ∼10 μm along the c axis of single crystals. The magnetic part of the resistivity ρ_m is obtained by subtracting the corresponding component of ρ of the nonmagnetic analog α- or β-$LuAlB_4$. The in-plane magnetic component $\rho_m^{(ab)}$ exhibits broad peaks at $T \sim 250$ K in both α-$YbAlB_4$ and β-$YbAlB_4$, which are close to the peak temperatures of $\chi_{ab}(T)$ and T_0 obtained from C_m. Therefore, these may be considered as the coherence peak providing the characteristic hybridization temperature scale. In contrast, $\rho_m^{(c)}$ in α-$YbAlB_4$decreases monotonically on cooling below 300 K.

Interestingly, ρ_c is much smaller than ρ_{ab} in α-$YbAlB_4$, i.e., the conductivity of the system exhibits quasi-one-dimensional anisotropy. The ratio ρ_{ab}/ρ_c increases at low temperatures, making a peak at $T \sim 6$ K [Fig. 4(a), solid black line]. At the peak, ρ_{ab}/ρ_c reaches ∼13 and approaches a constant value of ∼11 at the lowest temperatures. This low-temperature anisotropy is one order of magnitude larger than typical anisotropic heavy fermion systems such as $CeCoIn_5$,[27] $CeCu_2Si_2$,[28] CeNiIn,[29] and YbAgGe,[30] where the ratio is almost T independent and $\lesssim 3$ below 300 K. In contrast, strongly temperature-dependent anisotropic resistivity was reported in $CeRu_2Si_2$ (Ref. 31) and CeNiSn,[32] which increases up to 5 below 10 K. However, this is still two to three times smaller than that observed in α-$YbAlB_4$. In contrast,

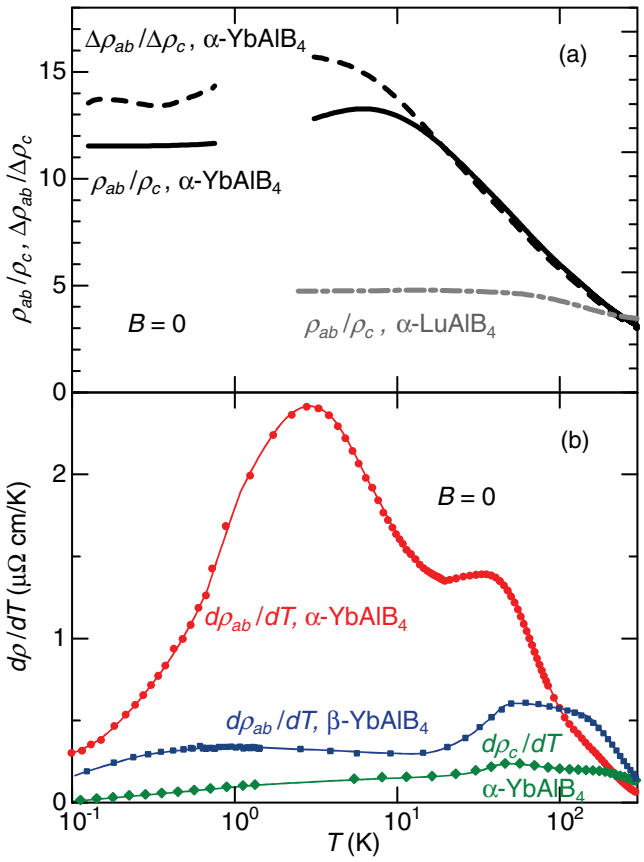

FIG. 4. (Color online) (a) Temperature dependence of the ratios ρ_{ab}/ρ_c and $\Delta\rho_{ab}/\Delta\rho_c$. Here $\Delta\rho$ is defined by $\Delta\rho \equiv \rho - \rho_0$ (see the text). (b) Temperature derivative of the resistivity $d\rho/dT$.

ρ_{ab}/ρ_c in α-LuAlB$_4$ is nearly temperature independent with a slight increase from 3.5 at 300 K to 4.8 at the lowest temperatures [Fig. 4(a), long-dash–short-dashed gray line]. This temperature-independent anisotropy in α-LuAlB$_4$ should come from the anisotropy of the Fermi surface. Interestingly, at $T \sim 300$ K, ρ_{ab}/ρ_c in α-YbAlB$_4$approaches a value similar to the one in α-LuAlB$_4$ although the f-electron contribution is dominant in α-YbAlB$_4$. This suggests that the topologies of the Fermi surfaces of these systems are similar to each other at high $T > T_0$.

The peak found in ρ_{ab}/ρ_c arises mainly from a rapid decrease in ρ_{ab} below $T \sim 10$ K (Fig. 3, inset). This can be clearly seen in the temperature derivative of ρ, $d\rho/dT$, 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 the Kondo lattice behavior, $T^* \sim 8$ K. This suggests that further coherence develops among f electrons due to the formation of heavy quasiparticles below this temperature. The absence of a similar increase in $d\rho_c/dT$ and large ρ_{ab}/ρ_c suggests that the associated heavy fermions are mobile only within the ab plane, but not along the c axis. This is consistent with the recent band calculation that 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 coming mostly 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 χ [Fig. 1(b)] where χ starts to show a 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 ρ_{ab}. Indeed, the recent band calculation suggests the smaller hybridization along the c axis in β-YbAlB$_4$.[26] Although the lower symmetry in α-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 in α- and β-YbAlB$_4$ based on the local Yb site symmetry if the crystal field ground doublet is made solely of $|J_z = \pm 5/2\rangle$.[19,33] In this case, the c-axis transport should come mostly from the conduction electrons and thus should have much larger conductivity because of almost no scattering by f electrons. The resultant anisotropy of the resistivity should be large when the ground state $|J_z = \pm 5/2\rangle$ is dominant at low temperatures. If f electrons start populating the excited CEF levels on heating, the ratio ρ_{ab}/ρ_c should decrease because the node is no longer well defined. Indeed, as shown in Fig. 4(a), ρ_{ab}/ρ_c has a large value below ∼10 K and rapidly decreases with a characteristic temperature scale close to the CEF gap energy of ∼80 K.[19]

The two theoretical indications above strongly support the existence of anisotropic hybridization. It is not likely that the Kondo lattice scale $T^* \sim 8$ K comes from the smaller hybridization scale along the c axis because no feature is observed at ∼8 K in ρ_c. Instead, as already discussed, $T^* \sim 8$ K should arise from the in-plane correlation among f electrons. To confirm this, the Yb-Yb intersite correlation effect should be clarified through, for example, the Lu dilution study in $Yb_{1-x}Lu_xAlB_4$ systems. Note that anisotropic hybridization due to the CEF and its effect on physical properties have been already discussed theoretically in several cases such as for magnetoresistance due to Ce impurities in metals,[34] metamegnetism found in $CeRu_2Si_2$,[35–38] and anisotropic resistivity in CeNiSn.[39] As already mentioned above, the anisotropy found in α-YbAlB$_4$ is more pronounced compared to $CeRu_2Si_2$ and CeNiSn. Therefore, α-YbAlB$_4$ and possibly β-YbAlB$_4$ provide the best systems to study clarifying anisotropic hybridization effects.

The temperature-dependent parts of the resistivity $\Delta\rho \equiv \rho - \rho_0$ at $T < 1$ K are shown in Fig. 5(a). Here ρ_0 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). The zero-temperature limits of the resistivity ρ_0 are 9.4 and 0.82 μΩ cm for ρ_{ab} and ρ_c, respectively, of α-YbAlB$_4$(RRR ∼ 20 and 70) and 0.49 μΩ cm for ρ_{ab} of β-YbAlB$_4$ (RRR ∼ 250). The anisotropy in ρ_0, which corresponds to $\rho_{ab}/\rho_c \sim 11$ at the lowest T, is almost the same as that of $\Delta\rho$ ($\Delta\rho_{ab}/\Delta\rho_c$), which is as large as 13 in the low-T limit [Fig. 4(a)]. The $\Delta\rho_{ab}$ of β-YbAlB$_4$ takes a

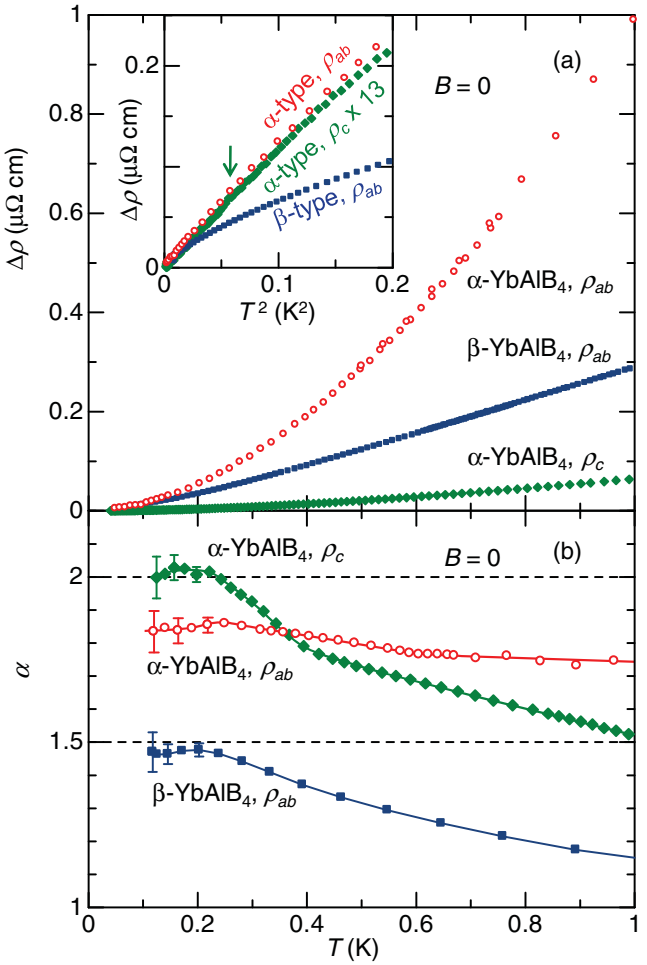

FIG. 5. (Color online) (a) Temperature dependence of $\Delta\rho \equiv \rho - \rho_0$. The inset shows $\Delta\rho$ versus T^2. Note that $\Delta\rho_c$ for α-$YbAlB_4$ is multiplied by a factor of 13 for clarity. The arrow indicates $T_F = 240$ mK estimated for ρ_c of α-$YbAlB_4$by using the resistivity exponent α (see the text). (b) Resistivity exponent α defined by $\Delta\rho = AT^{\alpha}$ (see the text).

value between $\Delta\rho_{ab}$ and $\Delta\rho_c$ of the α phase. In contrast, if we compare $\Delta\rho/\rho_0$, β-$YbAlB_4$ exhibits a much larger value than those in α-$YbAlB_4$. For instance, $\Delta\rho_{ab}/\rho_0 = 0.85$ at $T = 1$ K in β-$YbAlB_4$is ~10 times larger than the respective $\Delta\rho/\rho_0 = 0.11$ (ρ_{ab}) and 0.08 (ρ_c) for α-$YbAlB_4$. This cannot be explained only by the better sample quality in β-$YbAlB_4$and thus the quantum criticality in β-$YbAlB_4$ should also be responsible for the enhancement. Indeed, the application of the magnetic field, suppressing the criticality, decreases $\Delta\rho/\rho_0$ of β-$YbAlB_4$ to the same order as that in α-$YbAlB_4$. Note that even an α-$YbAlB_4$ sample with the highest RRR ~ 110 (estimated by ρ_c) does not exhibit superconductivity down to 35 mK (not shown).

To demonstrate the difference in the ground states of α- and β-$YbAlB_4$, we show the temperature dependence of the power law exponent α defined by $\Delta\rho(= \rho - \rho_0) = A'T^{\alpha}$ [Fig. 5(b)]. The α is obtained by using the equation $\alpha = d \log \Delta\rho / d \log T$. The ρ_0 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. The α is strongly dependent on ρ_0 and its error due to a 0.01% change in ρ_0 is shown in Fig. 5(b). While the exponent α in β-$YbAlB_4$ is small, $\lesssim 1.5$, at low temperatures, those in α-$YbAlB_4$ are much larger and approach the normal value of 2 expected for a FL on cooling. This can also be confirmed in the plot against T^2 [Fig. 5(a), inset], where $\rho_c(T)$ of α-$YbAlB_4$ shows a linear dependence on T^2 below $T_{FL} \sim 240$ mK. The observation of $\alpha \sim 2$ in the lowest temperatures in addition to almost saturating χ and C_m/T below $T^* \sim 8$ K indicates that the ground state of α-$YbAlB_4$is a Fermi liquid.

The T^2 coefficient A defined by $\Delta\rho = \rho_0 + AT^2$ was estimated by the linear fit in the inset of Fig. 5(a) below 240 mK. The values obtained for A are 0.094 and 1.27 $\mu\Omega$ cm/K^2 for ρ_c and ρ_{ab}, respectively. The Kadowaki-Woods ratio A/γ^2 estimated by using these anisotropic A values are 5.8 $\times 10^{-6}$ and 7.8 $\times 10^{-5}$ $\mu\Omega$ cm (K mol/mJ)^2 for ρ_c and ρ_{ab}, 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 K^2) was used. It is known that the ratio A/γ^2 is close to 1.0 $\times 10^{-5}$ $\mu\Omega$cm(K mol/mJ)^2 in many heavy fermion compounds of Kondo lattice systems.[44] In contrast, Tsujii *et al.* have

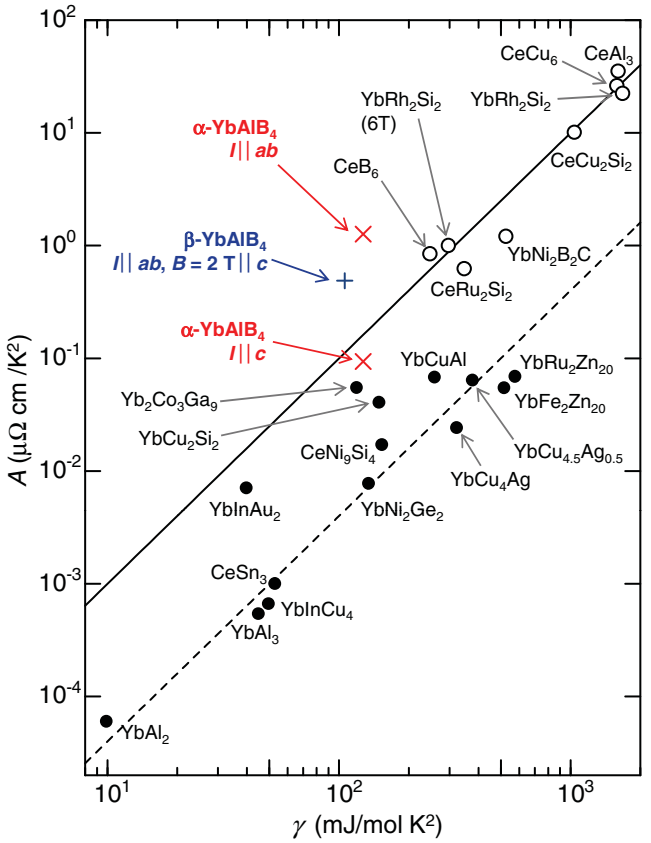

FIG. 6. (Color online) The T^2 coefficient A of the resistivity versus the T-linear coefficient of the specific heat γ for both α- and β-$YbAlB_4$ as well as for other Ce- and Yb-based heavy fermions.[40–42] The open circles denote Kondo lattice systems or the systems with crystal field ground-state degeneracy $N = 2$. The closed circles denote mixed-valence systems, Ce systems with $N = 6$, and Yb systems with $N = 8$. The solid line indicates the original Kadowaki-Woods ratio $A/\gamma^2 = 1.0 \times 10^{-5}$ $\mu\Omega$ cm (K mol/mJ)^2. The dashed line corresponds to $A/\gamma^2 = 4.0 \times 10^{-7}$ $\mu\Omega$ cm (K mol/mJ)^2, which is the typical value in transition metals.[43] The data of β-$YbAlB_4$ at $B = 2$ T $\parallel c$ (Refs. 7 and 9) is also shown.

suggested that the ratio is considerably smaller in intermediate-valence systems or, equivalently, systems with large orbital degeneracy N, i.e., the system with a large hybridization scale T_0 compared to CEF splitting Δ.[40,45] In this case, the expected ratio is close to the typical value known for transition metals $A/\gamma^2 = 0.4 \times 10^{-6}$ $\mu\Omega$ cm(K mol/mJ)^2, which is 25 times smaller than the above ratio for heavy fermions.[43] 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.[40–42] For most of the mixed-valence materials or materials with large N (in Ce systems $N = 6$ and in Yb systems $N = 8$) A/γ^2 is much smaller than the original Kadowaki-Woods ratio and has a value of the order of 10^{-7} $\mu\Omega$ cm(K mol/mJ)^2. Compared to these small values observed in mixed-valence materials, the ratio obtained for ρ_c and ρ_{ab} of α-$YbAlB_4$ is much larger and closer to the typical value for heavy fermions. In β-$YbAlB_4$, the ratio for ρ_{ab} also takes a similar value of 4.4×10^{-5} $\mu\Omega$ cm(K mol/mJ)^2 in magnetic field of 2 T along the c axis.[7,9] The large A/γ^2 in α-$YbAlB_4$ and β-$YbAlB_4$ ($B = 2$ T parallel to the c axis) indicate that the system behaves like Kondo lattices at low temperatures rather than mixed-valence materials. Interestingly, the ratio obtained for ρ_{ab} in both α- and β-$YbAlB_4$ 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.[46] 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 α-$YbAlB_4$ and β-$YbAlB_4$exhibit Kondo lattice behavior with a small renormalized temperature scale of $T^* \sim 8$ K in addition to a large valence-fluctuation scale of ~200 K. Below $T^* \sim 8$ K, α-$YbAlB_4$ forms a heavy-Fermi-liquid state with $\gamma \sim 130$ mJ/mol K^2 in contrast to the unconventional quantum criticality observed in β-$YbAlB_4$. The Kadowaki-Woods ratio takes a typical value for Kondo lattice systems that is considerably larger than those for mixed-valence systems. This is consistent with the Kondo lattice behavior found in the temperature dependence of the susceptibility and specific heat. The large Wilson ratio greater 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 resistivity with one of the strongest anisotropies in heavy fermions suggests that hybridization between $4f$ and conduction electrons is much stronger within the ab plane than along the c axis. Therefore, α-$YbAlB_4$ and possibly β-$YbAlB_4$ should be the best systems to study to deepen our understanding of the anisotropic hybridization effects. This strongly anisotropic hybridization and the large Wilson ratio are the keys to understanding the unusual Kondo lattice behavior and heavy-fermion formation in these mixed-valence compounds. Further work including neutron scattering measurements and studies of Lu dilution effect in $Yb_{1-x}Lu_xAlB_4$ systems is necessary to clarify the origin of these unusual behaviors.

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