Revisiting the heavy-fermion compound YbCdCu₄: Kondo temperature and the ground-state degeneracy

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YbCdCu₄ has been characterized as a heavy-fermion Kondo lattice system without long-range magnetic order. Thermodynamic and transport data of this compound have been partially analyzed, and the reported Kondo temperatures T_K are highly dependent on samples and estimation methods. In particular, the magnetic susceptibility $\chi(T)$ of this compound does not show the maximum corresponding to the Kondo resonance with a large degeneracy of the ground state, partly due to the sample quality. Thus, single crystals of YbCdCu₄ are grown to reinvestigate their physical properties by means of magnetization, specific heat, electrical resistivity, Hall resistivity, and thermoelectric power measurements. Unlike earlier studies, we found consistencies regarding the $T_K \sim 160$ K measurement estimated from thermodynamic and transport property measurements. The magnetic part of the specific heat C_m displays a broad maximum near 70 K, which follows well with the prediction made by the Coqblin-Schrieffer (C-S) model for whole J = 7/2 multiplets involved in the formation of the Kondo state. However, the local maximum in $\chi(T)$ does not match well with the C-S model prediction due to the crystalline electric field (CEF) effect, where the T_K value is still greater than the overall CEF splittings: $T_K > \Delta_{CEF}$. Electrical resistivity and Hall coefficient measurements indicate an extremum near ~70 K, while a negative minimum at ~130 K is observed in thermoelectric power.

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I. INTRODUCTION

Ytterbium (Yb)-based intermetallic compounds YbXCu₄ (X = Ag, Au, Zn, Cd, In, Mg, Pd, and Tl), with the cubic AuBe₅-type structure, present opportunities to study a broad range of phenomena, including valence fluctuation, heavy fermion, and long-range magnetic ordering, by changing X atoms in an isostructural condition [1-3]. An unusually large range of Kondo temperatures T_K is observed across the YbXCu₄ series: the highest $T_K \sim 855$ K for X = Mg is approximately 400 times larger than the lowest $T_K \sim 2$ K for X = Au [4]. YbInCu₄ exhibits the isostructural first-order valence transition at $T_v = 42$ K [5,6]. This valence transition is markedly absent from the other Yb-based compounds. The X = Au and Pd compounds order antiferromagnetically below 1 K with a moderately enhanced linear specific heat coefficient γ [1]. In these compounds, the Kondo and Ruderman-Kittel-Kasuya-Yosida interaction, in conjunction with crystalline electric field effects (CEFs), must be considered to understand their ground states [7]. The X = Mg, Zn, Cd, and Tl compounds exhibit no magnetic ordering, with each compound displaying properties in an intermediate state between the localized and intermediate valence regimes [2-4]; much less is known about the nature of these compounds, warranting further research.

Of great interest are the X = Ag and Cd compounds for their rather similar values of $\gamma \sim 200$ mJ/mole K² and zero temperature limit of magnetic susceptibility $\chi(T = 0) \sim$ 1.5×10^{-2} emu/mole [2], implying both compounds have nearly the same Kondo temperature. However, the Weiss temperature θ_p of YbCdCu₄ is roughly three times bigger than that of YbAgCu₄. The Kondo temperatures estimated from fitting the magnetic susceptibility data $\chi(T)$ to the Coqblin-Schrieffer (C-S) model [8] are $T_K \sim 200$ K for X = Cd and $T_K \sim 100$ K for X = Ag [4]. Note that unlike YbAgCu₄, there is no consensus for the Kondo temperature of YbCdCu₄ being highly dependant on sample quality and estimation methods. While a pronounced broad maximum in $\chi(T)$ appears at ~40 K in YbAgCu₄, a monotonic decrease with increasing temperature appears in YbCdCu₄ [2]. The susceptibility and specific heat maximum observed in YbAgCu₄ are clear signs of the Kondo effect with a large ground-state degeneracy; thus the maximum can be fit quantitatively to numerical predictions of the fully degenerate J = 7/2 C-S model [1-3,9,10]. For YbCdCu₄, however, the susceptibility maximum corresponding to the Kondo resonance has not been observed [2-4,11,12], and no specific heat analysis has been performed. In addition, only X = Ag has displayed a metamagnetic behavior at ~ 40 Tesla (T) in the magnetization curve, while X = Cd shows a gradual increase in magnetization up to 50 T [2,13]. The metamagnetic behavior of YbAgCu₄ can be explained by a theoretical model [14], reaching the valence crossover surface from a distance of approximately T = 40 K and 40 T. No maximum in the Hall coefficient, $R_H = \rho_H / H$, has been observed for the YbXCu₄ compounds with relatively small R_H , reflecting all good metals [15,16]. A large negative peak at $T \sim 50$ K is observed in the thermoelectric power (TEP) for X = Ag. This indicates a rather large Kondo temperature, implying that the Kondo effect acts on the full degeneracy of Yb ions [17]. For X = Cd, the TEP measurement has not been reported.

In this study we thoroughly investigated the physical properties of partially studied YbCdCu₄ by growing better quality single crystals. We have measured the magnetic susceptibility, magnetization, specific heat, electrical resistivity, Hall coefficient, and thermoelectric power to elucidate the Kondo contributions to these properties. We found both the magnetic susceptibility and electrical resistivity of YbCdCu₄ are sensitive to the growth conditions. Unlike previously studied samples, the low-temperature-grown single crystals clearly exhibit the local maximum in $\chi(T)$ associated with the Kondo effect. The Kondo temperature is estimated by applying the C-S model to the local maximum in magnetic susceptibility and specific heat. In particular, we resolve the inconsistency of earlier studies, where both X = Ag and Cd compounds show a similar $\chi(T = 0)$ and γ , while the Kondo temperature for X = Cd is twice that for X = Ag.

II. EXPERIMENTAL

Single crystals of YbCdCu₄ were grown by Cd-Cu flux. Further details of sample growth conditions for samples 1 (low-temperature growth) and 2 (high-temperature growth) are given below sample dependences. Power x-ray diffraction (XRD) measurements were performed at room temperature with Cu $K\alpha$ radiation using a Rigaku Miniflex diffractometer. Silicon powders as an internal standard were added to correct the instrument zero shift. The lattice parameter *a*, obtained from XRD analysis, is a = 7.146(3) Å for sample 1 and a = 7.133(8) Å for sample 2, which is consistent with an earlier report [2].

The chemical composition of the crystals was determined using an energy-dispersive x-ray (EDX) spectra obtained from a FEI Helios NanoLab 650 FIB/SEM system equipped with an EDAX Octane 60-mm² EDX detector, where the stoichiometry was checked at various locations. From EDX analysis it was found that the samples are homogeneous with atomic concentrations of Yb:Cd:Cu yielding 13.4:12.9:73.7, in line with a 1:0.96:5.50 composition for sample 1 and Yb:Cd:Cu yielding 13.3:13.7:73.0 in line with a 1:1.03:5.45 composition for sample 2. Note that the deviation of the stoichiometry from 1:1:4 is due to the experimental error. In order to have an absolute value of chemical composition, it is required to calibrate the EDX spectrum with known Yb-Cu and Cd-Cu binary compounds.

Magnetization was measured as a function of temperature, from 1.8 to 300 K, and magnetic field, up to 70 kOe, using a Quantum Design (QD) magnetic property measurement system. The specific heat was measured by relaxation technique down to 0.4 K in a QD physical property measurement system (PPMS). Four-probe AC resistivity measurements were performed down to 0.4 K in the QD PPMS. Hall resistivity measurements were performed in a four-wire geometry, for which the magnetic field directions were reversed to eliminate magnetoresistance effects due to voltage-wire misalignment. Thermoelectric power was measured in a homemade system with one heater and a two-thermometer setup.

III. RESULTS AND DISCUSSION

A. Sample dependences

Initially, YbCdCu₄ single crystals were prepared by a flux method similar to that described by Ref. [6]. High-purity Yb, Cd, and Cu with initial stoichiometry of



FIG. 1. (a) H/M of YbCdCu₄ along $H\parallel$ [111] for both highand low-temperature-grown samples. Top inset shows M/H of both cases. Bottom inset shows magnetization isotherms at 2 K for both cases. (b) Electrical resistivity of YbCdCu₄. See details in the text.

Yb_{0.125} (Cd_{0.28}Cu_{0.72})_{0.875} were placed in an alumina crucible and sealed in a quartz tube under partial Ar pressure. The ampoule was heated to 1100 °C and cooled to 800 °C at a rate of 10 °C/h, producing sample 2 (high-temperature growth), at which point the samples were separated from the flux by a centrifuge. The sample quality was checked by powder XRD, electrical resistivity, $\rho(T)$, and magnetic susceptibility $\chi(T)$ measurements. The $\chi(T)$ and $\rho(T)$ curves of sample 2 are displayed in Fig. 1.

The inverse magnetic susceptibility, $H/M = 1/\chi(T)$, for the sample 2 is shown in Fig. 1(a). The $1/\chi$ curve is linear in temperature between 100 and 300 K, following a Curie-Weiss law, $\chi(T) = C/(T - \theta_p)$, and shows a deviation from the linearity below 50 K. The values of the effective moment and Weiss temperature, obtained from the fit, are $\mu_{\text{eff}} = 4.4 \ \mu_B$ and $\theta_p = -100$ K, respectively, which are somewhat smaller than those of an earlier report [4]. The obtained effective moment is consistent with the theoretical value of $4.54 \ \mu_B$ for a free Yb³⁺ (J = 7/2) ion. As shown in the inset of Fig. 1, $\chi(T)$ increases with decreasing temperature continuously below a hump at around 30 K. Sample 2 is consistent with all previous magnetic susceptibility measurements, both single and polycrystalline YbCdCu₄ [2–4,11,12], where no clear maximum is observed in $\chi(T)$. Note that $\chi(0)$ and the characteristic temperature of the local maximum in Fig. 1 cannot be accurately determined due to the upturn below 20 K; this weak feature in earlier studies may result in large error bars for the characteristic temperature. Figure 1(b) shows the temperature dependence of the electrical resistivity. $\rho(T)$ of sample 2 indicates a metallic behavior at high temperatures, with a local maximum at around 60 K and a sharp rise in resistivity below the minimum around 20 K.

We suspect that the low temperature upturn below the hump in $\chi(T)$ is due to extrinsic effects such as impurities and disorder. Thus, various flux ratios and temperature profiles (cooling rate and decanting temperature) were used to minimize these extrinsic effects. Single crystals of YbCdCu₄ with a greatly reduced Curie-like tail in $\chi(T)$ can be obtained by decreasing Yb concentration and slightly increasing Cd concentration. Based on several attempts, sample 1 (low-temperature growth) was prepared with a nominal stoichiometry of Yb_{0.05} (Cd_{0.4}Cu_{0.6})_{0.95} and heated to 800 °C, maintained at this temperature for 5 h, then cooled to 700 °C at a rate of 1 °C/h. The grown single crystal has a tetrahedral shape with mirrored surfaces. Note that the cooling rate in the range of 1 °C/h to 10 °C/h produces a similar quality of samples, where $\chi(T)$ shows the local maximum.

The $\chi(T)$ and $\rho(T)$ curves of sample 1 are displayed in Figs. 1(a) and 1(b), respectively. Unlike sample 2, the $\chi(T)$ of sample 1 clearly shows a broad maximum around 50 K before reaching a constant value $\chi(0) \sim 1.01 \times 10^{-2}$ emu/mole at low temperature. The observed maximum is reminiscent for a Kondo lattice system with a large degeneracy: not only the CEF ground-state doublet, but also some of the excited CEF states or even the whole multiplet is involved in the formation of Kondo resonance, leading to a maximum in magnetic susceptibility and specific heat [8]. From the fit of $1/\chi(T)$ to the Curie-Weiss law above 150 K, $\mu_{\rm eff} = 4.4 \ \mu_B$ and $\theta_p = -130$ K are obtained. The effective moment of sample 1 is close to that of sample 2, while θ_p of sample 1 is bigger than that of sample 2. At T = 2 K, the magnetization isotherms M(H) of both samples 1 and 2 show linear field dependencies up to 70 kOe. The M(H) value of sample 2 at 70 kOe is roughly twice bigger than that of sample 1, as shown in the inset of Fig. 1(a). At high temperatures $\rho(T)$ of sample 1, displayed in Fig. 1(b), is similar to that of sample 2. However, the resistivity of sample 1 falls rapidly below 70 K and passes through a minimum at around 10 K before showing a single-impurity Kondo behavior. The $\rho(T)$ of both samples seems to follow Kondo lattice behavior with different scattering processes.

It has been shown for YbXCu₄ (X = In and Ag) [6] that the composition and concentration of the flux do not appear to be critical parameters for the ground-state physical properties. However, for YbCdCu₄, the growth condition, especially Yb and Cd concentration, affects the temperature dependence of the magnetic susceptibility and electrical resistivity. The lattice parameter *a*, obtained from XRD analysis, indicates basically no difference between samples 1 and 2 within the



FIG. 2. (a) Electrical resistivity of LuCdCu₄. Inset shows M/H at H = 70 kOe along [111] direction. (b) Specific heat $C_p(T)$ of LuCdCu₄. Inset shows C_p/T vs T^2 plot. The solid line represents the fit curve by $\gamma T + \beta T^3$.

resolution of the measurements. Based on the observations, it seems to be that the growth condition for sample 1 has low impurities or disorder. Therefore we used YbCdCu₄ single crystals prepared by the condition for sample 1 to measure specific heat, Hall resistivity, and thermoelectric power. The single crystals of nonmagnetic analog LuCdCu₄ are also obtained using the similar growth condition as sample 1.

B. LuCdCu₄

The physical properties of the nonmagnetic analog LuCdCu₄ are consistent with a paramagnetic metal. $\chi(T)$ of LuCdCu₄, taken at H = 70 kOe along the [111] direction, weakly depends on temperature with a small magnitude as shown in the inset of Fig. 2(a). The low temperature upturn below 50 K is likely due to the presence of paramagnetic impurities. Figure 2(a) shows $\rho(T)$ of LuCdCu₄, where $\rho(T)$ follows an ordinary metallic behavior (e.g., $d\rho/dT < 0$). The specific heat, $C_p(T)$, of LuCdCu₄ as a function of temperature



FIG. 3. $C_p(T)$ for YbCdCu₄ at H = 0 (open symbols) and 90 kOe (closed symbol). Inset shows C_p/T vs T^2 , where the solid line represents the fit to $\gamma T + \beta T^3$.

ture is present in Fig. 2(b). As the temperature increases, C_p increases monotonically without any signature of the phase transition and reaches a value close to the Dulong-Petit limit at 300 K. At low temperatures, the specific heat can be explained by considering electronic and phonon contributions: $C_p = \gamma T + \beta T^3$. The electronic specific heat coefficient γ and Debye temperature Θ_D , estimated from C_p/T vs T^2 plot shown in the inset, are 8.49 mJ/mole K² and 301 K, respectively. These values are consistent with the earlier report [2].

C. C-S model analysis

 $C_p(T)$ of YbCdCu₄ is displayed in Fig. 3. The absence of an anomaly in $C_p(T)$ confirms no long-range magnetic order at least down to 0.4 K. The plot of C_p/T vs T^2 , presented in the inset, indicates a large $\gamma \sim 110$ mJ/mole K² value. The obtained γ value is smaller than earlier studies ($\gamma = 180$ mJ/mole K²) [2,4]. The γ value obtained in this study naturally resolves an issue with an NMR investigation of YbXCu₄ (X = Au, Ag, In, Cd, Tl, and Mg) [4], where the variation of Knight shifts with various X roughly correlates with γ except for X = Cd. The deviation was caused by the anomalously large γ value of YbCdCu₄. The measurement of $C_p(T)$ at 90 kOe indicates no magnetic field dependence below 100 K.

The magnetic part of the specific heat, $C_m(T)$, plotted in Fig. 4(a), is estimated by subtracting $C_p(T)$ of LuCdCu₄ from $C_p(T)$ of YbCdCu₄. $C_m(T)$ clearly shows a well-defined maximum at ~70 K and converges below 5 K in a constant value $\gamma = 101$ mJ/mole K², estimated from the $C_m(T)/T$ curve below 5 K. Since the broad maximum implies the presence of the Kondo effect with a large ground-state degeneracy, N = 2J + 1, the $C_m(T)$ curve of YbCdCu₄ is compared with the Rajan's calculation for the C-S model. In Fig. 4(a) the values of $T_0 = 70$, 140, 220, and 300 K are used to generate the theoretical curves for J = 1/2, 3/2, 5/2, and 7/2, respectively. As shown in the figure, the numerical result for J = 7/2and $T_0 = 300$ K provides a better description of the $C_m(T)$



FIG. 4. (a) Magnetic part of the specific heat C_m (solid symbols) of YbCdCu₄, together with the Coqblin-Schrieffer model (Rajan's calculation) [8]. Values of $T_0 = 70$, 140, 220, and 300 K are used for J = 1/2, 3/2, 5/2, and 7/2 curves, respectively. Inset shows the magnetic entropy as a function of temperature. (b) Magnetic susceptibility of YbCdCu₄, together with the calculated results from the Coqblin-Schrieffer model. The magnetic susceptibility is scaled by the value $T_0 = 225$ K and $\chi(0) = 1.01 \times 10^{-2}$ emu/mole.

data than that for J = 5/2, suggesting that the ground-state degeneracy of YbCdCu₄ is N = 8. More precisely, the Kondo temperature in YbCdCu₄ compound is larger than the energy scale of overall CEF splitting, Δ_{CEF} : $T_K > \Delta_{CEF}$. Note that at high temperatures the difference between $C_m(T)$ and theoretical curve is likely attributed to an estimation of the phonon contribution (subtraction error). According to Ref. [18], the characteristic energy scale $T_0 = 300$ K can be converted into the Kondo temperature $T_K \sim 159.8$ K with N = 8, which is consistent with the value $T_K \sim 160$ K obtained from γ . The evolution of the magnetic entropy $S_m(T)$ as a function of temperature supports the high Kondo temperature. The inset to Fig. 4(a) shows the magnetic entropy obtained by integrating C_m/T as a function of temperature. $S_m(T)$ rises to $R \ln(4)$ by 95 K and does not reach $R \ln(8)$ at 300 K. The entropy evolution as a function of temperature suggests a rather weak CEF splitting, which seems to be in conjunction with the cubic crystal structure. The characteristic energy can also be estimated by taking twice the maximum temperature at which half of the full entropy $R \ln(8)$ is recovered [18], which leads to $T_K \sim 190$ K.

A further estimate for the characteristic energy scale can be gained from the magnetic susceptibility. The normalized magnetic susceptibility $\chi(T)/\chi(T=0)$ is compared with the theoretical results for the C-S model [8], where the value $T_0 = 225$ K and $\chi(0) = 1.01 \times 10^{-2}$ emu/mole are used to scale experimental data. As shown in Fig. 4(b), the numerical results of Ref. [8] (solid lines in the figure) do not provide an excellent description of the $\chi(T)$ curve of YbCdCu₄. At low temperatures the height of the maximum in $\chi(T)$ falls between J = 5/2 and 3/2, while at high temperatures $\chi(T)$ follows the theoretical curve with J = 5/2. In the previous study the model with J = 3/2 qualitatively captures the magnetic susceptibility at high temperatures [2]. Since the theoretical calculations do not provide a good description of experimental data, the Kondo temperature is estimated from the zero temperature limit of the magnetic susceptibility value $\chi(0) = 0.0101$ emu/mole, which gives $T_K \sim 172$ K. This T_K value is consistent with that obtained from the specific heat analysis. The Wilson ratio is estimated from $\chi(0)$ and γ to be ~ 1 . We note that $T_0 = 225$ K obtained from the magnetic susceptibility fit can be converted into the Kondo temperature $T_K \sim 120$ K with J = 7/2, smaller than that obtained from $\chi(0)$. For YbCdCu₄, the obtained T_K value is somewhat smaller than that determined by previous thermodynamic, transport, and photoemission spectra results [4,12,19,20].

In a Kondo lattice system, a development of the maximum in C_m has been explained well with the C-S model [21–26], while the maximum in $\chi(T)$ often deviates from the theoretical curves, mostly due to the finite CEF splitting. For YbCdCu₄, the reason that the C-S model for Yb³⁺ impurities does not provides such a good description of $\chi(T)$ seems to be due to the finite CEF splitting (however, still smaller than T_K), as evidenced by the NMR experiments [4]. It has been shown that the large increase in hyperfine fields below 140 K is attributed to a variation of CEF interactions as the system transformed into a mixed valence state. Note that the magnetic susceptibility of YbAgCu₄ is well explained by the fully degenerate C-S model with $T_0 = 150$ K. A similar situation is observed for YbT_2Zn_{20} (T = Fe, Ru, Os, Co, Rh, Ir) systems, where T = Fe is well interpreted by the fully degenerate C-S model, whereas the CEF gives rise to a gradual reduction of the effective ground-state degeneracy for remaining T [26]. Except for the fully degenerate groundstate case, the deviation is partly caused by the nature of the ground-state susceptibility that is sensitive to the ground-state wave functions. Therefore the proper ground-state wave functions should be taken into account to explain the observed magnetic susceptibility. Next we discuss the magnetization behavior between X = Ag and Cd. For X = Ag, a metamagneticlike enhancement of magnetization by applying magnetic field of 40 T has been observed and explained by the C-S model [2,13], while no such behavior has been observed for X = Cd. It is clear from the current investigation that the



FIG. 5. (a) Magnetic part of the electrical resistivity ρ_m of YbCdCu₄. Inset shows an enlarged plot of electrical resistivity at H = 0 and 90 kOe applied along [111] direction. (b) Hall coefficient $R_H = \rho_H/H$ of YbCdCu₄. Inset shows R_H vs $\chi(T) \times \rho_m$. Solid line is a guide to the eye. (c) Thermoelectric power S(T) of YbCdCu₄ at H = 0 and 90 kOe. Inset shows S/T vs T^2 plot.

magnetic susceptibility for X = Cd clearly indicates the local maximum around 30 K, and the γ value is roughly twice smaller than that for X = Ag. Thus it is expected that the field-induced metamagnetic behavior can also be observed for X = Cd by much higher external magnetic fields.

D. Transport properties

The temperature dependence of transport property measurements support the presence of Kondo interactions with a large characteristic energy scale. In Fig. 5(a) the temperature dependence of the magnetic part of the electrical resistivity $\rho_m(T)$ is plotted down to 0.4 K, where ρ_m is obtained by the subtraction of $\rho(T)$ of LuCdCu₄ from $\rho(T)$ of YbCdCu₄. On cooling from 300 K, $\rho_m(T)$ increases logarithmically, $\rho_m \propto -\log(T)$, reaches a maximum of the coherence peak at ~68 K, and then goes down into the coherent Kondo state. Below 10 K, $\rho(T)$ increases again (inset), following a single-ion Kondo behavior. Since the upturn depends weakly on magnetic field, it might not be attributed to magnetic origin. Due to the absence of the Fermi-liquid behavior, $\rho(T) = \rho_0 + AT^2$, we were not able to obtain the Kadowaki-Wood ratio.

Figure 5(b) shows the temperature dependence of the Hall coefficient, $R_H = \rho_H/H$, at H = 90 kOe. Note that Hall resistivity ρ_H is linear in fields up to 90 kOe for all measured temperatures. The R_H curve clearly indicates a broad peak structure at around 70 K, corresponding to the characteristic Kondo temperature, consistent with the resistivity result. At 300 K R_H approaches $-2.15 \times 10^{-12} \ \Omega \ \text{cm/Oe}$, which corresponds to 2.9×10^{22} carriers per cm³ in a simple oneband-model approximation. The obtained carrier density is consistent with an earlier study; however, their R_H indicates no broad feature near 70 K [2]. The negative sign of R_H suggests that transport properties are dominated by electronlike carriers. In the high-temperature incoherent regime, R_H is linear against $\chi \times \rho_m$, shown in the inset, as expected for the skew scattering mechanism. Thus, the local maximum in ρ_m and minimum in R_H can be ascribed to the beginning of the transition to the coherent regime.

The thermoelectric power, presented in Fig. 5(c), is measured with the heat current ΔT perpendicular to the [111] direction. Large absolute values, typical for heavy-fermion systems, with a negative sign over the whole temperature range are observed, as expected for an Yb-based Kondo lattice system [27]. A broad minimum develops at a temperature of $T \sim 130$ K with a magnitude of $S(T) = -45 \ \mu V/K$. The observation of more than one peak in S(T) for a Kondo lattice system has to be attributed to a superposition of both the Kondo and CEF effects. As only one single minimum is resolved in S(T) of YbCdCu₄, it can be assumed that the whole J = 7/2 is involved in the formation of the Kondo state. The minimum in S(T) is located at a much higher temperature than that of the resistivity and Hall results. The reason for this difference is not yet clear; however, one can conjecture it to be the complex interplay between finite CEF splitting and Kondo screening. For YbAgCu₄ the occurrence of a negative peak at $T \sim 50$ K in S(T) leads to a rather large Kondo temperature and indicates that the Kondo effect acts on the full degeneracy of Yb ions, assistant with the specific heat and magnetic susceptibility [17]. These observations imply that the Cu replacement results in an increase of the Kondo temperature from 100 K to 160 K with small CEF splittings of the J = 7/2 ground state. In addition, the In substitution drives $YbXCu_4$ (X = Cu and Ag) compounds out of the Kondo state to a mixed valence state. Below 4 K, the TEP of YbCdCu₄ extrapolates to zero, with an enhanced slope $S/T \sim -1.4 \,\mu V/K^2$, in accordance with a heavy Fermi-liquid ground state, shown in the inset of Fig. 5(c). A universal ratio of S/T and γ has been found for a number of metals [27], $q = (S/T)N_A e/\gamma$ with N_A as Avogadro's number and e as electronic charge. For YbCdCu₄, we found $q \sim 1.3$, in good agreement with this empirical law.

IV. SUMMARY

We have successfully synthesized single crystals of $RCdCu_4$ (R = Yb and Lu) and thoroughly characterized its thermodynamic and transport properties. By minimizing extrinsic effects such as impurities and disorder in R = Yb, we are able to observe the maximum in the magnetic susceptibility, typical for Kondo lattice systems with degenerate ground states. YbCdCu₄ is a paramagnetic Kondo lattice system which follows a Curie-Weiss behavior above 150 K, with the effective moment $\mu_{\rm eff} = 4.4 \, \mu_B$ and the Weiss temperature $\theta_p = -130$ K. YbCdCu₄ has a γ value of 110 mJ/mole K² and a nearly constant low-temperature susceptibility of χ (0) = 0.01 emu/mole, yielding a Wilson ratio $R \sim 1$. The temperature dependence of the magnetic specific heat C_m is well described by the degenerate J = 7/2 Coqblin-Schrieffer model (Rajan's calculation) with a characteristic temperature $T_0 = 300$ K, while the magnetic susceptibility curve deviates from it due to the finite CEF splittings (still Δ_{CEF} < T_0). Temperature dependences of the transport properties exhibit Kondo interactions, where the observed local extremum reflects the characteristic energy scales. Through comprehensive measurements of YbCdCu₄, we are able to obtain the Kondo temperature $T_K \sim 160$ K in a consistent way.

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- C. Rossel, K. N. Yang, M. B. Maple, Z. Fisk, E. Zirngiebl, and J. D. Thompson, Phys. Rev. B 35, 1914 (1987).
- [2] J. L. Sarrao, C. D. Immer, Z. Fisk, C. H. Booth, E. Figueroa, J. M. Lawrence, R. Modler, A. L. Cornelius, M. F. Hundley, G. H. Kwei, J. D. Thompson, and F. Bridges, Phys. Rev. B 59, 6855 (1999).
- [3] J. M. Lawrence, P. S. Riseborough, C. H. Booth, J. L. Sarrao, J. D. Thompson, and R. Osborn, Phys. Rev. B 63, 054427 (2001).
- [4] T. Koyama, M. Matsumoto, T. Tanaka, H. Ishida, T. Mito, S. Wada, and J. L. Sarrao, Phys. Rev. B 66, 014420 (2002).
- [5] I. Felner and I. Nowik, Phys. Rev. B 33, 617 (1986)

- [6] J. L. Sarrao, C. D. Immer, C. L. Benton, Z. Fisk, J. M. Lawrence, D. Mandrus, and J. D. Thompson, Phys. Rev. B 54, 12207 (1996).
- [7] A. Severing, A. P. Murani, J. D. Thompson, Z. Fisk, and C.-K. Loong, Phys. Rev. B 41, 1739 (1990).
- [8] V. T. Rajan, Phys. Rev. Lett. 51, 308 (1983).
- [9] M. J. Besnus, P. Haen, N. Hamdaoui, A. Herr, and A. Meyer, Phys. B: Condens. Matter 163, 571 (1990).
- [10] P. Schlottmann, J. Appl. Phys. 73, 5412 (1993).
- [11] K. Hiraoka, K. Kojima, T. Hihara, and T. Shinohara, J. Magn. Magn. Mater. **140-144**, 1243 (1995).
- [12] H. Anzai, K. Morikawa, H. Shiono, H. Sato, S.-I. Ideta, K. Tanaka, T. Zhuang, K. T. Matsumoto, and K. Hiraoka, Phys. Rev. B 101, 235160 (2020).
- [13] T. Graf, J. M. Lawrence, M. F. Hundley, J. D. Thompson, A. Lacerda, E. Haanappel, M. S. Torikachvili, Z. Fisk, and P. C. Canfield, Phys. Rev. B 51, 15053 (1995).
- [14] S. Watanabe, A. Tsuruta, K. Miyake, and J. Flouquet, J. Phys. Soc. Jpn. 78, 104706 (2009).
- [15] A. L. Cornelius, J. M. Lawrence, J. L. Sarrao, Z. Fisk, M. F. Hundley, G. H. Kwei, J. D. Thompson, C. H. Booth, and F. Bridges, Phys. Rev. B 56, 7993 (1997).
- [16] E. Figueroa, J. M. Lawrence, J. L. Sarrao, Z. Fisk, M. F. Hundley, and J. D. Thompson, Solid State Commun. 106, 347 (1998).
- [17] R. Casanova, D. Jaccard, C. Marcenat, N. Hamdaoui, and M. J. Besnus, J. Magn. Magn. Mater. 90-91, 587 (1990).

- [18] A. C. Hewson, *Kondo Problem to Heavy Fermion* (Cambridge University Press, Cambridge, England, 1993).
- [19] H. Shiono, S. Ishihara, K. Mimura, H. Sato, E. F. Schwier, K. Shimada; M. Taniguchi, S. Ideta, K. Tanaka, T. Zhuang, K. T. Matsumoto, K. Hiraoka, and H. Anzai, AIP Conf. Proc. 2054, 040013 (2019).
- [20] H. Sato, K. Hiraoka, M. Taniguchi, Y. Nishikawa, F. Nagasaki, H. Fujino, Y. Takeda, M. Arita, K. Shimada, H. Namatame, A. Kimura, and K. Kojima, J. Phys.: Condens. Matter 14, 4445 (2002).
- [21] H.-U. Desgranges and J. W. Rasul, Phys. Rev. B **32**, 6100 (1985).
- [22] H. Michor, K. Kreiner, N. Tsujii, K. Yoshimura, K. Kosuge, and G. Hilscher, Phys. B: Condens. Matter 319, 277 (2002).
- [23] H. Michor, S. Berger, M. El-Hagary, C. Paul, E. Bauer, G. Hilscher, P. Rogl, and G. Giester, Phys. Rev. B 67, 224428 (2003).
- [24] 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. USA 104, 9960 (2007).
- [25] M. Deppe, S. Hartmann, M. E. Macovei, N. Oeschler, M. Nicklas, and C. Geibel, New J. Phys. 10, 093017 (2008).
- [26] E. D. Mun, S. Jia, S. L. Bud'ko, and P. C. Canfield, Phys. Rev. B 86, 115110 (2012).
- [27] K. Behnia, D. Jaccard, and J. Flouquet, J. Phys.: Condens. Matter 16, 5187 (2004).