

Anomalous pressure dependence of the bulk modulus and Yb valence in cubic YbPd

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We investigate the Yb valence instabilities in the strongly correlated YbPd compound using resonant x-ray emission spectroscopy as a function of pressure across the charge-order (CO) transition. At a low temperature ($T = 30$ K) in the CO phase, the Yb $4f$ valence remains nearly constant up to a pressure $P_L = 1.5 \pm 0.2$ GPa, and then increases gradually at higher pressures. In contrast, at room temperature in the normal phase, an anomalous decrease of the Yb $4f$ valence is observed, without any accompanying structural phase transition. This behavior is corroborated by a systematic pressure-dependent decrease of the unit-cell volume. Based on a Birch-Murnaghan analysis, the compressibility indicates hardening of the lattice with applied pressure up to a distinct kink seen at $P_K = 1.6 \pm 0.2$ GPa. In contrast, for $P > P_K$, the Yb $4f$ valence saturates, and the compressibility reveals a counterintuitive pressure-induced softening. The results show a minimum in the compressibility of YbPd (with f^0 – f^1 hole-type mixed valence), reminiscent of the maximum in compressibility seen in the γ – α first-order isostructural phase transition in cerium (with f^0 – f^1 electron-type mixed valence).

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I. INTRODUCTION

Cerium- and ytterbium-based Kondo-lattice systems have attracted considerable attention over the past two decades owing to their outstanding physical properties, including unconventional superconductivity often associated with quantum phase transitions [1,2]. These original properties are due to the competition between the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction and the Kondo effect. These properties may be tuned through control parameters such as chemical doping, pressure (P), or magnetic field, and the competition between the two interactions is well summarized in the Doniach phase diagram [3]. When the hybridization between the $4f$ states and the conduction band is weak, the RKKY interaction dominates. This leads to a stable trivalent $4f$ state and to the formation of large local magnetic moments, which order magnetically at low temperatures. As

the hybridization strength increases, the Kondo interaction becomes dominant: The $4f$ magnetic moments are screened by the conduction electrons, and the valence deviates from the trivalent state.

The strongly correlated YbPd material is a unique Yb compound that exhibits both intermediate valence behavior and charge ordering (CO). It crystallizes in the cubic CsCl structure at room temperature and exhibits an antiferromagnetic phase transition at $T_N = 1.9$ K due to the competition between the RKKY interaction and the Kondo effect [4]. Upon cooling from room temperature to T_N , YbPd undergoes two transitions. Below $T_1 = 130$ K, the cubic structure with a unique Yb site transforms into a tetragonal structure [5,6] with two distinct Yb sites [Figs. 1(a) and 1(b)] that form an incommensurate CO phase characterized by the wave vector $(\pm 0.07 \pm 0.07 \ 1/2)$, which becomes a commensurate CO phase below $T_2 = 105$ K characterized by the wave vector $(0 \ 0 \ 1/2)$. At low temperature, a trivalent magnetic Yb^{3+} ion and strongly intermediate-valent nonmagnetic $\text{Yb}^{2.6+}$ ion are arranged alternatively along the c axis, which has also been termed valence order [7–10]. While the trivalent magnetic Yb^{3+} ions undergo an antiferromagnetic transition at T_N , the CO/valence order in YbPd is very intriguing since YbPd shows metallic electrical resistivity from above T_1 down to low temperature. Accordingly, many studies have been devoted to

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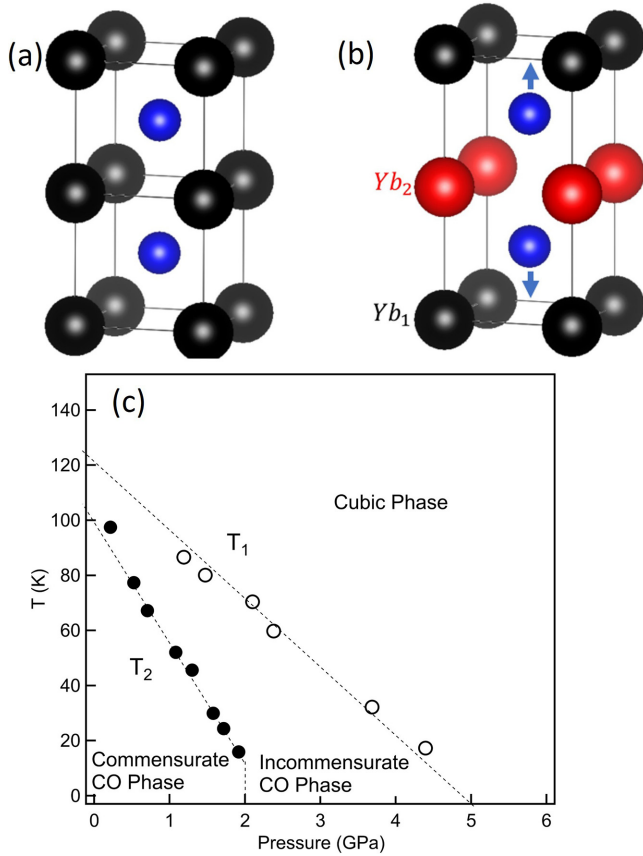


FIG. 1. Crystal structure of YbPd at ambient pressure (a) above $T_1 = 130$ K (cubic phase) and (b) below T_1 (tetragonal phase). YbPd shows only one type of Yb site in the normal cubic phase, but shows two types of Yb sites in the tetragonal CO phase. (c) P - T phase diagram reported in Refs. [12,24].

understanding the magnetic ordering in the intermediate valence YbPd compound as well as the origin of the CO transition [4,9,11–14]. Further, the intermediate valence of the nonmagnetic Yb site, which is indicative of strong hybridization, suggests that Kondo-like physics is at play in the electronic structure of YbPd. While earlier studies focused on the possibility of charge ordering originating from Fermi surface nesting, band structure calculations failed to find any evidence for Fermi surface nesting, but indicated strong correlation effects [4,15]. It is well known that high-energy spectroscopies in combination with model calculations are well suited to characterize the ground-state electronic structure as well as excitation spectra of strongly correlated materials [4,16–20]. In a recent study [16], Liao *et al.* showed that the bulk-sensitive temperature-dependent Yb $3d$ core level spectra as well as the valence band spectra of YbPd could be nicely explained by simplified single-impurity Anderson model (SIAM) calculations. In particular, it was shown that YbPd exhibits a dual Kondo effect originating in the two types of Yb ions, which in turn leads to zero thermal expansion in the CO phase. The study quantified the site- and T -dependent single-ion Kondo temperatures (T_{K1} and T_{K2}) across the incommensurate and commensurate CO transitions. Further, it was shown that the site- and T -dependent T_{K1}

and T_{K2} values follow the evolution of lattice parameters in the CO phase. Thus, the results showed a direct relation between the dual Kondo effect and structural changes associated with CO, leading to a zero thermal expansion in the CO phase [16].

Pressure-dependent ac-calorimetry and electrical resistivity measurements revealed another peculiar behavior of the YbPd compound. Unlike typical ytterbium compounds, where pressure usually stabilizes magnetic ordering, in YbPd the magnetic transition temperature T_N decreases with increasing pressure and is discontinuously suppressed at $P_C = 1.9$ GPa [7,12,13]. Both the CO transitions at T_1 and T_2 also decrease monotonically with increasing P [Fig. 1(c)]. A precise understanding of the exotic properties of YbPd requires detailed knowledge of the temperature and pressure dependence of the Yb valence. While temperature-induced valence changes have been investigated using high-energy spectroscopic techniques [4,16,17], the pressure dependence of the valence evolution in YbPd has not yet been reported.

In this study, we investigate the pressure dependence of the Yb valence in YbPd under high pressure at temperatures below and above the CO transition by means of high energy resolution fluorescence detected x-ray absorption spectroscopy (HERFD-XAS) and resonant x-ray emission spectroscopy (RXES) at the Yb L_3 edge, which are sensitive probes of the $4f$ valence state owing to their superior spectral resolution [18–20]. To study the interplay between the Yb valence and the CO, we measured the pressure dependence of the Yb valence below the CO transitions in the tetragonal phase (at $T = 30$ K) and in the cubic phase (at room temperature $T = 300$ K). At low temperature ($T = 30$ K), the valence is constant up to $P_L \sim 1.5 \pm 0.2$ GPa but it increases at higher P . Remarkably, the measurements at room temperature revealed a significant decrease of the Yb valence at low pressures ($P < P_K$, with $P_K = 1.6 \pm 0.2$ GPa), followed by a nearly constant mixed valence at higher P . This is accompanied by a kink at $P = P_K$ in the P -dependent unit-cell volume. Moreover, while the compressibility β of most materials generally increases with P , YbPd exhibits a minimum in β versus P at $P = P_K$. This behavior is reminiscent of the changes in β , albeit with a maximum instead of minimum, at the γ - α first-order isostructural phase transition in cerium metal [21–23].

II. EXPERIMENTAL DETAILS

The polycrystalline YbPd sample was prepared from high-purity (at least 99.9%) commercially available elements. A mixture with $Yb_{1.03}Pd$ starting composition to compensate for the Yb vaporization at elevated temperature was placed in a Ta crucible. The crucible was sealed by arc welding in a glove box under a pure Ar atmosphere. The crucible was heated up to ~ 1550 °C in an induction furnace for several minutes. Room-temperature x-ray diffraction (XRD) indicates that the sample is single phase with the CsCl-type cubic structure. The RXES and HERFD-XAS measurements were performed using the multianalyzer x-ray emission spectrometer “MULTIXS” on the resonant inelastic x-ray scattering (RIXS) end station of the GALAXIES beamline [25] at the SOLEIL synchrotron. The spectrometer was equipped with

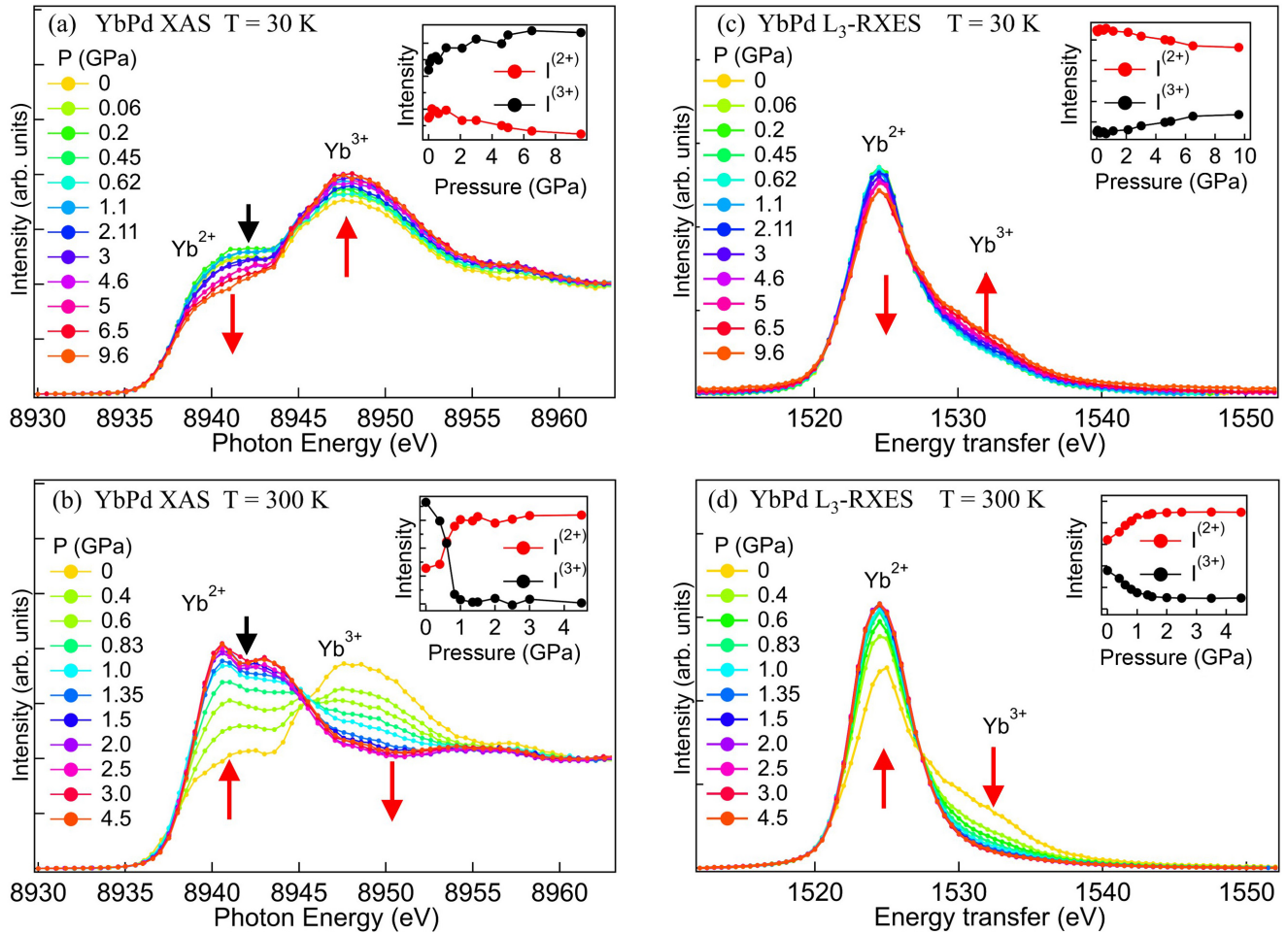


FIG. 2. Yb L_3 XAS and RXES spectra of YbPd as a function of pressure at (a) $T = 30$ K and (b) $T = 300$ K. The incident photon energy ($h\nu_{\text{in}} = 8942$ eV) of the RXES spectra was set to the maximum of the Yb^{2+} resonance at the Yb L_3 absorption edge indicated by the black arrows in panels (a) and (b). The inset shows the evolution of the integrated intensities of the Yb^{2+} and Yb^{3+} final states with pressure.

three Si(620) bent-crystal analyzers (with a bending radius $R = 1$ m), which were operated near the Bragg angle of 76.770° . The overall energy resolution was about 1 eV as determined by the full width at half maximum of the elastic line. To study the P - T dependence of Yb valence, the sample was mounted in a diamond anvil cell (DAC) with silicone oil as the pressure-transmitting medium and ruby as the pressure indicator. The cell was subsequently mounted in a specially designed high-pressure He flow cryostat. High-pressure XRD experiments were performed using the high-pressure XRD diffractometer at the Laboratoire de Physique des Solides. The instrument is equipped with a microfocused Mo x-ray tube ($\lambda = 0.71$ Å) coupled to Montel multilayer focusing optics, providing a beam size of approximately 100 μm at the sample position with a divergence of 0.5 mrad. Diffracted intensities were collected using a two-dimensional MAR345 image plate detector. The two-dimensional diffraction images were radially integrated to obtain one-dimensional diffraction patterns using the DIOPTAS software package [26]. High pressures were generated using a DAC equipped with 1 mm culet diamonds and a CuBe gasket. The sample chamber was loaded with powdered YbPd, together with gold used as an internal

pressure calibrant [27] and silicone oil as pressure-transmitting medium [28,29].

III. RESULTS

Figure 2(a) shows the pressure-dependent Yb L_3 HERFD-XAS spectra of YbPd measured at $T = 30$ K. The spectra were recorded by scanning the incident photon energy across the Yb L_3 absorption edge while monitoring the scattered intensity of the Yb $L_{\alpha 1}$ fluorescence line ($2p^5 3d^{10} \rightarrow 2p^6 3d^9$, $h\nu_{\text{out}} = 7417$ eV). The HERFD-XAS spectral shape is sharper than the standard XAS spectrum because the broadening is determined by the longer lifetime of the shallower $3d$ hole, rather than by that of the deep $2p$ hole [30]. The spectra exhibit two peaks at around 8940 and 8947 eV corresponding to the transition from the initial intermediate valence ground state $|g\rangle = a|f^{13}\rangle + b|f^{14}\rangle$ to a final state with mainly Yb^{2+} ($2p^6 4f^{14} \rightarrow 2p^5 4f^{14} 5d^1$) and Yb^{3+} ($2p^6 4f^{13} \rightarrow 2p^5 4f^{13} 5d^1$) characters. This intermediate valence character at low temperatures is consistent with early XAS at Yb L_3 edge (average valence $v = 2.80$) [4] as well as soft x-ray photoemission ($v = 2.75$) [17] and hard x-ray

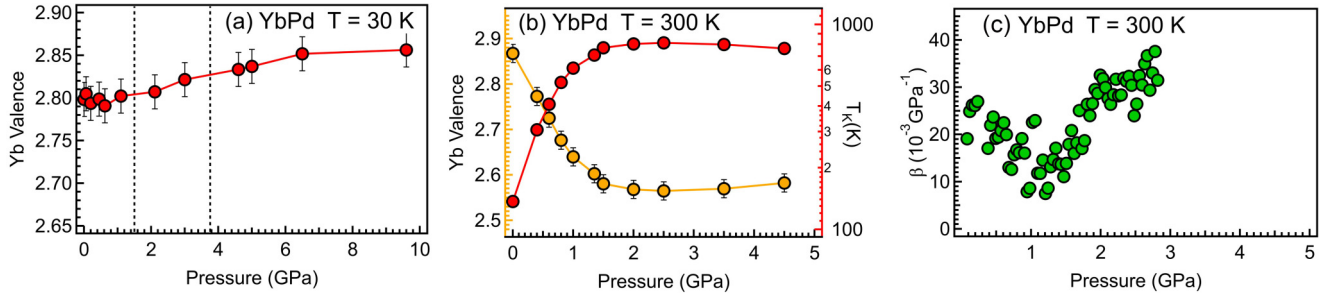


FIG. 3. (a) P dependence of the Yb valence at $T = 30$ K. (b) P dependence of the Yb valence (left scale) and the Kondo temperature T_K (right scale) at room temperature. (c) P dependence of the compressibility β at 300 K.

($v = 2.79$) [16] photoemission measurements. The Yb^{2+} structure is split into two peaks separated in energy by ~ 2.3 eV. This splitting is attributed to the $5d$ crystal field effect [31,32]. With increasing pressure, a clear spectral-weight transfer from Yb^{2+} to Yb^{3+} is observed. In contrast, at room temperature, the Yb^{3+} decreases and is transferred to Yb^{2+} with increasing pressure as shown in Fig. 2(b). It should be noted that the spectral shape of the Yb^{2+} structure does not change with pressure. In particular, the relative intensity between the peaks at ~ 8941 and ~ 8943 eV remains essentially constant with increasing pressure (see Appendix A for more details).

Extracting valence from XAS spectra is hampered by uncertainty in the edge-step position and background due to the edge-to-continuum jump. RXES is a more efficient way, thanks to its resonant character, which allows for the enhancement of either the Yb^{2+} or Yb^{3+} configuration, providing an accurate determination of the Yb valence [19,20,33,34]. The pressure-dependent Yb $L\alpha_1$ RXES spectra measured with the incident photon energy tuned at the Yb^{2+} emission line of the absorption edge for $T = 30$ K and $T = 300$ K are shown in Figs. 2(c) and 2(d). The spectra are plotted as a function of the transferred energy E_t and normalized to the area under the curve to highlight the spectral changes. The RXES spectra exhibit two structures corresponding to Yb^{2+} ($2p^6 4f^{14} \rightarrow 2p^5 4f^{14} 5d^1 \rightarrow 2p^6 3d^9 4f^{14}$) and Yb^{3+} ($2p^6 4f^{13} \rightarrow 2p^5 4f^{13} 5d^1 \rightarrow 2p^6 3d^9 4f^{13}$) final states and similar spectral changes with pressure are observed, as in the XAS spectra.

Let us now turn to the quantitative analysis of the Yb valence as a function of pressure. The spectra are analyzed using phenomenological fitting with a standard pseudo-Voigt function for each valence state (as detailed in Appendix B). The Yb valence v is quantitatively determined from the RXES spectra using the expression [19,20]

$$v = 3 - \frac{I^{(2+)}}{I^{(2+)} + \frac{I^{(3+)}}{\zeta}}, \quad (1)$$

where $I^{(2+)}$ and $I^{(3+)}$ are, respectively, the integrated intensities of the peaks Yb^{2+} and Yb^{3+} . The parameter ζ accounts for the reduced Yb^{3+} spectral weight observed in RXES spectra, which depends on the incident photon energy. A value of $\zeta = 8.5\%$ yields a valence at ambient pressure consistent with that determined by hard x-ray photoemission spectroscopy ($v = 2.79$ at 20 K) [16].

Figures 3(a) and 3(b) show the pressure dependence of the Yb valence obtained from RXES at $T = 30$ K and $T = 300$ K, respectively. In Fig. 3(a), the dotted vertical lines mark the pressure-induced transition as found from earlier work (as discussed in Ref. [12] and references therein); the commensurate CO state is stable up to $P_L \sim 1.5 \pm 0.2$ GPa, where it transforms into the incommensurate CO and the cubic phase is subsequently restored for P higher than 3.7 GPa at $T = 30$ K. The mean Yb valence increases globally upon increasing pressure, as expected. It is almost P independent in the commensurate CO state, increases throughout the incommensurate CO range, and gradually saturates in the normal cubic phase. In the low- P quadratic commensurate CO phase, there are two Yb sites with valencies of ~ 3.0 and ~ 2.6 for Yb_1 and Yb_2 , respectively [16], while in the cubic high- P phase, there is only a single Yb site with valence ~ 2.85 . Hence, while the pressure increase yields an unusual decrease of the Yb_1 valence, the mean valence does show a gradual increase from $v \sim 2.80$ to $v \sim 2.85$ [16], because a pressure increase leads to an increase of the Yb valence and the Yb^{3+} (f^{13}) ion is smaller compared to that of the Yb^{2+} (f^{14}) ion [35–44].

At room temperature, we directly—and unexpectedly—observe an anomalous pressure-induced change in the Yb valence. The Yb valence first drops at low pressure ($P \leq 1.5$ GPa), where it decreases from 2.87 to 2.58, before remaining almost constant upon further pressure increase. A similar pressure-induced anomalous valence reduction with pressure has recently been observed in YbCu_5 [45], $\text{YbCu}_{4.5}$ [46], and $\text{YbCu}_{6.5}$ [47]. In $\text{YbCu}_{4.5}$ [46], the Yb valence decrease is linked to a structural first-order transition, while in YbCu_5 [45] and $\text{YbCu}_{6.5}$ [47], it has been suggested that this anomalous behavior might be due to peculiar features of the density of states near the Fermi level.

IV. DISCUSSION

The pressure dependence of the Kondo temperature T_K in Ce- and Yb-based heavy-fermion compounds has been studied theoretically within the framework of the SIAM [48]. It was shown that for Ce compounds, T_K increases monotonically, whereas for Yb compounds it decreases at low pressure, shows a broad minimum, and then increases with further increase in pressure. At room temperature, YbPd might be located near the broad minimum. It is well known that,

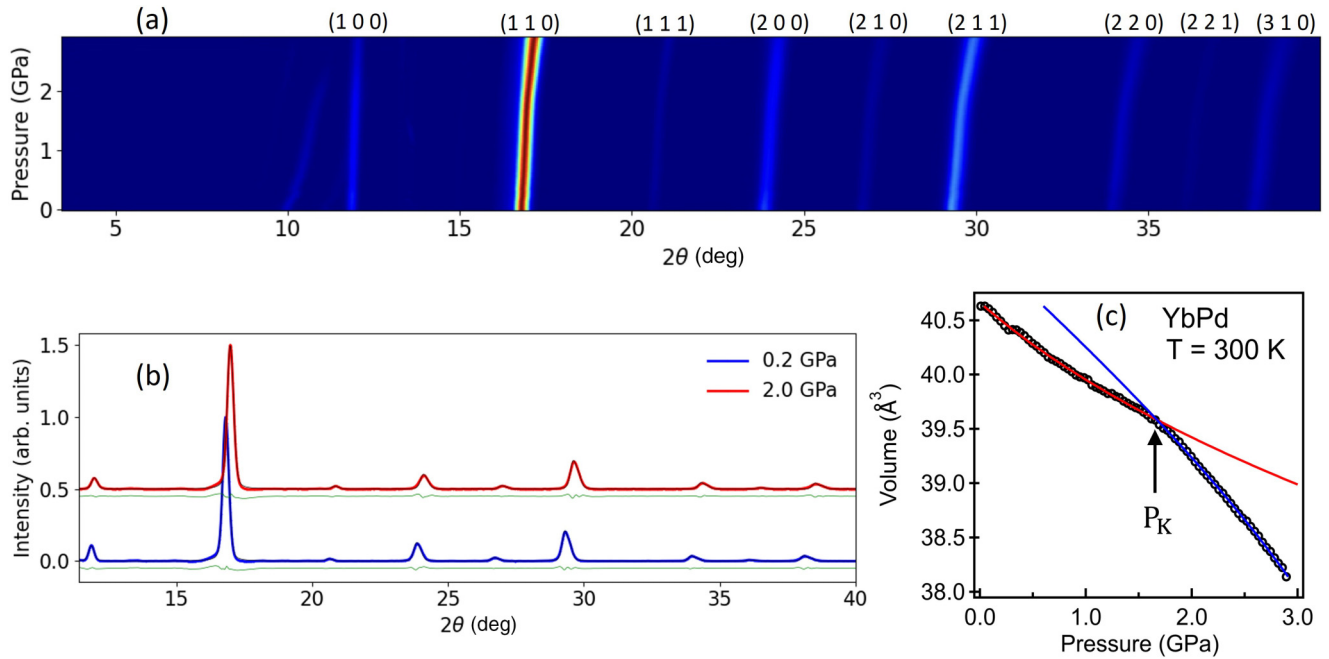


FIG. 4. (a) Contour map of x-ray diffraction intensities collected in the pressure range 0–3 GPa. (b) XRD patterns at $P = 0.2$ GPa (blue) and 2.0 GPa (red) together with the Le Bail fit (black) and the difference plot between the data and the fit (green). (c) Pressure dependence of the volume. The blue and red curves are the third-order Birch-Murnaghan's equation of state for $P \leq P_K$ and $P \geq P_K$, respectively. The arrow indicates the pressure P_K where the kink is observed.

in Yb-based compounds, the Kondo temperature T_K scales with the occupation of the $4f$ hole number n_h (the Yb valence $v = 2 + n_h$). By combining the Kondo energy scale T_K , extracted from thermodynamic measurements, with the $4f$ -state occupation determined by RIXS experiments, Kummer *et al.* [20] recently showed that this scaling follows a universal power law, $3 - v = aT_K^n$, with $n = 2/3$ and $a = 1/200$. We employed this power law to estimate the pressure dependence of the Kondo temperature for the single Yb site at room temperature, as shown in Fig. 3(b). The Kondo temperature increases markedly from 137 to 768 K at $P_K \sim 1.6 \pm 0.2$ GPa and then remains nearly unchanged upon further increase in pressure.

We performed room-temperature P -dependent XRD measurements to investigate how the Yb valence variation couples to the crystal structure. Figure 4(a) shows the contour map of x-ray diffraction intensities as a function of pressure; no structural phase transition is detected up to 3 GPa, as shown in Fig. 4(b). Figure 4(c) shows the pressure dependence of the unit-cell volume extracted from the XRD data. The lattice parameter a was obtained using a Le Bail fit for each pattern [see Fig. 4(b)]. The unit-cell volume decreases with pressure with a kink near $P_K \sim 1.6 \pm 0.2$ GPa.

At low P , the compressibility $\beta = -\frac{1}{V} \frac{\partial V}{\partial P}$ is found to decrease with increasing pressure as is typical for solids [49]. On increasing P further, it exhibits a minimum near $P_K \sim 1.6 \pm 0.2$ GPa, the pressure at which the Yb valence saturates, and beyond P_K , the compressibility increases at higher pressures indicating an unusual behavior not reported in Yb-based mixed-valent materials to date [Fig. 3(b), right scale]. This indicates a change of stiffness that we quantify using a Birch-Murnaghan analysis in the following. We

extracted the bulk modulus K_0 (the reciprocal of the compressibility), which characterizes the compression behavior under hydrostatic pressure, and its pressure derivative $K'_0 = \frac{\partial K_0}{\partial P}$ that defines the change in stiffness using the standard third-order Birch-Murnaghan equation of state [50]:

$$P(V) = \frac{3}{2} K_0 \left[\left(\frac{V_0}{V} \right)^{\frac{7}{3}} - \left(\frac{V_0}{V} \right)^{\frac{5}{3}} \right] \times \left[1 + \frac{3}{4} (K'_0 - 4) \left(\left(\frac{V_0}{V} \right)^{\frac{2}{3}} - 1 \right) \right], \quad (2)$$

where P and V are the pressure and volume, respectively, and V_0 and K_0 are the values of V and K at zero pressure. The data for the $V(P)$ functions, illustrating the fitted equation of state, are shown in Fig. 4(c). We obtain the parameter of $K'_0 = 22.3$, $K_0 = 47.18$ GPa, and $\text{\AA}^3$ for $P < P_K$ and $K'_0 = -6.1$, $K_0 = 38.32$ GPa, and $V_0 = 39.6 \text{ \AA}^3$ for $P > P_K$. Remarkably, we observe a sign change in the derivative of the bulk modulus K'_0 .

Negative values of K'_0 are quite rare and correspond to an effect known as *pressure-induced softening* [51–58], a counterintuitive phenomenon in which a material becomes increasingly compressible under pressure. Pressure-induced softening has been observed in a few but very different materials, including cerium metal [21], the amorphous silica [51], ZrW_2O_8 [53], $\text{Zn}(\text{CN})_2$ [54], and ScF_3 [58]. Among these materials, cerium exhibits strong similarities with YbPd. Indeed, cerium undergoes an isostructural face-centered cubic γ – α phase transition, accompanied by a volume collapse of approximately 15% at room temperature under pressure. The bulk modulus of cerium exhibits a minimum at the γ – α

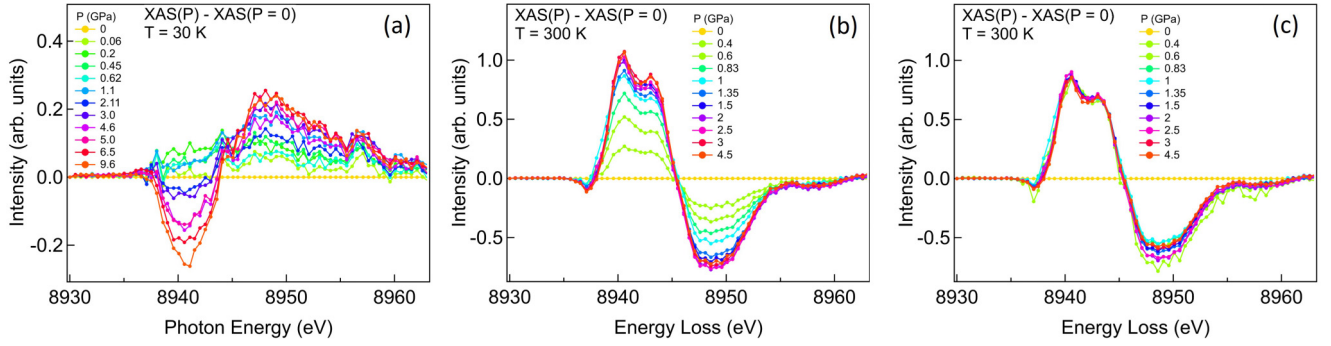


FIG. 5. Experimental difference $XAS(p) - XAS(p = 0)$, at (a) $T = 30$ K and (b) $T = 300$ K. Panel (c) shows the difference spectra at $T = 300$ K normalized to the intensity of the peak at ~ 8941 eV.

volume-collapse transition that persists even above the critical endpoint in the supercritical state [21–23,59,60]. Moreover, the phase transition in Ce is marked by a pronounced increase in the Kondo temperature, occurring concomitantly with the volume collapse under pressure. The f electrons play a central role in this transition. As shown in Fig. 3(b), YbPd likewise

exhibits a rapid increase in the Kondo temperature at low pressures up to P_K —at which the compressibility reaches a minimum—and then becomes nearly constant above P_K . The sign change of K'_0 is reminiscent to that observed in Ce. Both the pressure-temperature phase diagram and the temperature dependence of the bulk modulus of cerium have been

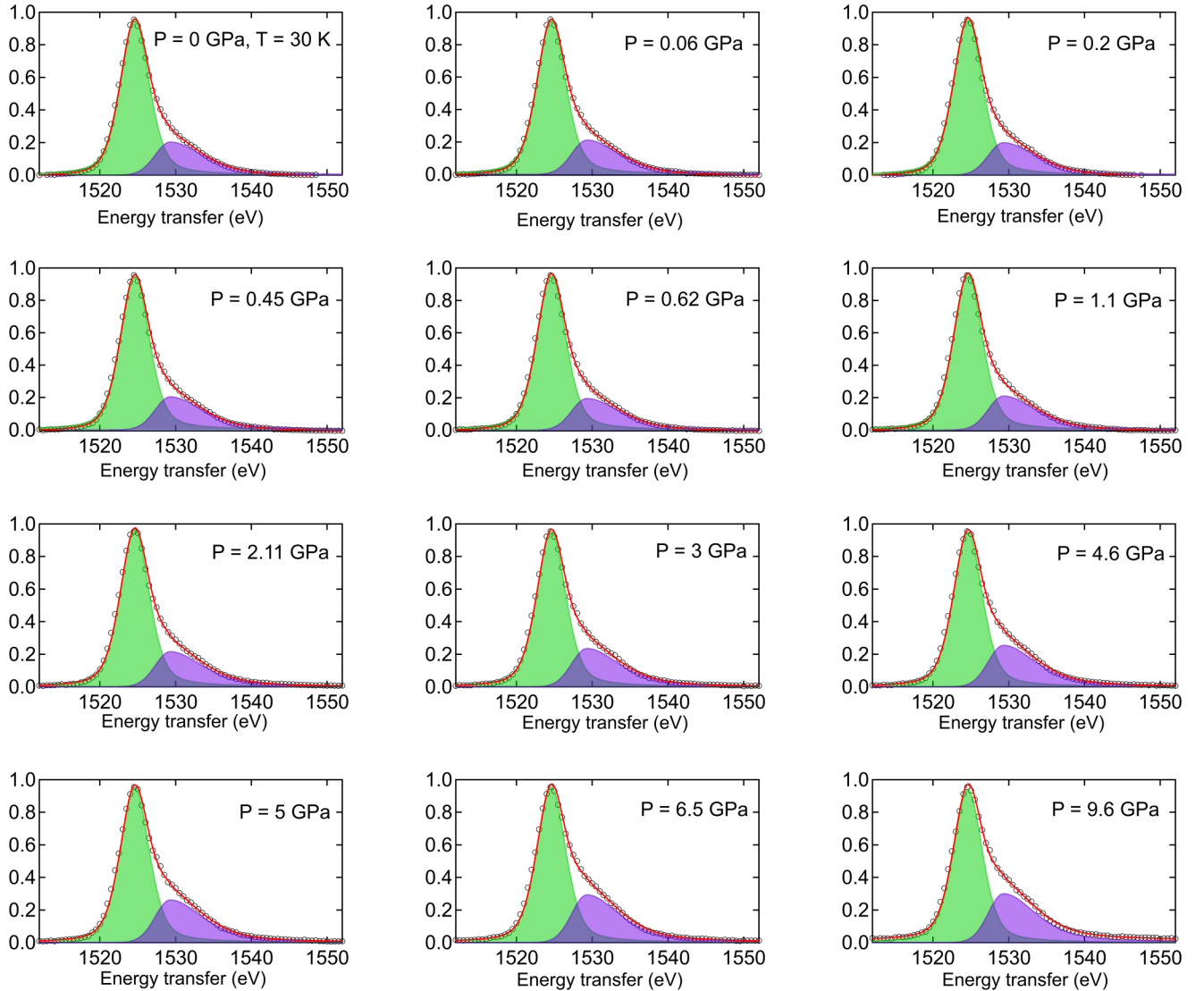


FIG. 6. Results of the fit analysis of the RXES spectra for all measured pressures at $T = 30$ K.

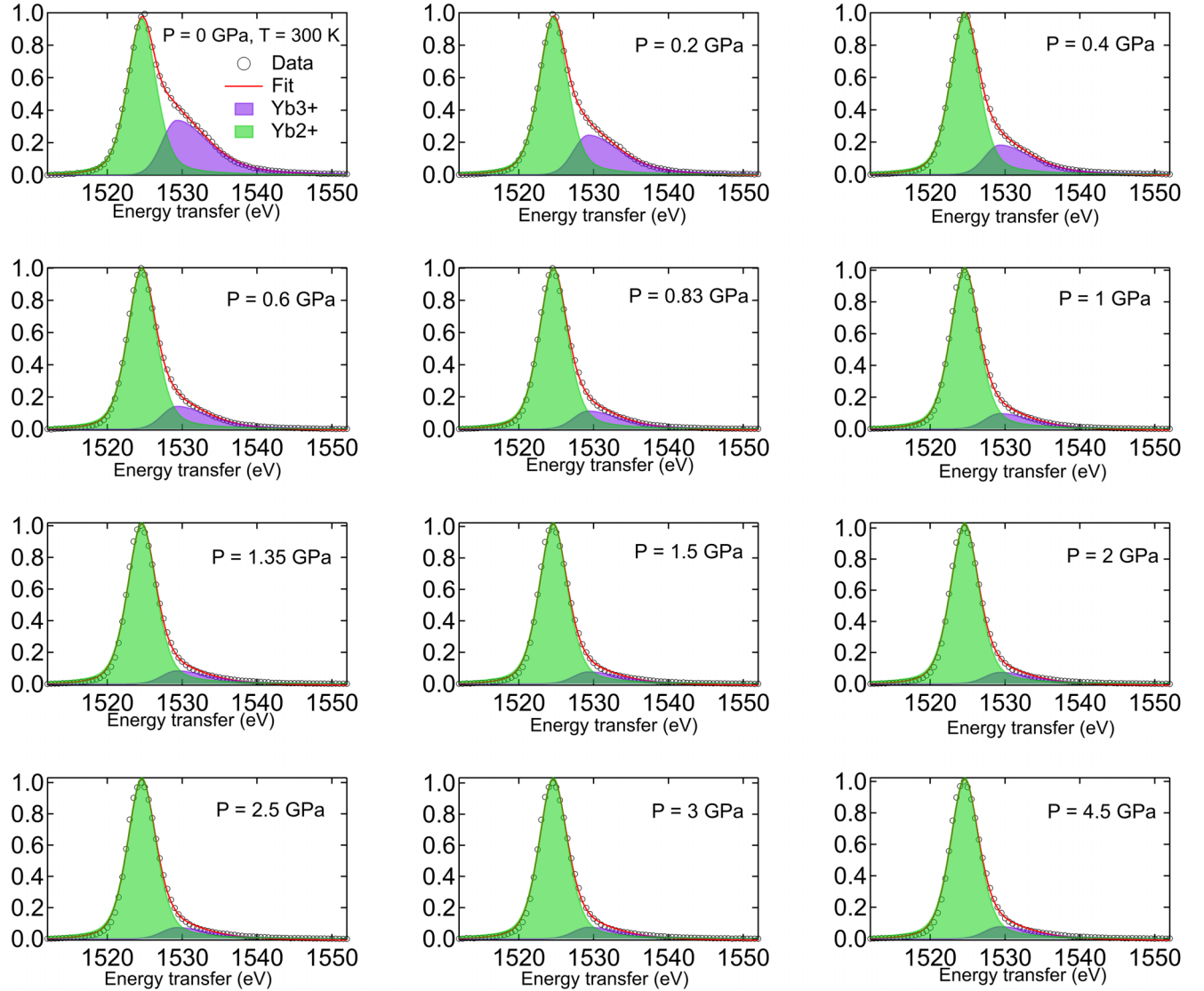


FIG. 7. Results of the fit analysis of the RXES spectra for all measured pressures at $T = 300$ K.

successfully reproduced within the Kondo volume-collapse model [61–63] in which a rapid pressure-induced increase of the Kondo temperature drives the transition. Our experimental findings identify strongly correlated YbPd as an interesting system to test the applicability of the Kondo volume-collapse model in describing pressure-induced changes resulting in a pronounced hardening-to-softening transition.

V. CONCLUSION

In conclusion, we have investigated the pressure dependence of the Yb valence in YbPd using Yb L_3 resonant x-ray emission spectroscopy. Our RXES data in the low-temperature charge ordering phase show a gradual increase in the mean valence with increasing pressure. In contrast, the RXES data at room temperature in the normal phase showed that the Yb valence decreases from 2.87 to 2.58 at 1.6 GPa and subsequently saturates with further increase in pressure. This unexpected and unusual evolution of Yb valence is coupled with an uncommon hardening-to-softening

phenomenon. This behavior is somewhat surprising and has not been reported in previous studies, likely because this region of the pressure-temperature phase diagram of YbPd (as well as other Yb-based mixed-valent systems) has remained largely unexplored. Most of the earlier P -dependent studies focused on investigating the low-temperature behavior. By analogy with Ce, this pronounced minimum in the pressure dependence of the compressibility suggests proximity to a first-order transition in the (P, T) phase diagram of YbPd.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

APPENDIX A: PRESSURE-DEPENDENT XAS DIFFERENCE SPECTRA

Figures 5(a) and 5(b) show the difference plots, $XAS(p) - XAS(p = 0)$, corresponding to Figs. 2(a) and 2(b). The spectral shape of the difference spectra as a function of pressure is very similar at both temperatures. More importantly, these difference plots do not reveal any significant change in spectral shape with increasing pressure within the Yb^{2+} and Yb^{3+} final states. In particular, the relative intensity between the peaks at ~ 8941 and ~ 8943 eV remains essentially constant with increasing pressure, as shown more clearly in Fig. 5(c), where the difference spectra are normalized to the intensity of the peak at ~ 8941 eV. The apparent changes in the shape of the Yb^{2+} structure seen in Fig. 2(b) arise from the overall increase in the Yb^{2+} intensity, rather than from any intrinsic change in the electronic structure.

APPENDIX B: FIT ANALYSIS OF THE RXES SPECTRA

Figures 6 and 7 show results of the fit analysis of the RXES spectra for all measured pressures at 30 and 300 K,

respectively. The spectra were fitted using two pseudo-Voigt functions $f_2(x; E_2)$ and $f_3(x; E_3)$ for the Yb^{2+} and Yb^{3+} emission lines, respectively. The total fitting function is given by $f(x; E) = f_2(x; E_2) + f_3(x; E_3)$, where

$$f_2(x; E_2) = a_0 \left[(1 - a_1) \exp \left[-\ln 2 \left(\frac{x - E_2}{a_2} \right)^2 \right] + \frac{a_1}{1 + \left(\frac{x - E_2}{a_2} \right)^2} \right],$$

$$f_3(x; E_3) = b_0 \left[(1 - b_1) \exp \left(-\ln 2 \left(\frac{x - E_3}{b_2} \right)^2 \right) + \frac{b_1}{1 + \left(\frac{x - E_3}{b_2} \right)^2} \right].$$

x denotes the photon energy, and $E_2 = 1524.6$ eV and $E_3 = 1529.3$ eV are the peak positions of the Yb^{2+} and Yb^{3+} emission lines. $a_1 = 0.399$ and $a_2 = 2.275$ eV. The asymmetry of the Yb^{3+} line due to the onset of fluorescence at the absorption edge was accounted for by using the sum of a Lorentzian and a Gaussian line shape with different widths and relative weights below and above the emission line. For $x \leq x_0$, $b_1 = 0$ and $b_2 = 2.349$ eV, and for $x > x_0$, $b_1 = 0.569$ and $b_2 = 5.115$ eV. a_0 and b_0 were considered as fitting parameters, while the other parameters were held fixed at the same values.

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