Magnetic, thermal, and transport properties on single crystals of antiferromagnetic Kondo-lattice Ce_2PdSi_3

S. R. Saha, H. Sugawara, T. D. Matsuda, Y. Aoki, and H. Sato
Department of Physics, Tokyo Metropolitan University, Hachioji-Shi, Tokyo 192-0397, Japan

E. V. Sampathkumaran
Tata Institute of Fundamental Research, Homi Bhabha Road, Colaba, 400005 Mumbai, India

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Magnetization, heat capacity, electrical resistivity, thermoelectric power, and Hall effect have been investigated on single-crystalline Ce_2PdSi_3. This compound is shown to order antiferromagnetically below Néel temperature (T_N) $\sim$ 3 K. The Sommerfeld coefficient far below T_N is found to be about 110 mJ/K^2 mol Ce, which indicates the heavy-fermion character of this compound. The transport and magnetic properties exhibit large anisotropy with an interplay between crystalline-electric-field (CEF) and Kondo effects. The sign of thermoelectric power is opposite for different directions at high temperatures and the ordinary Hall coefficient is anisotropic with opposite sign for different geometries, indicating the anisotropic Fermi surface. The CEF analysis from the temperature dependence of magnetic susceptibility suggests that the ground state is $|\pm\frac{1}{2}\rangle$. The first and the second excited CEF doublet levels are found to be located at about 30 and 130 K, respectively. The Kondo temperature is estimated to be the same order as T_N, indicating the presence of a delicate competition between the Kondo effect and magnetic order.

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I. INTRODUCTION

There has been considerable interest in understanding the interplay among the crystalline-electric-field (CEF) effect, the indirect exchange [Ruderman-Kittel-Kasuya-Yoshida (RKKY)] interaction among the $4f$ magnetic moments and the Kondo effect in Ce compounds, since these are the decisive factors of the physical properties in these compounds. It is therefore worthwhile to carry out careful investigation in new Ce compounds. With this motivation, we report here the results of magnetic susceptibility (χ), magnetization (M), heat-capacity (C), electrical-resistivity (ρ), thermoelectric-power (S), and Hall-coefficient (R_H) measurements on single-crystalline Ce_2PdSi_3, grown for the first time.

This compound has been reported to form in an AlB_2-derived hexagonal crystal structure and to exhibit Kondo effect.[1] The intensity of investigation in the RE_2XSi_3 (RE = rare earth, X = transition metal) series, crystallizing in the above-mentioned structure,[2] increased only in the recent years and these compounds have been reported to exhibit many unusual features in the magnetic, thermal, and transport properties (see, for instance, Refs. 1–10 and references therein). Gd_2PdSi_3 exhibits Kondo-lattice-like anomalies, e.g., a resistivity minimum above T_N accompanied by a large negative magnetoresistance.[3,4] These features, presumably due to a novel mechanism, are not common to Gd compounds. Ce_2CoSi_3 is a mixed-valent compound, a small La substitution for Ce induces a non-Fermi-liquid behavior in ρ.[5] Eu_2PdSi_3 exhibits two distinct magnetic transitions, with the possibility of quasi-one-dimensional magnetism for the high-temperature transition[6] and unusual magnetic characteristics.[7] Particularly considering that the Gd-based compound in the Pd series has been found to show many interesting anisotropic features,[3] it is tempting to carry out detailed studies on the single-crystalline Ce_2PdSi_3 as well. Previous magnetic, electrical-resistance, and heat-capacity measurements on this compound were performed only in the polycrystalline form[1] and no clear magnetic ordering could be detected. Thus the present studies extended to much lower temperatures, particularly on single crystals, serve as a first thorough characterization of the bulk properties of this compound.

II. EXPERIMENTAL DETAILS

Single crystals of Ce_2PdSi_3 have been prepared by the Czochralski pulling method using a tetra-arc furnace in an argon atmosphere. The single-crystalline nature has been confirmed by back-reflection Laue technique. The magnetic measurements were carried out with a Quantum Design superconducting quantum interference device (SQUID) magnetometer. The heat capacity was measured by a quasiadiabatic heat- pulse method using a dilution refrigerator. The electrical-resistivity and Hall-effect measurements have been performed by a conventional dc four-probe method in the temperature interval of 0.5–300 K. The thermoelectric-power data have been taken by the differential method using a Au-Fe (0.07%)-chromel thermocouple.

III. RESULTS AND DISCUSSIONS

Figure 1(a) shows the temperature dependence of the inverse magnetic susceptibility $\chi^{-1}(T)$, measured in a magnetic field $H = 1$ kOe for both $H//[10\bar{1}0]$ and $H//[0001]$. There is a large difference in the absolute values of χ for two geometries, apparently due to CEF effect. The effective magnetic moment (μ_{eff}) and the paramagnetic Curie temperature Θ_P are estimated from the high-temperature linear region to be about $2.60\mu_B$/Ce and 3.6 K for $H//[10\bar{1}0]$ and $2.65\mu_B$/Ce and -107 K for $H//[0001]$, respectively. These values of μ_{eff} are very close to that expected for a free trivalent Ce ion ($2.54\mu_B$). The large negative value of Θ_P for $H//[0001]$ and a small positive value for $H//[10\bar{1}0]$ are presumably due to CEF effect.[11] The large anisotropy in Θ_P may also indicate the existence of anisotropy in the exchange interaction which depends on the CEF level scheme. The expanded view of the temperature dependence of χ is shown in the inset of Fig. 1(a). There is no difference between the field-cooled and the zero-field-cooled measurements of χ down to 1.9 K within the experimental accuracy, indicating the absence of any spin-glass-like behavior. This fact is in contrast to the formation of spin-glass state in the isostructural U_2PdSi_3.[9] There is a peak in $\chi(T)$ at around 2.8 K for $H//[10\bar{1}0]$ and at 2.5 K for $H//[0001]$. These peaks can be ascribed to the antiferromagnetic ordering. The occurrence of the peak at slightly different temperatures for two directions might be due to the anisotropic field dependence of T_N for two directions, since the heat-capacity measurement in absence of a magnetic field shows a peak at around 3 K as shown below. Recent neutron-diffraction data on polycrystals also suggests the occurrence of antiferromagnetic (AF) ordering below 2.5 K in a sinusoidally modulated AF structure.[10]

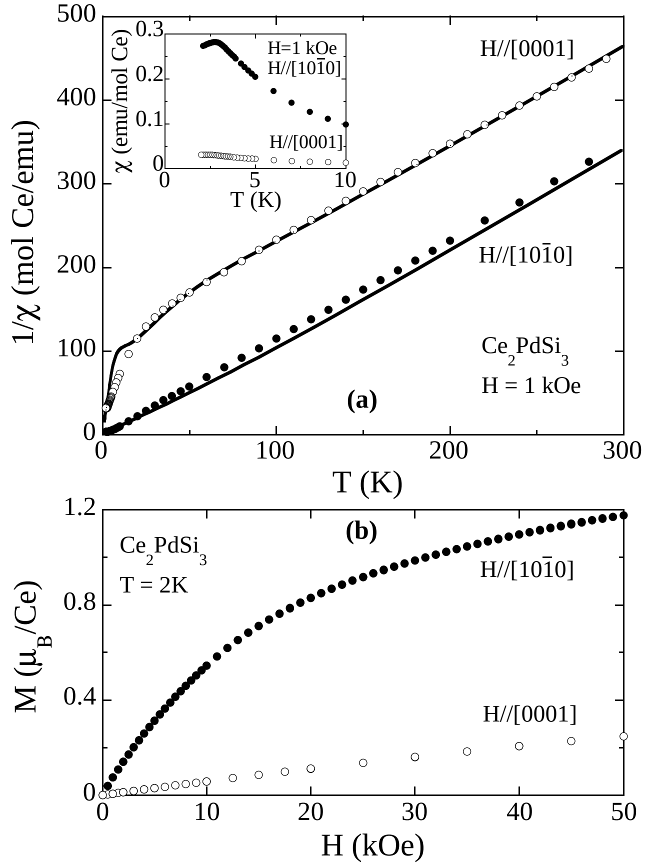

FIG. 1. (a) The inverse magnetic susceptibility versus temperature (2–300 K) for single crystals of Ce_2PdSi_3. The values calculated considering the CEF model (see text) are shown by the solid curves. The inset shows the expanded view of the magnetic susceptibility at low temperatures. (b) The isothermal magnetization behavior at 2 K for $H//[10\bar{1}0]$ and $H//[0001]$.

We have tried to analyse the $\chi(T)$ data using a CEF model, considering hexagonal site symmetry[10] of Ce in Ce_2PdSi_3. According to the Hutchings' notation,[12] the CEF Hamiltonian for $J = 5/2$ ion with the hexagonal point symmetry is given by

$$\mathcal{H} = B_2^0 O_2^0 + B_4^0 O_4^0, \tag{1}$$

where B_n^m and O_n^m represent the CEF parameters and the Steven's equivalent operators, respectively. The results of this CEF analysis using Eq. (1), employing the $\chi(T)$ data at the paramagnetic region, leads to $B_2^0 \simeq 10.1$ K, and $B_4^0 \simeq 0.11$ K. This set of parameters corresponds to a crystal-field level scheme with the three doublets $\mid \pm\frac{1}{2}\rangle$, $\mid \pm\frac{3}{2}\rangle$ and $\mid \pm\frac{5}{2}\rangle$ at around 0, 28 K (Δ_1), and 130 K (Δ_2), respectively. Accordingly, the calculated values of χ^{-1} are shown by the solid lines in Fig. 1(a), which indicates that the anisotropy in $\chi(T)$ is mainly induced by the CEF effect. However, there is a deviation of the calculated χ^{-1} from the experimental values, particularly at low temperatures. The following explanations can be offered to this deviation: According to Szytula *et al*,[10] Pd and Si atoms are random in this crystal structure (space group $P6/mmm$). Therefore this randomness or disorder between Pd and Si sites may produce local modification of the CEF effects due to the distribution of the CEF parameters. Alternatively, if Pd and Si are well ordered (space group $P6_3/mmc$, see Ref. 10), there are two different crystallographic environments for Ce ions,[8,13] in which case the CEF effect may be different for these two sites. All these factors are neglected in the present CEF calculations.

The isothermal magnetization at 2 K is shown in Fig. 1(b). M varies distinctly in different ways with the applied magnetic field for $H//[10\bar{1}0]$ and $H//[0001]$. This anisotropy in M is presumably due to the CEF effect. The magnetic moments at $H = 50$ kOe for two directions are ~ 1.18 and $\sim 0.25\mu_B$/Ce for $H//[10\bar{1}0]$ and $H//[0001]$, respectively. The larger magnetic moment for $H//[10\bar{1}0]$ indicates the a-b plane as the easy plane of magnetization, and the $|\pm\frac{1}{2}\rangle$ doublet as the ground state, in agreement with the CEF analysis from $\chi(T)$ described above. These two facts are also consistent with the proposed magnetic structure of Ce_2PdSi_3 based on neutron-diffraction experiment, i.e., the Ce magnetic moments lie in the a-b plane.[10]

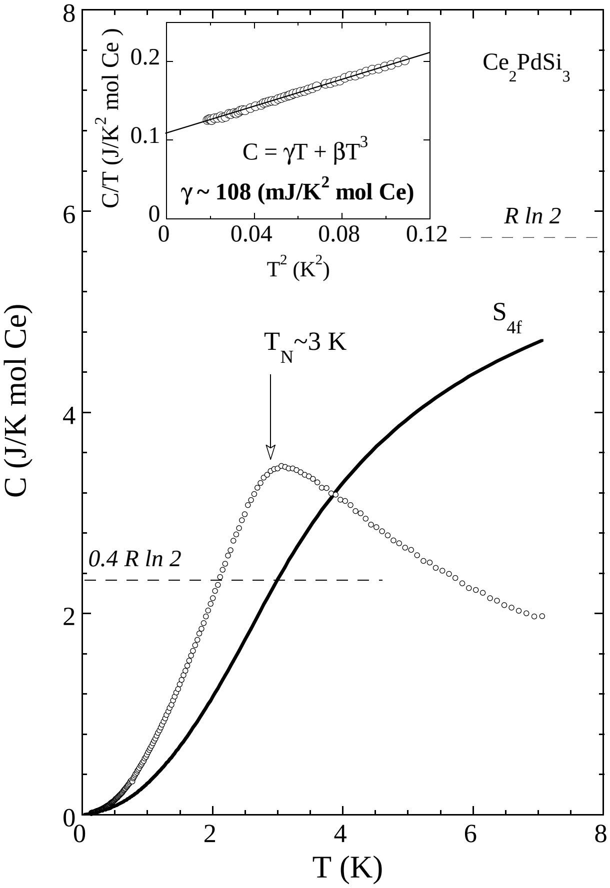

FIG. 2. The temperature dependence of heat capacity $C(T)$ for Ce_2PdSi_3. The magnetic ($4f$ contribution) entropy is presented by the solid line. The inset shows the C/T vs T^2 plot at temperatures well below T_N where phonon contribution can be neglected.

Figure 2 shows the temperature dependence of heat capacity, $C(T)$, for Ce_2PdSi_3. A peak in $C(T)$ at around 3 K indicates the existence of the antiferromagnetic ordering below $T_N \simeq 3$ K, supporting the conclusion from the $\chi(T)$ behavior. The transition is, however, not very sharp, which might be due to the presence of site disorders. The inset shows the C/T vs T^2 plot at temperatures far below T_N where phonon contribution is negligible. The linear coefficient of specific heat (γ), named as the Sommerfeld coefficient, estimated from this plot at such low temperatures is 108 mJ/K^2 mol Ce. At this temperature range the specific heat can be expressed as $C = \gamma T + \beta T^3$ with $\beta \simeq 862$ mJ/K^4 mol Ce and the parameter β mainly comes from the contribution of an antiferromagnetic-magnon part. For Y_2PdSi_3, the nonmagnetic compound serving as a reference for phonon contribution, the γ value is 4.5 mJ/K^2 molY.[1] The moderately large value of γ for Ce_2PdSi_3 suggests that even in the magnetically-ordered state Ce_2PdSi_3 may be classified as a heavy fermion. The solid line in Fig. 2 shows the magnetic entropy (S_{4f}) estimated from the $4f$ contribution (C_m) to C. C_m is obtained by employing the C values of Y_2PdSi_3 (Ref. 1) as a reference for the lattice contribution taking into account the difference of Debye temperatures of the two compounds by using the procedure suggested in Ref. 14. At T_N, $S_{4f} \simeq 2.3$ J/K mol Ce is $\sim 40\%$ of R ln 2 expected for a complete removal of the two fold degeneracy of a CEF ground-state doublet. This reduced entropy value might be due to the substantial Kondo-derived reduction of the Ce moments and/or a presence of short-range correlations above T_N.[15] Tentatively assuming a Kondo-derived reduction, the magnetic entropy S_{4f} ($\simeq 0.4$ R ln 2) at T_N yields the Kondo temperature $T_K \simeq 8$ K, according to the Bethe-ansatz for a spin-$\frac{1}{2}$ Kondo model (see Refs. 16 and 17). The theoretical calculations of $C(T)$ using $T_K \simeq 8$ K, however, cannot reproduce the experimental curve above T_N. This deviation indicates the presence of short-range-AF correlations above T_N; the presence of these correlations is, in fact, detected in a recent neutron-scattering experiment,[18] and a possible contribution from an excited CEF level (28 K) even at the measured temperature range.

The temperature dependence of resistivity $\rho(T)$ for Ce_2PdSi_3 with the current $J//[10\bar{1}0]$ and $J//[0001]$ as well as in polycrystalline Y_2PdSi_3 is shown in Fig. 3. ρ for both current directions gradually decreases with decreasing temperature down to about 20 K showing a broad hump around 100 K; below 20 K, there is a weak upturn giving rise to a minimum at around 20 K, followed by a drop below 8 K. The $\rho(T)$ in Y_2PdSi_3 shows usual metallic behavior, however there is a drop in ρ below 6 K, presumably attributable to the presence of traces of the superconducting phase YPdSi.[1] As known for many other Ce alloys, the broad hump in Ce_2PdSi_3 can be ascribed to the combined effect of CEF and Kondo effect. The magnetic contribution to the resistivity $\rho_m = \rho(Ce_2PdSi_3) - \rho(Y_2PdSi_3)$, obtained by using $\rho(T)$ data for $J//[0001]$ in Ce_2PdSi_3, is shown in the inset; the ratio of the slopes of the ρ_m vs ln T plot at high to low temperature turns out close to 35/3 confirming that the ground state is a doublet (see Ref. 19). It clearly reveals the presence of a peak at around 50 K which might be related with T_K enhanced by CEF effect as proposed by Hanzawa *et al.*[20] Correspondingly, there is also a broad hump in $S(T)$ (see Fig. 4). The minimum in $\rho(T)$ around 20 K is either due to the Kondo effect or a consequence of magnetic-precursor effect as noted for some Gd alloys.[4] It may be noted that the drop in ρ sets in at 8 K, much above the T_N, similar to $CePt_2Ge_2$,[21]

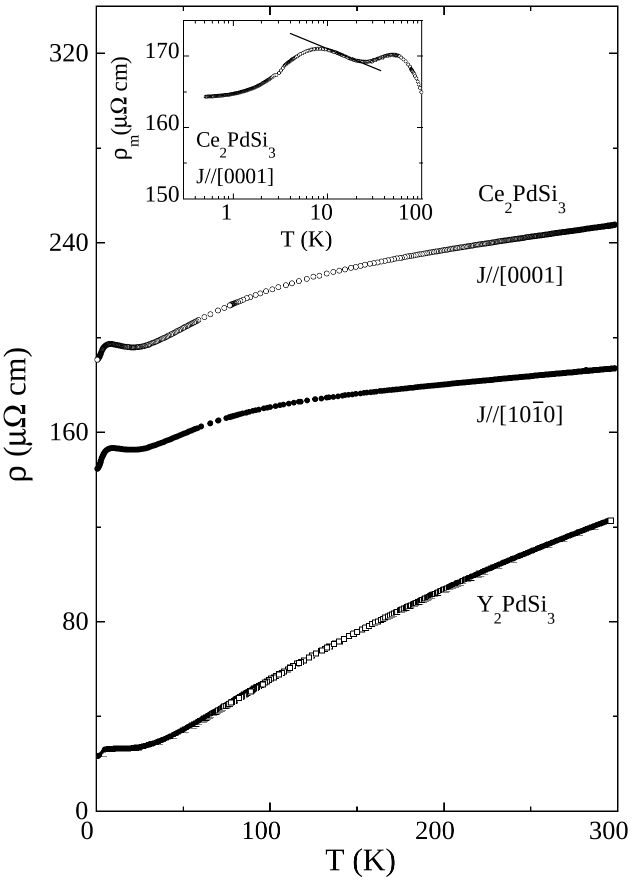

FIG. 3. The electrical resistivity (ρ) of single-crystalline Ce_2PdSi_3 as a function of temperature (0.5–300 K) for $J//[10\bar{1}0]$ and $J//[0001]$ and polycrystalline Y_2PdSi_3. The inset shows the magnetic contribution to the resistivity as a function of ln T.

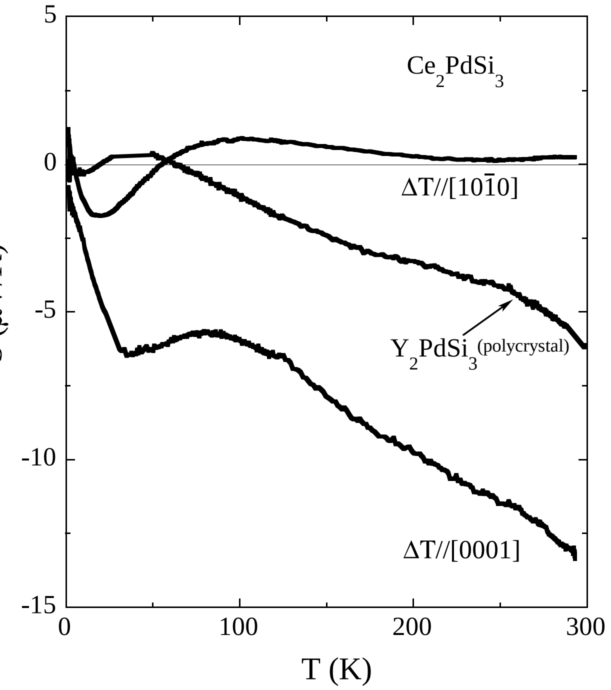

FIG. 4. The thermoelectric power as a function of temperature for two different directions of thermal gradient in Ce_2PdSi_3 single crystals, along with the data for polycrystalline Y_2PdSi_3.

and such a feature in magnetically ordered Kondo lattices arises from a combination of indirect exchange interaction and the Kondo effect.[22] There is a small difference in absolute values for two geometries, which might be due to the combined effect of anisotropy in the Fermi surface and preferably oriented microcracks. It may also be remarked that the residual resistivity is large even for the single crystal, which might be due to a presence of crystallographic (Pd-Si) disorder or a combined effect of disorder and a dominance of Kondo contribution even in the magnetically ordered state.

Figure 4 shows the temperature dependence of thermoelectric power (S) in Ce_2PdSi_3 as well as in Y_2PdSi_3 (polycrystal). In Ce_2PdSi_3, for the temperature gradient $\Delta T//[10\bar{1}0]$, S is positive at room temperature, then it gradually increases with decreasing temperature and shows a broad hump around 100 K. There is a change of sign around 50 K with a minimum around 17 K. On the other hand, for $\Delta T//[0001]$, S has a large negative value at room temperature and decreases with decreasing temperature showing a broad hump around 100 K. $S(T)$ for this direction also shows a minimum, however, at a temperature slightly higher than that for $\Delta T//[10\bar{1}0]$. Thus $S(T)$ in Ce_2PdSi_3 is highly anisotropic with the directions of the thermal gradient. The anisotropy in the Fermi surface might be one reason behind this anisotropy, since $S(T)$ in the isostructural Gd_2PdSi_3 is also anisotropic.[3] For nonmagnetic Y_2PdSi_3, $S(T)$ has a large negative value at room temperature and decreases gradually with temperature. In Ce_2PdSi_3, the broad hump can be attributed to the interplay between CEF and Kondo effect.[23] The origin of the minimum at $T_{min} \simeq 17$ K is not clear yet, however, the possible explanation may be the Kondo scattering in the CEF ground state or the growth of AF correlations as in the case of $CeAuAl_3$.[24] Tentatively assuming the Kondo-derived origin, $T_K \simeq 8$ K would be obtained using the relation $T_K \simeq \frac{1}{2}T_{min}$ that holds for $CeAl_2$ and $CeCu_2$ (see Ref. 15). However, the temperature dependence of C above T_N suggests that this estimation of T_K is a rough one, indicating that both Kondo effect and AF correlations may play a role for this minimum. For $\Delta T//[10\bar{1}0]$, $S(T)$ is similar to the behavior in the typical magnetically ordered heavy-Kondo compounds, e.g., $CeCu_2$ and $CeAl_2$,[15] though the overall temperature dependence of S in Ce_2PdSi_3 is rather weaker. If the crystallographic disorder is the dominant origin of the large residual resistivity, the Kondo contribution to $S(T)$ could be suppressed. Since, according to the Gorter-Nordheim rule, the thermoelectric power for more than one scattering mechanisms can be expressed as $S_{alloy} = [\rho_1 S_1 + \rho_2 S_2]/\rho$, where the subscripts 1 and 2 correspond to different scattering mechanism:[25] In the present case, 1 represents the Kondo scattering and 2 represents the other scatterings. Therefore a large ρ_2 can suppress the Kondo contribution S_1. The large negative thermoelectric power in Y_2PdSi_3 might arise from the $4d$ band of Pd, as in the case of the $3d$ band of Co in YCo_2.[26]

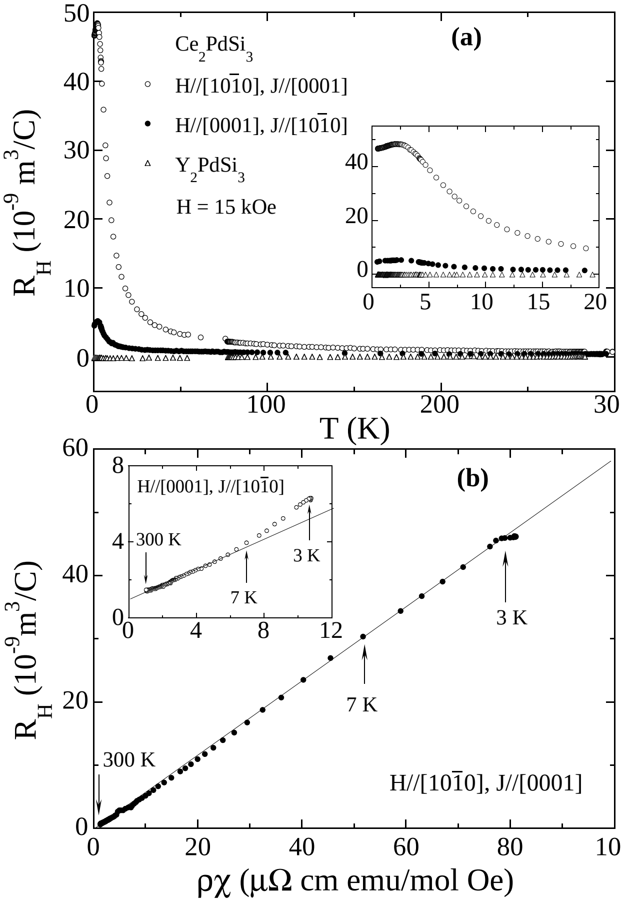

FIG. 5. (a) The Hall coefficient R_H (employing a magnetic field of 15 kOe) as a function of temperature for Ce_2PdSi_3 single crystals with two different orientations and for polycrystalline Y_2PdSi_3. The inset shows the expanded view of low-temperature $R_H(T)$. (b) R_H as a function of the electrical resistivity times magnetic susceptibility (with temperature as an intrinsic parameter).

The $S(T)$ curve for $\Delta T//[0001]$ at high temperatures is almost parallel to that of Y_2PdSi_3, therefore a significant effect of the Pd-$4d$ band on $S(T)$ even in Ce_2PdSi_3 cannot be ruled out. The temperature dependence of Hall coefficient (R_H) for H = 15 kOe, shown in Fig. 5(a), also reflects the anisotropic nature of this material. For both geometries (as labeled in the figure) R_H is positive at room temperature and increases gradually with decreasing temperature. At low temperatures R_H becomes highly anisotropic and shows a positive peak for both geometries, in the vicinity of T_N (see the inset). The large anisotropy observed in R_H is also reflected in the $\chi(T)$ data taken at H = 15 kOe (not shown), indicating that the anisotropy in R_H is of magnetic origin. The positive value of R_H at all temperatures and a positive peak at the vicinity of T_N are similar to those in the antiferromagnetic Kondo-lattice compound $CeAl_2$.[27] In contrast, R_H ($\simeq 0.9\times10^{-10}$ m^3/C at 300 K) in Y_2PdSi_3 is almost temperature independent. Clearly, there is a dominant $4f$ contribution in Ce_2PdSi_3. The Hall coefficient in magnetic materials like those in Ce compounds is generally a sum of two terms; an ordinary Hall coefficient (R_0) due to Lorentz force and an anomalous part arising from magnetic scattering (skew scattering);[27] in the paramagnetic state $R_H = R_0 + A\rho\chi$, where A is a constant. Using this relation, R_0 is estimated by plotting R_H versus $\rho\chi$ [Fig. 5(b)]. From Fig. 5(b), it is obvious that the plot is linear for both $H//[10\bar{1}0]$ and $H//[0001]$ in the paramagnetic state with a value of $R_0 \simeq -3.2\times 10^{-10}$ m^3/C and 1.0×10^{-9} m^3/C, and $A \simeq 7.4\times10^{-16}$ mol/C and 4.8×10^{-16} mol/C, respectively. This linear behavior indicates the presence of dominant skew scattering in Ce_2PdSi_3. In the vicinity of T_N, however, the data deviate from the high-temperature linear variation. R_0 of different sign with the anisotropic values for two geometries indicates the presence of anisotropy in the Fermi surface, in agreement with the $S(T)$ data.

IV. SUMMARY

Summarizing, we have investigated the magnetic behavior of recently synthesized Ce_2PdSi_3 in the single-crystalline form and the results show strong anisotropic behavior of the measured properties. The paramagnetic Curie temperature for $H//[10\bar{1}0]$ is positive, however, the value is negative for $H//[0001]$. The sign of the thermoelectric power is different for the two measured crystallographic orientations at high temperatures. Distinct features due to an interplay between CEF and Kondo effect have also been observed in the thermoelectric power and resistivity data. The ordinary Hall-coefficient is anisotropic with opposite sign for the two measured geometries. The results establish that this compound is an antiferromagnetic Kondo lattice with T_N= 3 K. The magnitude of T_K is also estimated to be of same order as T_N and this fact suggests a delicate competition between the

Kondo effect and indirect exchange interaction. Therefore it would be of interest to investigate this compound under high pressure.

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