Pressure-induced changes of valence fluctuation in β-$YbAlB_4$ probed by x-ray absorption spectroscopy

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We report detailed valence fluctuation phenomena in the unique quantum-critical compound β-$YbAlB_4$ under pressures and at low temperatures. We directly observed the drastic change in the pressure dependence of the mean Yb valence at about 3 GPa with no pressure-induced structural transition. Below 3 GPa, the anomaly in the temperature dependences of the mean Yb valence was observed at about 55 K under ambient pressure, corresponding to a previously observed reduced Kondo coherence temperature, and this temperature rapidly decreases to 22 K at 2.2 GPa, in contrast to the monotonous decrease of the high Kondo temperature (~250 K at ambient pressure). The present results reveal that this reduced Kondo coherence temperature is responsible for the unconventional quantum critical behaviors in β-$YbAlB_4$. Above 3 GPa, it was found that the temperature dependence of the mean Yb valence exhibits a discontinuous change at low temperature (10.5 and 9 K under 4.8 and 5.5 GPa, respectively).

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I. INTRODUCTION

In intermetallic rare-earth compounds with heavy-fermion characters, tuning the ground state by a nonthermal parameter is an important experimental technique to study anomalous phenomena, such as unconventional superconductivities and non-Fermi liquid states which appear in the vicinity of a quantum critical point. In these compounds, rare-earth ions have an almost integer valence and these phenomena are characterized by a small energy scale called a Kondo temperature [1]. In contrast, intermediate-valence rare-earth compounds display a much larger characteristic energy scale owing to stronger hybridizations of the $4f$ electrons in rare-earth ions with conduction (c) electrons [2]. In these compounds, valence fluctuation phenomena reflect degrees of the $4f$-c hybridizations.

The first Yb-based heavy-fermion superconductor β-$YbAlB_4$ exhibits quantum critical (QC) behaviors at low temperature (above $T_c \sim 80\,\mathrm{mK}$) without any tuning parameters [3,4]; these QC behaviors are unconventional, such as the magnetic susceptibility $\chi \sim T^{-0.5}$, the electronic specific heat coefficient $C_e/T \sim \log T$, and the electrical resistivity $\rho \sim T$, which are not accounted for by the spin-density wave description. In contrast to other known QC compounds, the mean Yb valence in β-$YbAlB_4$ was evaluated to be about 2.75 by the x-ray absorption-based methods [5,6] and thus β-$YbAlB_4$ has the high Kondo temperature $T_K \sim 250\,\mathrm{K}$ [4]. β-$YbAlB_4$ crystallizes in a layered orthorhombic *Cmmm* structure [3,7]. At ambient pressure, Yb ions in the Yb and Al layers are centered between seven-member boron rings in the B layers with an accidentally local sevenfold-like symmetry. The theoretical predictions [8,9] suggest that the high value of T_K comes from anisotropic hybridizations between the Yb $4f$ electrons and the conduction electrons in the B layers with the unusual sevenfold-like symmetry around Yb ions. Despite valence fluctuations in β-$YbAlB_4$, incoherent skew scattering appears in Hall resistivities down to $T_{coh} \sim 40\,\mathrm{K}$ [10], one order of magnitude smaller than T_K. Recently, a quasiparticle peak at about 4 meV below the Fermi energy was observed in energy distribution curves obtained by angle-resolved photoemission spectroscopy [11]. This energy scale is consistent with T_{coh}. These results suggest that this reduced Kondo coherence temperature plays a crucial role in anomalous QC behaviors in β-$YbAlB_4$.

In the intermediate-valence rare-earth compounds, hydrostatic pressure, one of the nonthermal parameters, influences $4f$-c hybridizations and radii of the rare-earth ions through decreases of lattice parameters without introducing any disorder. The electrical resistivity of β-$YbAlB_4$ measured under pressure at low temperature indicates suppression of a strange metallic state with the QC behavior at ambient pressure, reaching the Fermi-liquid state at around 1 GPa [12]. No symmetry change in the structure was observed up to around 11 GPa and down to 7 K by synchrotron-radiation (SR)

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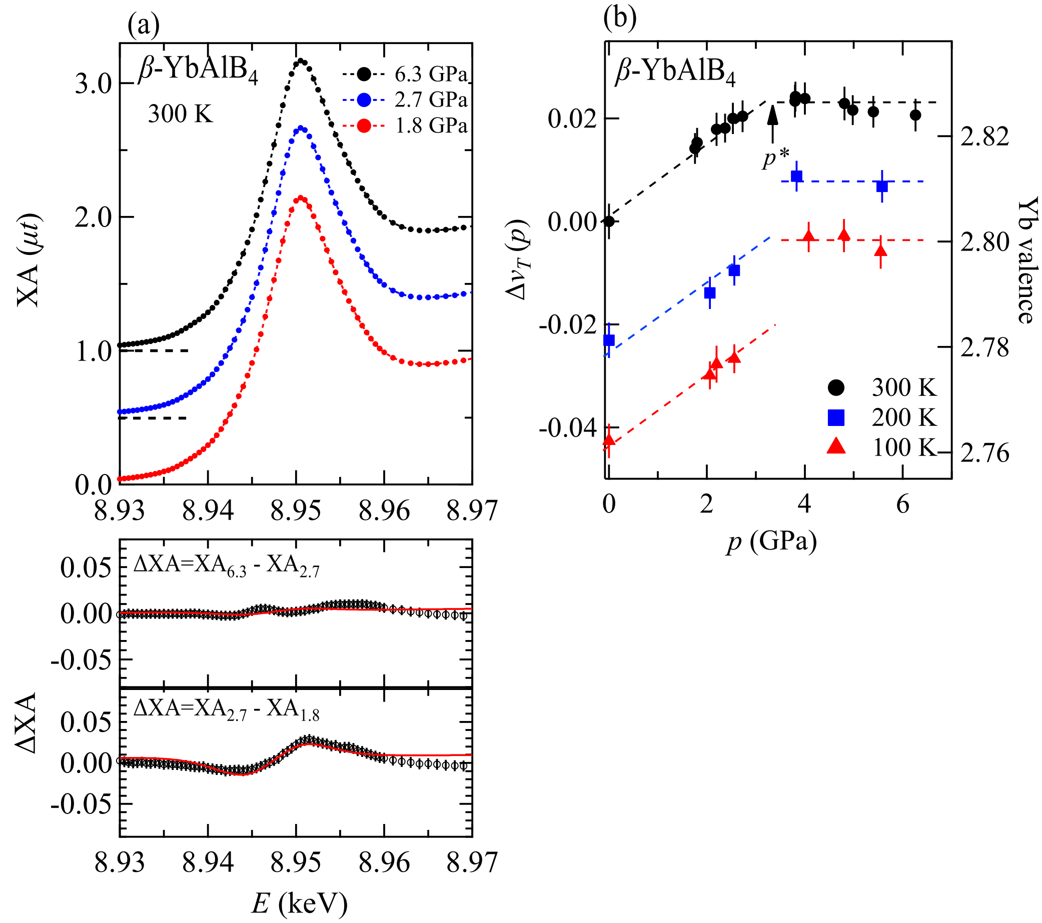

FIG. 1. (a) Selected x-ray absorption (XA) spectra observed under pressure at 300 K and the difference (ΔXA) between two spectra at 300 K and (b) change of the mean Yb valence, $\Delta v_T(p)$, of β-$YbAlB_4$ with pressure at 300, 200, and 100 K. The observed XA spectra have been normalized to an edge jump of unity. In the bottom panel in (a), the red solid lines represent the fitting results using two pseudo-Voigt functions. In (b), the right vertical axis is the mean Yb valence evaluated and the broken lines are visual guides.

powder x-ray diffractions [12,13]. Consequently, there are particular interests in valence fluctuations in β-$YbAlB_4$ under pressure to understand detailed hybridization effects underlying the anomalous QC behaviors.

In this paper, we have systematically studied valence fluctuations in β-$YbAlB_4$ under pressures at low temperatures by x-ray absorption spectroscopy. With the results of our experiments, it was confirmed that the pressure dependence of the mean Yb valence changes at around 3 GPa. Below 3 GPa, the anomaly in the temperature dependence of the mean Yb valence under pressure revealed that the reduced Kondo coherence temperature rapidly decreases above 2 GPa. Above 3 GPa, furthermore, we report experimental evidence of the first-order-like valence change of the Yb ions in β-$YbAlB_4$ at lower temperatures with local structural change around the Yb ions within the orthorhombic *Cmmm* structure.

II. EXPERIMENTAL DETAILS

High-purity single crystals of β-$YbAlB_4$ were grown by the Al-flux method [7]. Energy-dispersive x-ray analysis indicated no impurity phases, no inhomogeneities, and a ratio Yb:Al of 1:1 within the detection limits of the equipment. The powder x-ray diffraction patterns of β-$YbAlB_4$ were collected as a function of temperature at ambient pressure by conventional x-ray diffractometer using the Cu $K\alpha$ radiations.

The experiments of x-ray absorption (XA) spectroscopy at the Yb L_{III} edge were carried out using the single crystalline samples under pressure at the beamline BL39XU on SPring-8, Japan [14] to track the pressure and temperature variation of the Yb valences in β-$YbAlB_4$. Nanopolycrystalline diamonds were used in a clamp-type diamond-anvil cell (DAC) to avoid glitches in XA spectra under pressure [15,16]. The single crystalline sample was loaded into a sample cavity of the Inconel alloy gasket in the clamp-type DAC with ruby crystals and mixtures of methanol-ethanol or Daphne7474 as a pressure-transmitting medium to ensure no pressure-medium dependence in XA spectra observed at low temperatures. Pressure was calibrated by measuring the wavelength shift of the R_1 luminescence line of the ruby crystals in the clamp-type DAC at each experimental condition. In our experiments, the c axis of the single crystalline β-$YbAlB_4$ sample was aligned along the propagation vector of the incident x ray.

III. RESULTS AND DISCUSSION

The selected XA spectra observed at 300 K under pressure are shown in the top panel of Fig. 1(a). The observed

XA spectra have been normalized to a jump of unity at the Yb L_{III} transition. As seen in this figure, the Yb L_{III} transitions are observed in the spectra as a prominent peak at ∼8.95 keV with a tail feature in the lower energy region, reflecting intermediate-valence characters of the Yb ions in β-$YbAlB_4$ at ambient pressure. The pressure dependences of these characteristic features are not significant in the XA spectra observed. Thus, an unambiguous determination of the Yb valences in β-$YbAlB_4$ from these spectra is hindered by the broader line shapes which reflect the short lifetime of the final excited state. To clarify the change in the spectra observed under pressure at 300 K, we evaluated the difference (ΔXA) between two XA spectra observed at different pressures. As shown in the bottom panel of Fig. 1(a), the ΔXA spectrum between 2.7 and 1.8 GPa exhibits the negative and positive peaks at 8.944 and 8.951 keV, respectively. These energies in the Yb L_{III} edge consist with the L_{III} transition ones of the Yb^{2+} and Yb^{3+} ions in compounds. On the other hand, there is no characteristic feature in the ΔXA spectrum between 6.3 and 2.7 GPa within our experimental accuracies. It is suggested that the variation of the Yb valence in β-$YbAlB_4$ is not monotonous with pressure at 300 K.

We have analyzed the ΔXA spectra using two pseudo-Voigt functions with the same intensity and width corresponding to the two peaks of the Yb^{2+} and Yb^{3+} contributions to evaluate the change of the mean Yb valence, $\Delta v_T(p)$, from that at ambient conditions. At 300 K, as seen in Fig. 1(b), $\Delta v_T(p)$ linearly increases with pressure up to $p^* \sim 3.3$ GPa and then almost maintains at about +0.022 with no discontinuity at p^*. Previous SR powder x-ray diffractions of β-$YbAlB_4$ at 300 K [12] represented that the lattice parameters smoothly decrease with pressure up to 11 GPa and the pressure dependence of the unit-cell volume was explained by the equation of state. Although the lattice parameters show no anomaly up to 11 GPa at 300 K, the pressure dependence of the mean Yb valence in β-$YbAlB_4$ substantially changes at p^* and 300 K.

We evaluated the $\Delta v_T(p)$ values from the ΔXA spectra at 200 and 100 K, where the $\Delta v_T(0)$ values were estimated from the temperature dependence of the XA spectra at ambient pressure as shown later. As seen in Fig. 1(b), the $\Delta v_T(p)$ values at 200 and 100 K linearly increase with pressure up to $p^* \sim 3$ GPa and then hardly depend on pressure within our experimental accuracy, which is similar to that at 300 K. Meanwhile, the discontinuous jump in $\Delta v_T(p)$ is observed at p^* in the pressure dependences of $\Delta v_T(p)$ at 200 and 100 K. These results indicate that not only the pressure but also the temperature dependences of the mean Yb valence are different below and above p^*.

The top panels in Figs. 2(a) and 2(b) show selected XA spectra observed at 0.1 MPa ($< p^*$) and 4.8 GPa ($> p^*$), respectively, as a function of temperature. Since the temperature dependences of the spectra are obscure under pressures, ΔXA spectra were evaluated from two XA spectra observed at different temperatures under the same pressure, which are shown in the bottom panels in Figs. 2(a) and 2(b). As seen in the bottom panel in Fig. 2(a), at 0.1 MPa, there is no peak in the ΔXA spectra between 2 and 25 K and between 25 and 40 K. Thus, the mean Yb valence is almost independent of temperature below ∼40 K. At 4.8 GPa, meanwhile, the ΔXA spectrum between 9 and 15 K exhibits a two-peak structure as seen in the bottom panel in Fig. 2(b), suggesting that the mean Yb valence shifts toward divalent around 10 K with decreasing temperature.

We analyzed ΔXA spectra using the two pseudo-Voigt functions to evaluate the change of the mean Yb valence, $\Delta v_p(T)$, from that at 300 K under pressure. Figure 3 shows the typical temperature dependences of $\Delta v_p(T)$ below and above p^*. Since $\Delta v_T(p)$ is less than 1% of the mean Yb valence up to 6 GPa at 300 K as shown in Fig. 1(b), $\Delta v_p(T)$ is similar to the temperature dependence of the normalized mean Yb valence under pressure. As shown in this figure, it is confirmed that the temperature dependences of $\Delta v_p(T)$ are different below and above p^*. At 0.1 MPa, $\Delta v_p(T)$ linearly decreases with temperature down to $T_1 = 55$ K and then is constant at approximately −0.055. The temperature dependence of $\Delta v_p(T)$ maintains in the high-temperature region ($T > T_1$) below p^* within our experimental accuracies and T_1 decreases to 22 K at 2.2 GPa with pressure. Above p^*, the temperature dependence of $\Delta v_p(T)$ becomes weaker and exhibits a discontinuous change of about −0.01 at $T_2 = 10.5$ and 9 K under 4.8 and 5.5 GPa, respectively. We carefully evaluated T_1 and T_2 below and above p^*, respectively, from the temperature variations in ΔXA.

The Yb valences in β-$YbAlB_4$ at ambient pressure have been evaluated by several experimental methods [5,6]. The intrinsic limitation of each experimental method [17] prevents the precise determination of the Yb valence in β-$YbAlB_4$ due to its small mixed valence. However, $\Delta v_p(T)$ was evaluated to be −0.06 at ambient pressure by using several XA-based methods, which is consistent with our result of −0.055 as seen in Fig. 3. The lifetime-broadening-suppressed XA [6] and the bulk-sensitive hard x-ray photoemission spectroscopies [5] evaluated the mean Yb valence in β-$YbAlB_4$ to be 2.76(8) at 2 K and 2.75(2) at 20 K, respectively. As seen in Fig. 3, the low-temperature mean Yb valence in β-$YbAlB_4$ maintains up to T_1 at ambient pressure. We tried to fit the XA spectrum observed at 2 K and ambient pressure with the mean Yb valence of 2.75 using the Voigt and arctangent functions for each valence. As shown in Fig. 2(a), the observed XA spectrum is explained by the mean Yb valence of 2.75. Thus, the pressure and temperature variations of the mean Yb valence were estimated from $\Delta v_p(T)$ and $\Delta v_T(p)$ with the mean Yb valence of 2.75 at 2 K and ambient pressure. The selected pressure and temperature dependences of the mean Yb valence in β-$YbAlB_4$ are shown in the right vertical axis in Fig. 1(b) and the inset of Fig. 3, respectively.

The anomalies observed in the temperature and the pressure dependences of the mean Yb valence are summarized in the pressure vs temperature (p-T) diagram as shown in the top panel of Fig. 4. Although the SR powder x-ray diffractions of β-$YbAlB_4$ under pressure [12,13] represented that the lattice parameters smoothly decrease with pressure to ∼11 GPa, four different regions are observed in the p-T diagram of β-$YbAlB_4$ which are denoted by I, II, III, and IV. As shown in the bottom panel of Fig. 4, the mean Yb valence linearly increases with pressure up to p^* at 3 K (in the I region) at the rate of ∼0.007/GPa which is similar to those at 300, 200, and

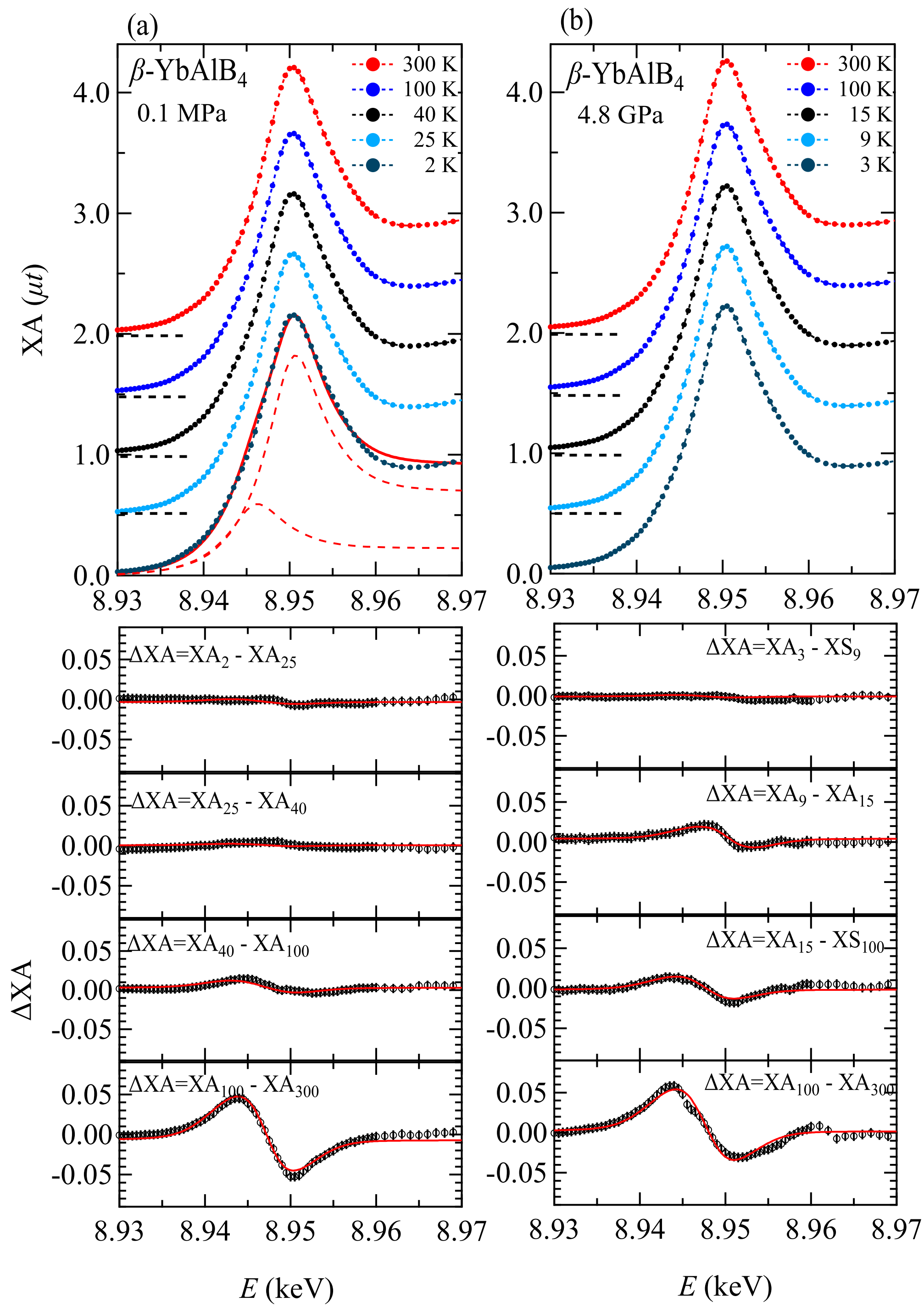

FIG. 2. Selected x-ray absorption (XA) spectra and the difference (ΔXA) between two spectra of β-$YbAlB_4$ observed at (a) 0.1 MPa and (b) 4.8 GPa as a function of temperature. The observed XA spectra have been normalized to an edge jump of unity. The red solid lines in the bottom panels represent the fitting results using two pseudo-Voigt functions. In the XA spectrum at 2 K of the top panel in (a), the red solid and broken lines represent the fitting result with the mean Yb valence of 2.75 and two components of the Yb^{2+} and Yb^{3+} ionic states, respectively, in this fitting result.

100 K below p^* (in the II region). In the III region, the mean Yb valence maintains at approximately 2.78 at 3 K. Accordingly, the pressure dependences of the mean Yb valence are much different below and above $p^* \sim 3$ GPa in β-$YbAlB_4$.

Below p^* (in the I and II regions), the pressure moves the Yb ions in β-$YbAlB_4$ to the localized Yb^{3+} ionic state. These isothermal pressure dependences of the mean Yb valence consist with those in Yb intermediate-valence compounds [18,19]. As seen in Fig. 5 where the date was taken from our previous SR powder x-ray diffraction under pressure at 7 K [13], the pressure dependence of the unit-cell volume, V, was well reproduced by Murnaghan's equation of state. Despite

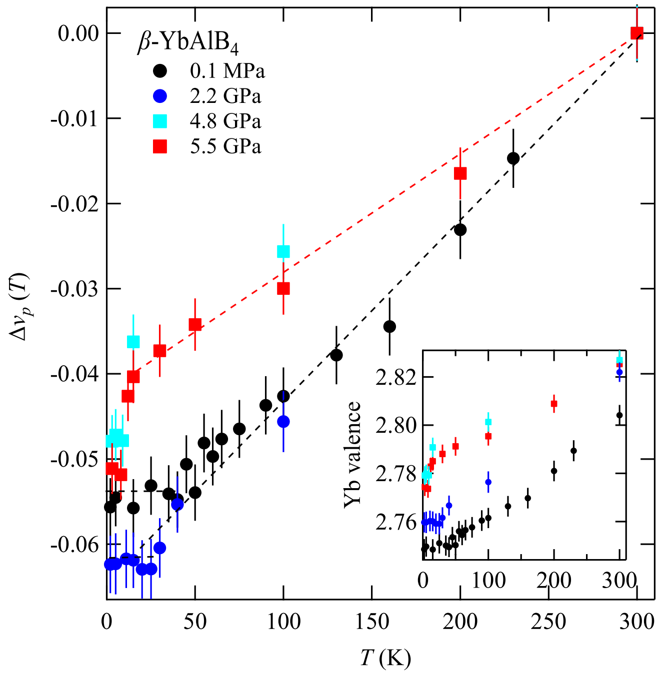

FIG. 3. Change of the mean Yb valence, $\Delta v_p(T)$, at 0.1 MPa, 2.2, 4.8, and 5.4 GPa of β-$YbAlB_4$ as a function of temperature. The broken lines are visual guides. The inset shows the temperature dependences of the mean Yb valence evaluated at 0.1 MPa and 2.2, 4.8, and 5.4 GPa.

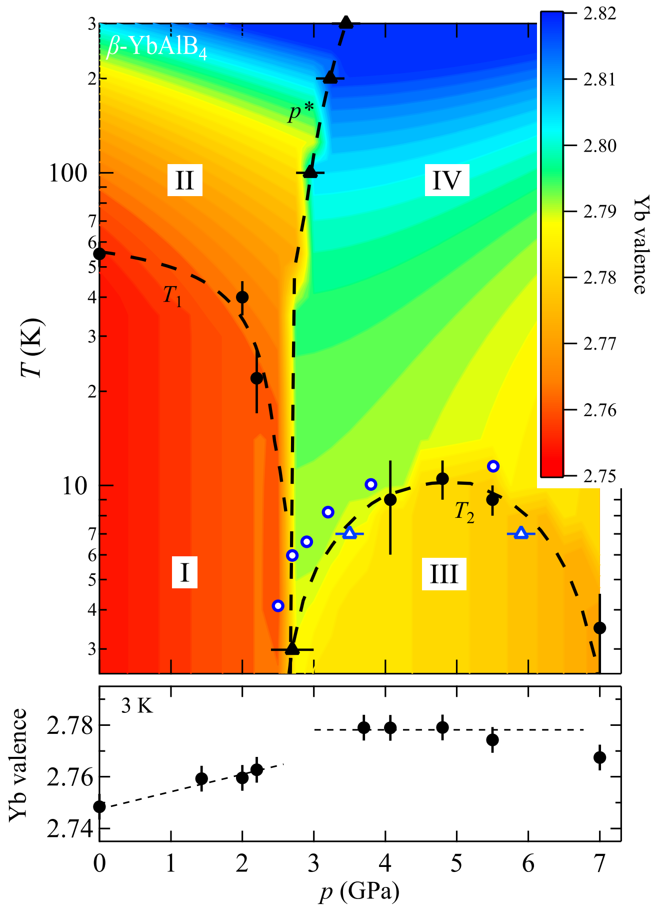

FIG. 4. Pressure vs temperature diagram (top panel) and pressure dependence of the mean Yb valence at 3 K of β-$YbAlB_4$ (bottom panel). In the top panel, closed black circles and triangles with error bars are determined from the temperature and pressure dependences of the mean Yb valence, respectively. Two open blue triangles with error bars at 7 K represent the local structure changes in β-$YbAlB_4$ with the orthorhombic *Cmmm* symmetry obtained by SR x-ray diffraction under pressure [13]. The open circles are the anomalies in the temperature dependences of electrical resistivities under pressure [12]. A false-color plot in the top panel is a guide for the pressure and temperature dependences of the mean Yb valence. The broken lines in the top and bottom panels are visual guides.

no pressure-induced structural transition, the two-dimensional network and the accidental sevenfold-like symmetry in the B layers around the Yb sites significantly change at 3.5 and 5.8 GPa within the orthorhombic *Cmmm* structure. Thus, the local volume around the Yb site is thought to be correlated with the mean Yb valence under pressure. We evaluated the heptagonal-prism volume, $V_{\rm hpt}$, at the Yb site under pressure using the lattice and the atomic coordination parameters refined in Ref. [13]. As shown in Fig. 5, $V_{\rm hpt}$ smoothly decreases with pressure up to 3.5 GPa (in the I region) which is represented by Murnaghan's equation of state using the same parameters as those refined by the pressure dependence of V. The decrease of the number of $4f$ electrons to 13 in Yb ions leads to local pressure that produces a local lattice shrinking. The mean Yb valence linearly increases with pressure in the I and II regions as seen in Figs. 1(b) and 4, and the temperature dependence of the normalized mean Yb valence maintains in the II region as seen in Fig. 3. These results reveal that the $4f$-c hybridizations decrease with pressure in the I and II regions of β-$YbAlB_4$, suggesting that $T_{\rm K}$ with larger characteristic energy scale monotonously decreases from ∼250 K up to p^*.

The T_1, indicating the boundary between the I and II regions, decreases with pressure as shown in the p-T diagram of β-$YbAlB_4$. At ambient pressure, the temperature dependence of the lattice parameters were evaluated by the conventional powder x-ray diffractometer. Figure 6(a) shows selected x-ray diffraction patterns of β-$YbAlB_4$ with Si standard. All diffraction lines in all x-ray diffraction patterns are labeled with the indices of β-$YbAlB_4$ with the orthorhombic *Cmmm* structure and of Si with the cubic diamond structure [20]. The temperature dependences of the (0 0 ℓ) diffraction lines are much different from those of the (h k 0) diffraction lines as seen in Figs. 6(b) and 6(c). The observed x-ray diffraction patterns were analyzed to evaluate the lattice parameters of β-$YbAlB_4$ [21]. As seen in Fig. 6(d), the evaluated lattice parameters, a and b, exhibit anomalous temperature dependences below $T_{ab} \sim 60$ K. At ambient pressure, the T_1 and T_{ab} values are comparable to $T_{\rm coh}$ determined by the Hall effect measurements [10]. Furthermore, the quasiparticle peak was observed at about 4 meV (∼47 K) by angle-resolved photoemission spectroscopy [11]. This reduced Kondo coherence temperature of β-$YbAlB_4$ is characterized by T_1 in the temperature dependence of the mean Yb valence at ambient pressure. Thus, T_1 evaluated under pressure reveals that the reduced

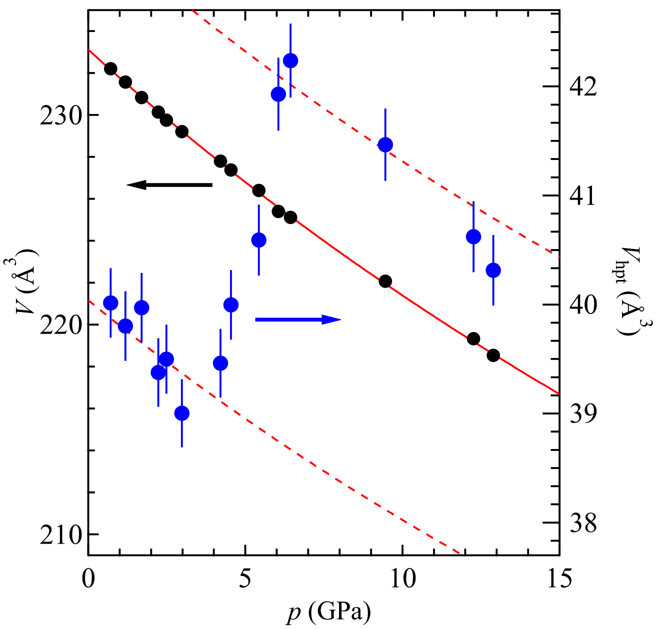

FIG. 5. Pressure dependences of the unit-cell volume, V, of β-$YbAlB_4$ and the heptagonal-prism volume, V_{hpt}, of the distorted YbB_{14} in β-$YbAlB_4$ at 7 K. The V data were taken from Ref. [13] and the V_{hpt} values were evaluated from the lattice and atomic coordination parameters refined using the x-ray diffraction patterns at 7 K [13]. The red solid line represents the curve obtained using Murnaghan's equation of state, $\frac{V}{V_0} = [1 + (\frac{B'}{B})p]^{-1/B'}$, where V_0 is the ambient-pressure volume at 7 K, the bulk modulus, B, is 169(3) GPa, and a pressure derivative of B, B', is 5.1(4) [13]. In two red broken lines, the V_{hpt} values at ambient pressure below 3.5 and above 5.8 GPa were evaluated by Murnaghan's equation of state using the same B and B' values as those in the pressure dependences of V.

Kondo coherence temperature of β-$YbAlB_4$ rapidly decreases from 40 K at 2.0 GPa to 22 K at 2.2 GPa. Under pressure, the strange metallic behaviors in the electrical resistivity at low temperature are suppressed up to about 1 GPa and furthermore, the superconductivity is suppressed around 0.6 GPa [12]. Consequently, the present results strongly suggest that this reduced Kondo coherence temperature is responsible for the unconventional QC behaviors and possibly the superconductivity in β-$YbAlB_4$.

The Fermi surface of β-$YbAlB_4$ calculated by local density approximation contains a cylindrical tube connected quasi-two-dimensional sheets [8,9]. In the phenomenological theoretical model [22], these Fermi surfaces were predicted to have a nodal character in momentum space where a smaller hybridization is expected. The reduced Kondo coherence temperature may be attributed to this characteristic feature in the Fermi surface. In these calculations, the Yb ions are in an almost nominal $4f^{13}$ configuration although β-$YbAlB_4$ is the intermediate-valence rare-earth compound. Recently, unusual slow valence fluctuation between the Yb^{2+} and Yb^{3+} ionic states in β-$YbAlB_4$ is observed below $T^* \sim 8$ K at ambient pressure by SR-based ^{174}Yb Mössbauer spectroscopy [23]. This unusual slow valence fluctuation extends up to ~1 GPa at 2 K, after which a conventional valence fluctuation state with faster valence fluctuation appears in the pressure-induced

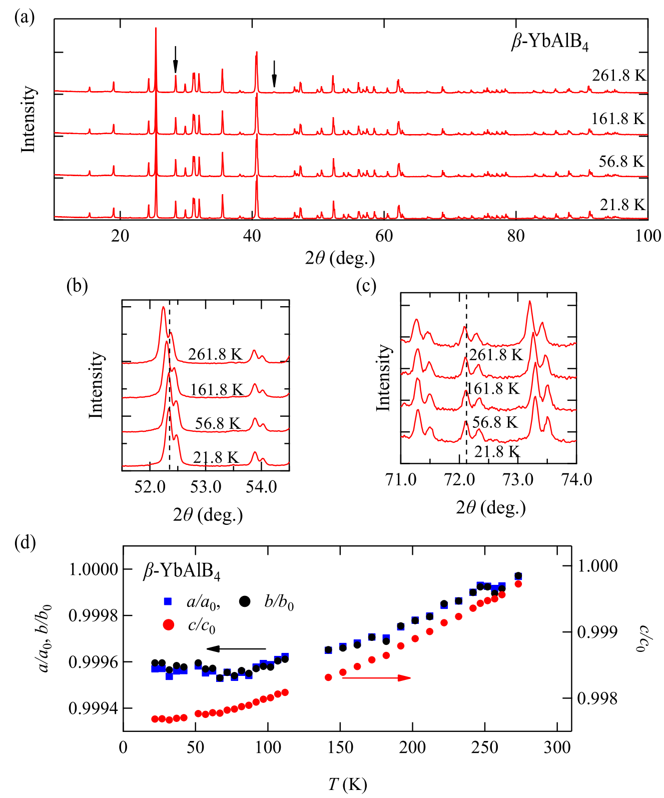

FIG. 6. (a) Selected powder x-ray diffraction patterns of β-$YbAlB_4$ at ambient pressure, (b) and (c) expanded diffraction patterns around 52.3° and 72.35° for the (0 0 2) and (1 7 0) diffraction lines, respectively, and (d) normalized lattice parameters a/a_0, b/b_0, and c/c_0, at 0.1 MPa of β-$YbAlB_4$ as a function of temperature. The a_0, b_0, and c_0 are represented by the lattice parameters at ambient conditions. In (a), the arrows indicate the diffraction lines from the Si standard. In (b) and (c), the dotted vertical lines are visual guides for the (0 0 2) and (1 7 0) diffraction lines.

Fermi liquid state of β-$YbAlB_4$. Thus, detailed theoretical investigations will be important to clarify the interplay between these unusual valence fluctuations and the reduced Kondo coherence temperature in β-$YbAlB_4$ as a function of pressure.

Above p^* (in the III and IV regions), the mean Yb valence evaluated maintains with pressure within our experimental accuracies although the pressure dependences of V at 7 and 300 K are well reproduced by the equations of state. No pressure dependence of a Yb valence reveals local expansion stress around the Yb ions in the crystal applied by pressure. The SR powder x-ray diffractions indicate that not only the inclination angles but also the B-B bond lengths in the B layers strongly depend on pressure from 3.5 to 5.8 GPa at 7 K [13]. The V_{hpt} evaluated in the III region suggests local stress at Yb ions in β-$YbAlB_4$ under pressure. As shown in Fig. 5, V_{hpt} increases with pressure from 3.5 to 5.8 GPa (in the III region). The first-order-like increase of the mean Yb valence at p^* is associated with the change of the local structure on the Yb site in β-$YbAlB_4$ with the orthorhombic *Cmmm* structure. Although pressure dependences of the lattice parameters were observed at 300 K, detailed structural analyses have never been done in the IV region of β-$YbAlB_4$. Since the mean Yb valence discontinuously decreases at T_2 in its temperature dependence above p^*, this anomaly may be related to local structural changes in the orthorhombic *Cmmm* structure. As suggested by theoretical predictions and the band calculations [8,9], the pressure-induced broken in the sevenfold-like symmetry in the B layers directly affects the valence fluctuation phenomena above p^* in β-$YbAlB_4$ through the stronger $4f$-c hybridizations.

As seen in the p-T diagram of β-$YbAlB_4$, the previous electrical resistivity measurements under pressure represented the anomalies in these temperature dependences where the transition may occur from a nonmagnetic to an antiferromagnetic state [12]. As seen in the top panel of Fig. 4, the anomaly at 10 K in the temperature dependence of resistivity under 3.8 GPa consists with that at 9 K in the temperature dependence of the mean Yb valence at 4 GPa. Furthermore, the anomaly at 8.2 K in the temperature dependence of resistivity under 3.2 GPa almost corresponds to the local structural change at 3.5 GPa and 7 K obtained by the SR powder x-ray diffraction [13]. Our recent SR-based ^{174}Yb Mössbauer spectroscopy indicates no magnetic hyperfine field at ^{174}Yb nuclei in β-$YbAlB_4$ down to 2 K at 4.1 GPa and down to 4 K at 5.0 GPa [24]. Consequently, the origin of these anomalies observed in the temperature dependences of resistivity up to $\sim$4 GPa comes from the discontinuous decrease of the mean Yb valence with the local structural changes.

IV. CONCLUSION

In summary, we investigated valence fluctuations in β-$YbAlB_4$ as functions of pressure and temperature using XA spectroscopy at the Yb L_{III} edge. Our results confirm the drastic change in the pressure dependence of the mean Yb valence at $p^* \sim 3$ GPa with no pressure-induced structural transition. Below p^*, the mean Yb valence increases at a rate of $\sim$0.007/GPa with pressure, suggesting that T_K monotonously decreases from $\sim$250 K at ambient pressure up to p^*. The temperature dependence of the mean Yb valence shows an anomaly at T_1 corresponding to the reduced Kondo coherence temperature. T_1 decreases above 2 GPa with pressure, with the consistency of the previous electrical resistivity measurements under pressure. Thus, the present results strongly suggest that this reduced Kondo coherence temperature is responsible for the unconventional QC behaviors in β-$YbAlB_4$.

Above p^*, a discontinuous decrease in the mean Yb valence is observed at T_2 in the temperature dependence, which is related to local structural changes in the orthorhombic *Cmmm* structure. The pressure dependence of T_2 represents the domelike region centered around 5 GPa in the p-T diagram of β-$YbAlB_4$.

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