# Resistivity and magnetic-susceptibility studies in the $RPd_2Al_3$ ($R$=La, Ce, Pr, Nd, and Sm) system

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In this paper we report magnetic susceptibility and resistivity studies on hexagonal $RPd_2Al_3$ ($R$=La, Ce, Pr, Nd, and Sm) compounds from 1.5 to 300 K. We observe that $NdPd_2Al_3$ shows antiferromagnetic ordering (AFM) at 6.5 K from the susceptibility and resistivity studies. On the other hand, $PrPd_2Al_3$ does not show any ordering down to 1.5 K but its susceptibility saturates below 15 K. The susceptibility measurements on $CePd_2Al_3$ reveal a peak at 2.8 K which is an indication of AFM in this compound. In addition, we have carried out resistivity and magnetization measurements on $SmPd_2Al_3$. Our resistivity studies show at least two magnetic transitions that are in agreement with the previous heat-capacity studies but unlike their resistivity measurements show only one transition at 12 K in the same compound. Further, the detailed measurements of magnetization ($M$) vs field ($H$) isotherms show evidence for two field-induced transitions in this compound at 2.5 K.

## I. INTRODUCTION

The discovery of the two heavy-fermion superconductors $UNi_2Al_3$ (Ref. 1) and $UPd_2Al_3$ (Ref. 2) has given fresh impetus to study superconductivity and magnetism in heavy-fermion compounds. This discovery has revived the search for similar systems in lanthanide compounds. Kitazawa *et al.*[3] have succeeded in finding $CePd_2Al_3$ as the antiferromagnetic ($T_N$=2.8 K) heavy-fermion ($\gamma$=380 mJ/mole $K^2$) compound in the equivalent lanthanide series. The susceptibility ($\chi$) of this compound shows Curie-Weiss dependence (corresponding to free $Ce^{3+}$) from 100 K to 300 K. This indicates that the Ce $4f$ electrons are localized at high temperatures. Recent neutron-scattering measurements on $CePd_2Al_3$ (Ref. 4) suggests that the observed antiferromagnetism could be due to simple collinear antiferromagnetic structure with moments in the $ab$ plane and the extrapolated moment value at 0 K is $0.38\pm0.05\mu_B$/Ce. It is found that the antiferromagnetism exhibited by this sample seems to be quite sensitive to defects and impurities. In fact the single crystal grown by Kitazawa's group[4] does not order down to 1.5 K while its $\gamma$ value reaches about 1 J/mole $K^2$.[5] They have suggested that such a behavior could arise due to defects or disordering in the crystal.

In their first report, Kitazawa *et al.*[3] reported an additional transition in $CePd_2Al_3$ around 6 K whose origin was not understood. In order to study the antiferromagnetism in this compound and its homologues (Pr, Nd, and Sm), we have measured the resistivity and magnetic susceptibility of $RPd_2Al_3$ ($R$=La, Ce, Pr, Nd, and Sm) series. To the best of our knowledge, the compound $NdPd_2Al_3$, has been synthesized by us for the first time. Although, x-ray photoemission spectroscopy and bremsstrahlung isochromat spectrocopy studies have been reported on $PrPd_2Al_3$,[6] a comprehensive study on the resistivity and susceptibility has not been made. In the case of $SmPd_2Al_3$, Kitazawa *et al.*[3] have observed three magnetic transitions at 12 K, 4.3 K, and 4.0 K using susceptibility and heat-capacity measurements. However their resistivity data showed only the transition at 12 K. In this paper, we report our high-resolution resistivity studies as well as magnetization studies on $SmPd_2Al_3$.

## II. EXPERIMENTAL DETAILS

All the samples of $RPd_2Al_3$ system were made by melting the individual constituents in an arc furnace under high purity argon atmosphere. The purity of the rare earth elements is 99.9% whereas the purity of Pd and Al is 99.99%. The samples were annealed at 900 °C for two weeks in a sealed quartz tube with He atmosphere. All the samples were found to have the hexagonal $P6/mmm$ ($UPd_2Al_3$ structure) and the lattice constants $a$ and $c$ (of La, Ce, and Sm) agrees with the previously published values.[7] The values of $a$ and $c$ of newly synthesized $NdPd_2Al_3$ along with the other members of the series is shown in Fig. 1. The magnetic susceptibility ($\chi$) was measured using a superconducting quantum interference device magnetometer (Quantum Design, U.S.) in a field of 5 kOe in the temperature range of 2 K to 300 K and a home built ac susceptometer[8] was used in the temperature range from 4.2 K to 1.4 K. The resistivity was measured using a four probe dc technique and the contacts were made using ultrasonic solder (with nonsuperconducting solder) on a cylindrical sample of 2 mm diameter and 10 mm length. The temperature was measured using a calibrated Si diode (Lake Shore Inc., U.S.) sensor. The sample voltage was measured with a Keithley nanovoltmeter with a current of 25 mA using a 20 ppm stable HP current source. All the data are collected using an IBM compatible PC/AT via IEEE-488 interface.

 

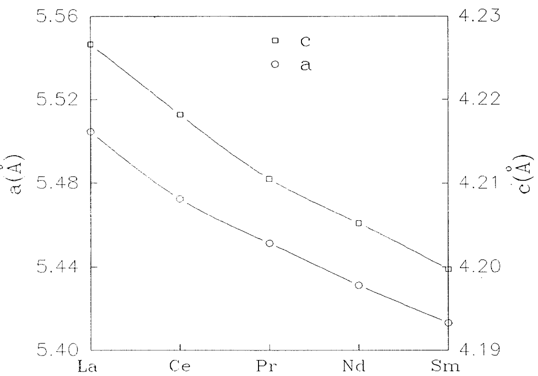


FIG. 1. Values of the lattice constants $a$ and $c$ of $RPd_2Al_3$ ($R$=La, Ce, Pr, Nd, and Sm) series at room temperature. The solid lines are guide to the eyes.

## III. RESULTS AND DISCUSSION

### A. Susceptibility studies

The temperature dependence of the inverse susceptibility ($1/\chi$) for $CePd_2Al_3$ in the temperature range 2 K to 300 K is shown in Fig. 2. The inset ($1/\chi$ vs $T$) shows the antiferromagnetic ordering at 2.8 K which agrees with the previously published data.[3] However, we do not see any additional transition at 6 K as reported in the previous study. The high temperature (100 K to 300 K) $\chi$ exhibits Curie-Weiss dependence and this data is shown as a continuous line in Fig. 2. The effective moment of Ce is $2.42\mu_B$ which is close to its free ion value. This suggests that the moments are well localized at high temperatures.

The $1/\chi$ data for $NdPd_2Al_3$ are shown in Fig. 3. The $\chi$ data shown as an inset clearly demonstrate the antiferromagnetic ordering at 6.5 K in this sample. The magnetization ($M$) vs field ($H$) isotherm data (up to $5T$ at 5 K and 6 K) for the same sample is shown in Fig. 4. The $M$ vs $H$ shows a clear positive curvature which is consistent with an antiferromagnetic ordering. The high temperature $\chi$ data (100 K to 300 K) shows Curie-Weiss dependence (shown as a continuous line) and the effective moment is found to be $3.6\mu_B$ which is close to the free Nd ion moment.

The high temperature $1/\chi$ data for $SmPd_2Al_3$ is shown in Fig. 5. The low temperature $1/\chi$ data (shown in the inset) reveal at least two magnetic transitions at 12 K and at 4.3 K. However there could be a third transition below 4.3 K whose value depends on the applied field. This data agrees with the previously published results.[9]

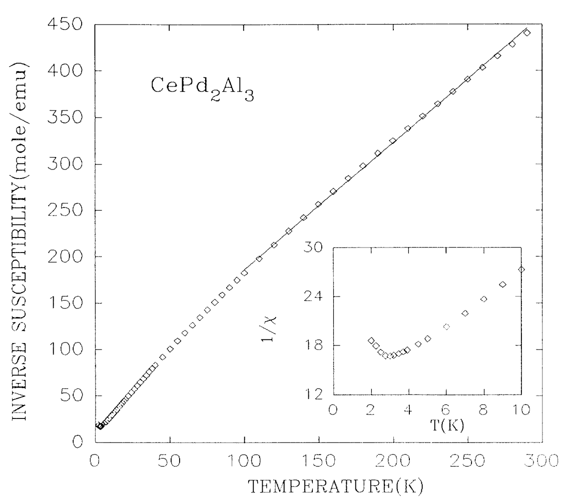


FIG. 2. Variation of inverse susceptibility ($1/\chi$) of $CePd_2Al_3$ from 2 K to 300 K. The solid line indicates the fit to Curie-Weiss law from 100 K to 300 K. The inset shows the antiferromagnetic transition at 2.8 K.

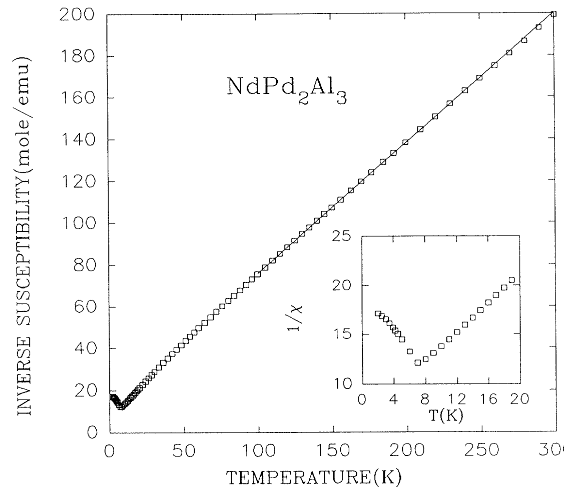


FIG. 3. Variation of inverse susceptibility ($1/\chi$) of $NdPd_2Al_3$ from 2 K to 300 K. The solid line indicates the fit to Curie-Weiss law from 100 K to 300 K. The inset shows the antiferromagnetic transition at 6.5 K.

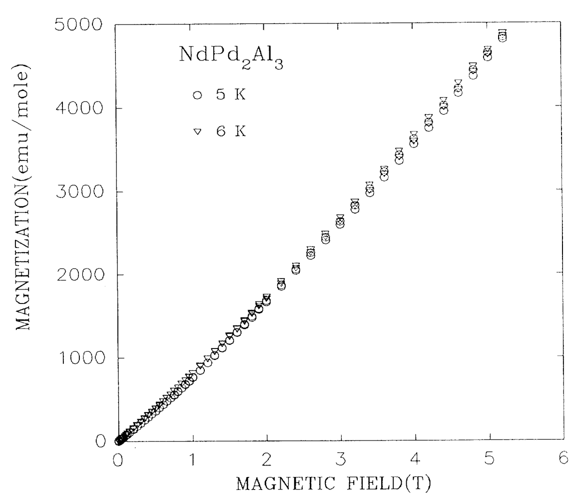


FIG. 4. Magnetization vs field isotherm of $NdPd_2Al_3$ at 5.0 K and 6.0 K.

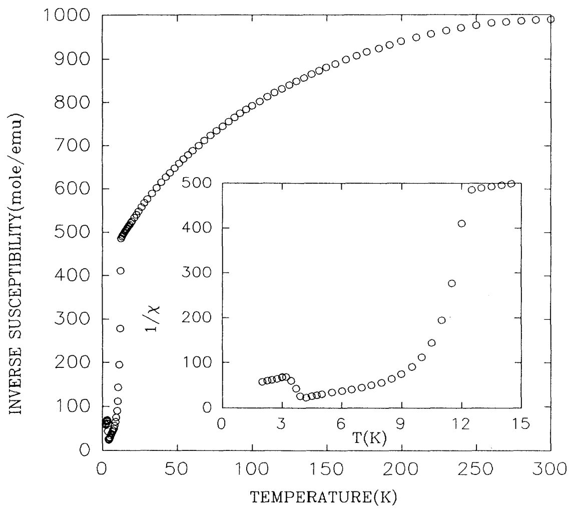


FIG. 5. Variation of inverse susceptibility ($1/\chi$) of $SmPd_2Al_3$ from 2 K to 300 K. The inset shows at least two magnetic transitions.

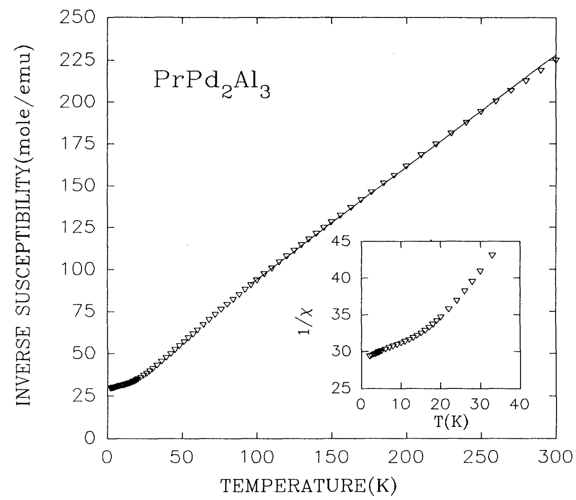


FIG. 7. Variation of inverse susceptibility ($1/\chi$) of $PrPd_2Al_3$ from 2 K to 300 K. The solid line indicates the fit to Curie-Weiss law from 100 K to 300 K. The inset shows the low temperature $\chi$ data.

However, our detailed measurements of magnetization ($M$) vs field ($H$) isotherm (shown in Fig. 6) at 2.5 K clearly indicate two field-induced magnetic transitions in this sample. The $M$-$H$ data at higher temperatures (5 K and 10 K) show small deviation from linearity. Our data coupled with the previous results tend to indicate that this compound exhibits a variety of magnetic transitions, the details of which need to be investigated further.

The $1/\chi$ data for $PrPd_2Al_3$ is shown in Fig. 7. This compound unlike the three previous compounds does not order down to 1.5 K. However, the $\chi$ tend to saturate below 15 K which is shown in the inset of Fig. 7. The measurements were repeated at fields as low as 200 Oe

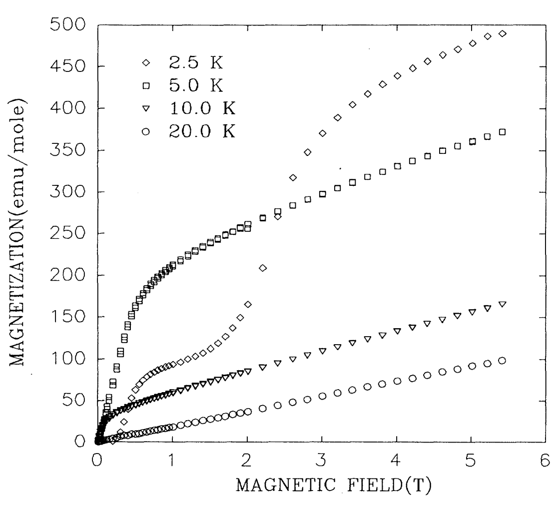


FIG. 6. Magnetization vs field isotherm of $SmPd_2Al_3$ at 2.5 K, 5.0 K, 10 K, and 20 K. The data at 2.5 K show two field-induced transitions.

which gave the same result. Hence one can rule out the possibility of magnetic impurities causing this effect. The temperature dependence of $\chi$ data fits well to the Curie-Weiss relation, $\chi = C/(T - \theta_p)$ (shown as a continuous line) from 100 K to 300 K. Here again the effective moment is found to be $3.45\mu_B$ which is nearly equal to the Pr free ion moment. The values $T_N$, $\theta_p$, C, $\mu_{\text{eff}}$, and $\mu_{\text{free}}$ for Ce, Pr, and Nd are given in Table I. We have also carried out ac susceptibility measurements on the Pr sample down to 1.5 K which do not reveal any magnetic ordering.

A preliminary analysis of the susceptibility has been carried out in terms of the crystal field Hamiltonian of the form,

$$H = B_2^0\, O_2^0 + B_4^0\, O_4^0 + B_6^0\, O_6^0 + B_6^6\, O_6^6 \qquad (1)$$

which is appropriate to the site symmetry $6/mmm$ of Pr ion. The effect of exchange interaction has been incorporated in terms of the molecular field approximation. Details of the procedure are given elsewhere.[10] Good fit to the $\chi$ data is obtained for several combinations of crystal field parameters. One such fit is shown in Fig. 8. The calculated anisotropic susceptibility along $a$ and $c$ axes for this combination of $B_n^m$'s is also shown. The anisotropy depends strongly on $B_n^m$'s. Therefore, the present analysis should be regarded as tentative un-

TABLE I. Magnetic properties of $R\text{Pd}_2\text{Al}_3$ ($R$=Ce, Pr, Nd, and Sm).

| $R$ | $\theta_p$ | $T_N$ | $C$ | $\mu_{\text{eff}}$ | $\mu_{\text{free}}$ |
|---|---|---|---|---|---|
| Ce | 36.0 K | 2.8 K | 0.73 emu/mole K | $2.42\mu_B$ | $2.54\mu_B$ |
| Pr | 40.4 K | ··· | 1.49 emu/mole K | $3.45\mu_B$ | $3.58\mu_B$ |
| Nd | 21.8 K | 6.7 K | 1.61 emu/mole K | $3.59\mu_B$ | $3.62\mu_B$ |

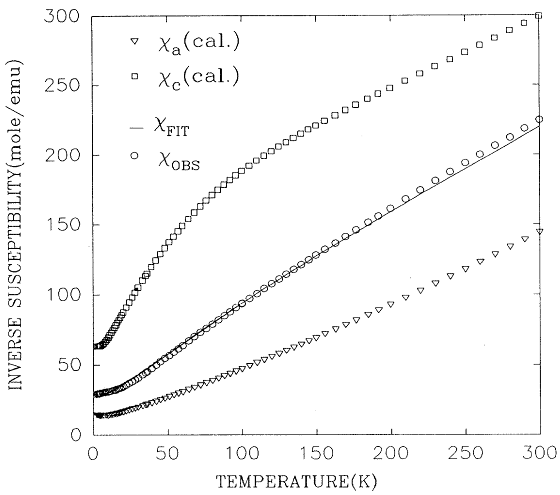


FIG. 8. Temperature vs $(1/\chi)$ fit for $PrPd_2Al_3$ from crystal field calculations. The calculated $\chi_a$ and $\chi_c$ are also shown.

til more data, in particular, $\chi$ values on single crystal, are available. The values of the $B_n^m$ parameters are $B_2^0 = -10.3$ K, $B_4^0 = -0.056$ K, $B_6^0 = 1.31 \times 10^{-2}$ K, and $B_6^6 = 4.36 \times 10^{-3}$ K. The value of the exchange parameter $\lambda = -18.8$ mole/emu.

### B. Resistivity studies

The temperature dependence of resistivity ($\rho$) for $CePd_2Al_3$ is shown in Fig. 9. The inset shows the magnetic transition below 3 K. This is in agreement with the

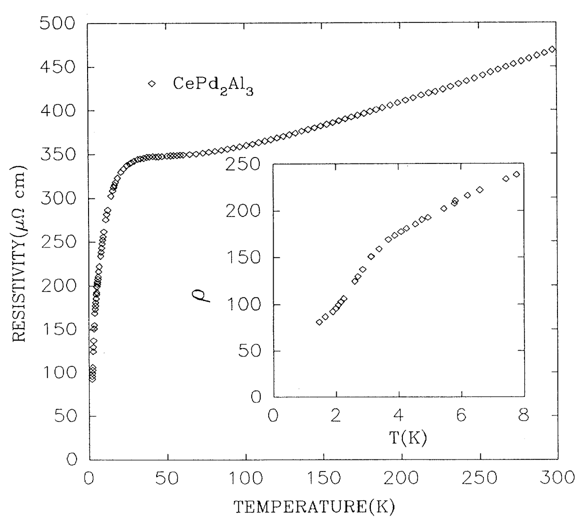


FIG. 9. Temperature dependence of resistivity $\rho(T)$ of $CePd_2Al_3$ from 1.5 K to 300 K. The inset shows the magnetic transition below 3 K.

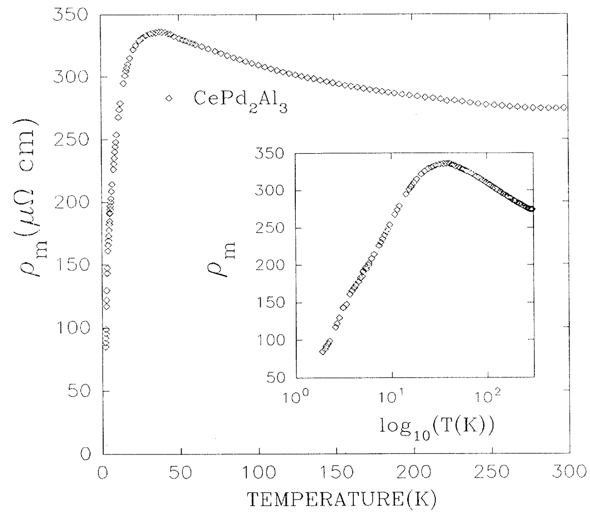


FIG. 10. Temperature dependence of the magnetic part of resistivity $\rho_m(T)$ of $CePd_2Al_3$ from 1.5 K to 300 K. The inset ($\chi$ vs $\log_{10}T$) shows maximum in $\rho$ around 30 K.

$\chi$ data. The $\rho$ data below $T_N$=3.0 K fits fairly well to a $T^{2.5}$ dependence. However, our $\rho$ data did not reveal any additional transition at 6 K which again is in agreement with our $\chi$ data. The magnetic part of $\rho$ (after substracting the $LaPd_2Al_3$ resistivity) at high temperatures is shown in Fig. 10. Here one observes a broad peak in $\rho$ around 30 K. The $\log_{10} T$ behavior (from 50 K to 300 K) suggest that this is a Kondo system and further, the large decrease of $\rho$ below the maximum at 30 K clearly exhibits the onset of coherence and the formation of a Kondo lattice as reported by Kitazawa *et al.*[3] The

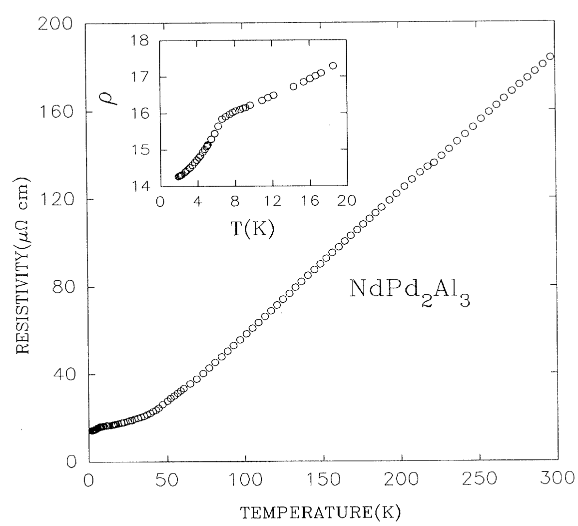


FIG. 11. Temperature dependence of the resistivity $\rho(T)$ of $NdPd_2Al_3$ from 1.5 K to 300 K. The inset shows the magnetic transition below 6.5 K.

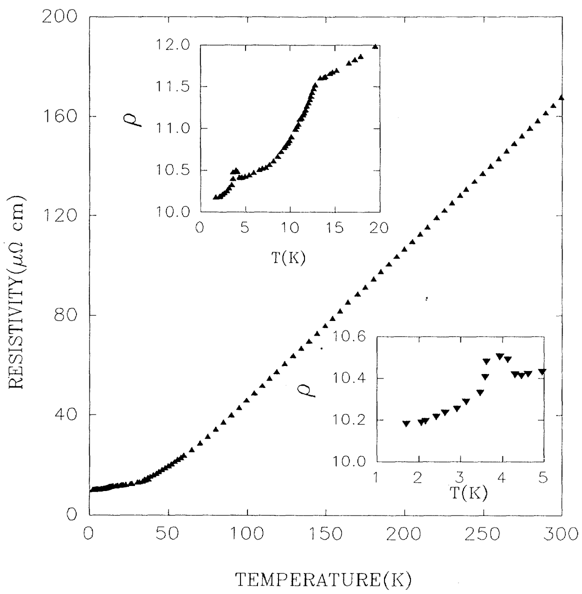


FIG. 12. Temperature dependence of resistivity $\rho(T)$ of $SmPd_2Al_3$ from 1.5 K to 300 K. The insets show at least two magnetic transitions at 12 K and at 4.3 K.

inset shows the maximum in $\rho$ at around 30 K.

The $\rho$ data for $NdPd_2Al_3$ are shown in Fig. 11. The inset shows low temperature $\rho$ of this compound. A change of slope occurs here at 6.5 K which corresponds to the antiferromagnetic transition in this compound. This is in agreement with our $\chi$ data. Although the magnetic ordering temperature is more than twice that of $CePd_2Al_3$,

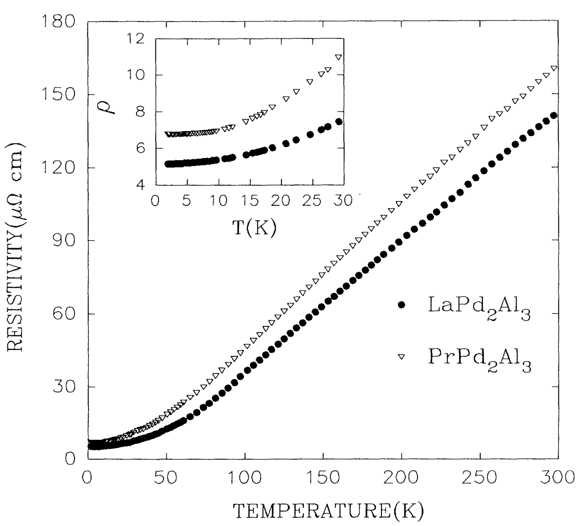


FIG. 13. Temperature dependence of resistivity $\rho(T)$ of $R$Pd$_2$Al$_3$ ($R$=La and Pr) from 1.5 K to 300 K. The inset shows the low temperature $\rho$ data.

TABLE II. Resistivity fit for $R$Pd$_2$Al$_3$ ($R$=La, Ce, Pr, Nd, and Sm) $\rho = \rho_0 + \rho_1 T^n$.

| $R$ | $\rho_0$($\mu\Omega$ cm) | $\rho_1$($\mu\Omega$ cm/K$^n$) | n | Range |
|---|---|---|---|---|
| La | 5.133 | $2.69 \times 10^{-3}$ | 2.0 | 1.5 K< $T$ < 30 K |
| Ce | 66.10 | 3.84 | 2.5 | 1.5 K< $T$ < 3 K |
| Pr | 6.739 | $9.3 \times 10^{-4}$ | 2.5 | 1.5 K< $T$ < 30 K |
| Nd | 14.12 | $3.96 \times 10^{-2}$ | 2.0 | 1.5 K< $T$ < 6.5 K |
| Sm | 10.29 | $1.94 \times 10^{-2}$ | 2.5 | 5 K< $T$ < 11 K |

the $\rho$ value is very much smaller than that of the Ce sample (Fig. 9). The large $\rho$ value of the latter is due to its Kondo behavior at high temperature. The $\rho$ data of the Nd sample can be fitted to a $T^2$ dependence from 1.5 K to 6.5 K (below $T_N$). This is in contrast to the $T^{2.5}$ seen in $CePd_2Al_3$.

The $\rho$ data for $SmPd_2Al_3$ is shown in Fig. 12. The inset shows low temperature $\rho$ of this compound. A change of slope occurs here at 12 K which corresponds to the first antiferromagnetic transition in this compound. The data also show another kink at 4.3 K and a rapid fall below 4.0 K which could correspond to two further magnetic transitions as reported from the suceptibility and heat-capacity data of Kitazawa.[9] However their $\rho$ data did not reveal the transitions at 4.3 K and 4.0 K. The $\rho$ data of the Sm sample can be fitted to a $T^{2.5}$ dependence from 5.0 K to 11 K (below the first $T_N$). Our $\chi$ data also indicate at least two transitions.

The $\rho$ data for $LaPd_2Al_3$ and $PrPd_2Al_3$ is shown in Fig. 13. The inset shows the low temperature $\rho$ of these two compounds. There is no anomaly seen in the resistivity down to 1.5 K. The $\rho$ data fits well to a $T^{2.5}$ dependence for the Pr sample and $T^2$ dependence for the La sample from 1.5 K to 30 K. At higher temperatures, the resistivities of these two samples are quite similar. The low temperature resistivity fit for the whole series $R$Pd$_2$Al$_3$ ($R$=La, Ce, Pr, Nd, and Sm) is given in Table II.

## IV. CONCLUSION

The detailed susceptibility and resistivity studies in $CePd_2Al_3$ shows an antiferromagnetic transition at 2.8 K which is in agreement with the previously published data.[3] However we do not see any evidence of second transition at 6 K. The antiferromagnetism at 6.5 K in $NdPd_2Al_3$ (which is not a heavy fermion) is reported. The $\chi$ data of $PrPd_2Al_3$ show a tendency to saturate below 15 K which can be explained by crystal field effects. The $SmPd_2Al_3$ sample shows three transitions as observed by Kitazawa *et al.*[9] However our high resolution $\rho$ data clearly show at least two transitions apart from the first antiferromagnetic transtion at 12 K. The magnetism exhibited by this series is quite intriguing and must await detailed neutron diffraction studies. Preliminary neutron diffraction studies on $CePd_2Al_3$ have already been reported.[4]

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