PHYSICAL REVIEW B VOLUME 45, NUMBER 2 1 JANUARY 1992-II

# Magnetic-susceptibility and electrical-resistivity measurements on *R*PdSn ($R$ = Ce–Yb) compounds

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(Received 7 February 1991; revised manuscript received 4 September 1991)

Magnetic-susceptibility and electrical-resistivity measurements have been carried out on equiatomic ternary compounds, *R*PdSn ($R$=Ce–Yb), in the temperature range 4.2 and 300 K. The compounds EuPdSn and YbPdSn have been synthesized. As-cast *R*PdSn ($R$=Ce–Yb) compounds crystallize in the orthorhombic TiNiSi-type structure. However, on annealing at 950 °C, Er- and Tm-containing compounds transform to hexagonal $Fe_2P$-type structure. Magnetic-susceptibility measurements reveal that the compounds with $R$=Ce, Sm, Eu, Gd, Tb, Dy, and Er order antiferromagnetically with Néel temperatures ($T_N$) of 7.5, 11, 13, 14.5, 23.5, 11.4, and 5.6 K, respectively. The compounds with $R$=Pr, Nd, Ho, and Tm are paramagnetic down to 4.2 K. Susceptibility of YbPdSn shows Curie-Weiss behavior between 300-150 K with $\mu_{\text{eff}}=1.45\mu_B$ and deviates considerably from it below 150 K. The electrical resistivity of all these compounds exhibits metallic behavior and shows a sharp drop at the respective Néel temperatures. The $T_N$ of the *R*PdSn series does not follow de Gennes scaling and, instead, peaks at Tb. This behavior of $T_N$ can be understood on the basis of crystalline-electric-field effects.

## I. INTRODUCTION

During the past few years, the intermetallic compounds containing rare-earth elements and the actinides have been the subject of intensive studies because of the interesting physical properties which some of these exhibit, such as the Kondo effect, superconductivity, spin fluctuation, valence instability, magnetic ordering with anomalously high ordering temperatures, and heavy-fermion behavior.[1–6] The family of compounds represented by the formula *RTX* ($R$= rare earth, $T$= transition metal, and $X$=Si, Ge, Al, Ga, As, In, and Sn) is one of the many families of ternary intermetallic compounds which have been reported in the literature.[7–15] Compounds of this family crystallize in a variety of structure types such as the cubic (LaIrSi, MgAgAs type), the hexagonal ($CaIn_2$, $Fe_2P$ type), the orthorhombic (TiNiSi, $CeCu_2$ type), the tetragonal (LaPtSi type), etc. Therefore, this family of compounds provides the possibility of a systematic study of the influence of the transition metals and the metalloids on the electronic structure of rare-earth ions and also the effect of the various crystal structures on the magnetic and transport properties of these compounds. Keeping this in mind we have initiated a systematic investigation of the *RTX* compounds. In an earlier paper, we presented the results of magnetic-susceptibility, electrical-resistivity, and heat-capacity measurements on CePdSn.[16] In this paper we report the results of crystal-structure, magnetic-susceptibility, and electrical-resistivity measurements on all the *R*PdSn ($R$=Ce–Yb) compounds. The Eu and Yb compounds of this series have also been synthesized. It is observed that the compounds with $R$=Ce, Sm, Eu, Gd, Tb, Dy, and Er order antiferromagnetically with Néel temperature ($T_N$), ranging from 7.5 K for Ce compound to 23.5 K for the Tb compound. However, the compounds with $R$=Pr, Nd, Ho, and Tm are paramagnetic down to 4.2 K. The susceptibility of CePdSn shows deviation from Curie-Weiss behavior below 100 K which is attributed to crystalline electric fields and Kondo effect. The Eu and Yb ions appear to be in a divalent state in EuPdSn and YbPdSn, respectively. The susceptibility of YbPdSn shows a Curie-Weiss behavior between 300 and 150 K with $\mu_{\text{eff}}=1.45\mu_B$ and deviates considerably from it below 150 K. The electrical resistivity of all these compounds exhibits metallic behavior and shows a sharp drop at the respective Néel temperatures. The Néel temperatures of the *R*PdSn series do not follow de Gennes scaling, and peak at Tb instead of at Gd. This behavior can be understood as a consequence of crystalline electric fields acting on the rare-earth ions.

## II. EXPERIMENT

The *R*PdSn ($R$=Ce–Yb) compounds were prepared by arc melting of the stoichiometric amounts of the constituent elements (purity 99.9% for rare-earth elements and 99.99% for Pd and Sn) in an inert atmosphere of argon. Powder x-ray diffraction studies were performed on all the samples using Siemen's x-ray diffractometer equipped with Cu $K_\alpha$ radiation. Magnetic-susceptibility measurements were made using the Faraday method in the temperature range of 4.2–300 K. Four-probe dc resistivity measurements were made in the temperature range of 4.2–300 K for which contacts to the samples were made with a conducting silver paint. In order to neutralize the effects due to thermal emf's, resistance measurements were made by passing the current in the forward and the reverse direction at each temperature and the average of the two was taken to compute the resistivities.

## III. RESULTS AND DISCUSSIONS

### A. Crystal structure

The *R*PdSn compounds with $R = \mathrm{Ce}$ to Sm, Gd, Tb, and Dy have been shown to crystallize in the orthorhombic TiNiSi-type structure (space group *Pnma*), while those with $R = \mathrm{Er}$, Tm, Lu, and Sc crystallize in the hexagonal $Fe_2P$-type structure (space group $P\bar{6}2m$).[7,9] The compound HoPdSn may stabilize in either of the two structure types.[7] Our powder x-ray diffraction (XRD) studies on the as-cast *R*PdSn compounds with $R = \mathrm{Ce}$, to Sm, Gd, Tb, and Dy, are in agreement with those reported in the literature.[9] However, the XRD studies on the as-cast samples of Ho, Er, and Tm prepared in our laboratory show that these too crystallize in the orthorhombic TiNiSi-type structure. A minority impurity phase (less than 8%) was detected in the Tm compound. The Eu and Yb compounds of the *R*PdSn series have been synthesized during the course of this investigation. These compounds are single-phase materials and their x-ray patterns are similar to those of orthorhombic *R*PdSn compounds. To study the effect of heat treatment on the structural stability, some selected compounds with the light rare-earth elements (e.g., Ce) and some with the heavy rare-earth elements (Tb, Dy, Er, and Tm) were annealed for one week at a temperature of 950 °C in an evacuated quartz capsule. Powder x-ray diffraction patterns of the annealed samples of ErPdSn and TmPdSn showed entirely different set of lines than those present in the as-cast samples, and could not be indexed on the basis of the orthorhombic TiNiSi-type structure. Instead, all the observed reflections could be indexed on the basis of the hexagonal $Fe_2P$-type structure. However, powder x-ray diffraction studies on annealed samples of CePdSn, TbPdSn, and DyPdSn reveal no change in the x-ray patterns compared to those of the as-cast samples. Thus the present work indicates that the structure of *R*PdSn compounds, with heavier rare-earth elements ($R = \mathrm{Ho}$, Er, and Tm) only, depends on the heat treatment and that ErPdSn and TmPdSn may also be stabilized either in the orthorhombic TiNiSi-type or in the hexagonal $Fe_2P$-type structure depending on the heat treatment.

The TiNiSi-type structure has four formula units per unit cell and the $Fe_2P$-type structure has three formula units per unit cell. The rare-earth atoms form a chain along the *c* axis in TiNiSi structure, while in the $Fe_2P$-type structure these atoms form a layered triangular lattice where each rare-earth atom is joined to another triangular lattice. In the TiNiSi-type structure of *R*PdSn compounds, the rare-earth element occupies the Ti site, Pd occupies the Ni site, and Sn occupies the Si site. A structural correlation exists[17] between the TiNiSi- and $Fe_2P$-type structures, both structures having the same coordination number.

The lattice parameters of the *R*PdSn compounds obtained by a least-squares fit of the observed *d* values are given in Table I. It is noted that the lattice parameter in the *a* direction across the series decreases at a faster rate compared to the lattice parameters in the other two directions. This is due to the fact that the rare-earth atoms form a zigzag chain in the *a* direction with relatively short separation which controls this lattice parameter. Figure 1 shows the cell volume, normalized to one formula unit of *R*PdSn, as a function of the rare-earth ion. The unit-cell volume of all the *R*PdSn compounds, except that of EuPdSn and YbPdSn, follows the lanthanide contraction expected for trivalent rare-earth ions. The deviation in the unit-cell volumes of EuPdSn and YbPdSn from the lanthanide contraction indicates that Eu and Yb ions are not in a trivalent state in these compounds but instead may be in the divalent state or in a mixed valent state.

TABLE I. Structure type, lattice parameters (*a*, *b*, and *c*), effective paramagnetic moment ($\mu_{\mathrm{eff}}$), paramagnetic Curie temperature ($\Theta_P$), magnetic ordering temperature ($T_N$), and de Gennes factor ($G'$) of the trivalent rare-earth ion (normalized to that of $Gd^{3+}$ or $Eu^{2+}$) in *R*PdSn compounds.

| Compound | Structure type | $a$ (Å) | $b$ (Å) | $c$ (Å) | $\mu_{\mathrm{eff}}$ ($\mu_B$) | $\Theta_p$ (K) | $T_N$ (K) | $G'$ |
|---|---|---|---|---|---|---|---|---|
| CePdSn | ortho | 7.526 | 4.742 | 7.931 | 2.67 | −68 | 7.5 | 0.011 |
| PrPdSn | ortho | 7.523 | 4.679 | 7.966 | 3.60 | −2 | | 0.051 |
| NdPdSn | ortho | 7.473 | 4.654 | 7.941 | 4.93 | −8 | | 0.116 |
| SmPdSn | ortho | 7.332 | 4.629 | 7.927 | NCW[b] | | 11 | 0.283 |
| EuPdSn | ortho | 7.498 | 4.682 | 8.039 | 8.27 | 5 | 13 | 1 |
| GdPdSn | ortho | 7.264 | 4.616 | 7.919 | 8.16 | −27 | 14.5 | 1 |
| TbPdSn | ortho | 7.196 | 4.595 | 7.893 | 10.17 | −16 | 23.5 | 0.667 |
| DyPdSn | ortho | 7.160 | 4.587 | 7.895 | 11.1 | −2 | 11.4 | 0.450 |
| HoPdSn | ortho | 7.111 | 4.577 | 7.881 | 11.07 | −7 | | 0.386 |
| ErPdSn | ortho | 7.094 | 4.570 | 7.872 | 9.51 | −0.3 | 5.6 | 0.162 |
| ErPdSn[a] | hexa | 7.419 | | 3.938 | | | | 0.162 |
| TmPdSn | ortho | 7.030 | 4.547 | 7.915 | | | | 0.074 |
| TmPdSn[a] | hexa | 7.495 | | 3.840 | 7.98 | −0.1 | | 0.074 |
| YbPdSn | ortho | 7.154 | 4.586 | 7.885 | 1.45 | −5 | | 0.020 |

[a] Sample annealed at 950 °C for a week.

[b] Non-Curie-Weiss behavior.

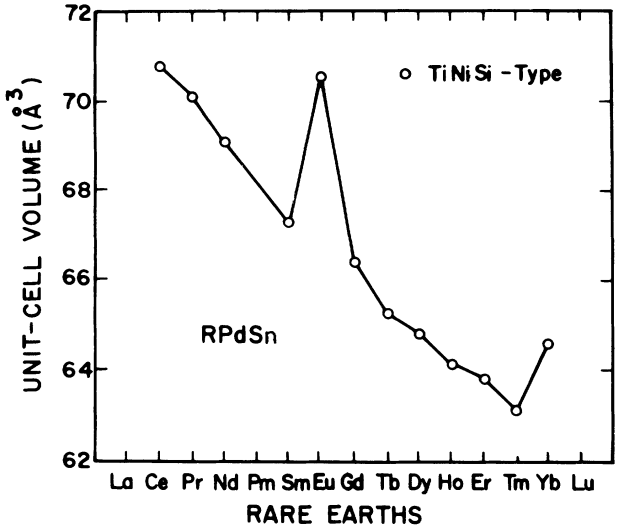

FIG. 1. Unit-cell volume of the *R*PdSn compounds as a function of the rare-earth element.

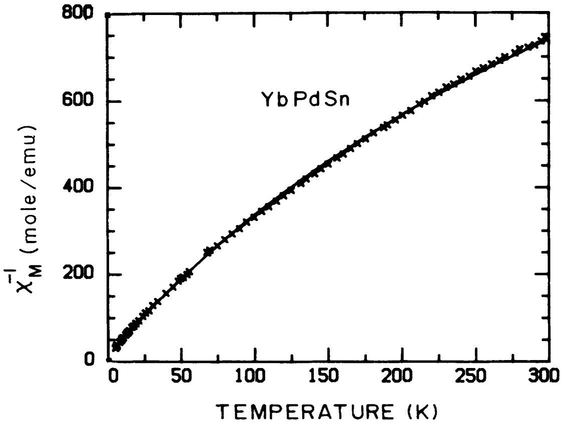

FIG. 3. Inverse magnetic susceptibility of YbPdSn as a function of temperature. Solid line shows the fit (see text).

### B. Magnetic susceptibility and electrical resistivity

Magnetic susceptibility of all the *R*PdSn compounds was measured in the temperature range of 4.2–300 K. The results are discussed below and largely summarized in Table I.

Among the *R*PdSn compounds, those with $R$ = Pr, Nd, Tm, and Yb remain paramagnetic down to 4.2 K while those with $R$ = Ce, Sm, Eu, Gd, Tb, and Er order magnetically. The plot of inverse susceptibility versus temperature for some of the *R*PdSn compounds is shown in Figs. 2–4. Except SmPdSn and YbPdSn, the susceptibility of all the *R*PdSn compounds follows Curie-Weiss behavior. The effective magnetic moment, $\mu_{\text{eff}}$, and paramagnetic Curie temperature, $\Theta_P$, obtained from a Curie-Weiss fit to the experimental data for all these compounds are given in Table I. The susceptibility of YbPdSn exhibits Curie-Weiss behavior between 150 and 300 K but deviates considerably from it below 150 K. The observed susceptibility from 4.2 to 300 K can be fitted to a Curie-Weiss term along with a temperature-independent susceptibility ($\chi_0$). The parameters obtained are $\mu_{\text{eff}} = 1.43\mu_B$, $\Theta_p = -4$ K, and $\chi_0 = 5.1 \times 10^{-4}$ emu/mole. The observed value of $\mu_{\text{eff}}$ of the Yb ion in YbPdSn is smaller than that of the free $\text{Yb}^{3+}$ ion ($\mu_{\text{eff}} = 4.54\mu_B$), suggesting that Yb ions are either in a divalent state or in an unstable valence state in this compound. This is consistent with the anomaly observed in the unit-cell volume of this compound in relation to that of other *R*PdSn compounds.

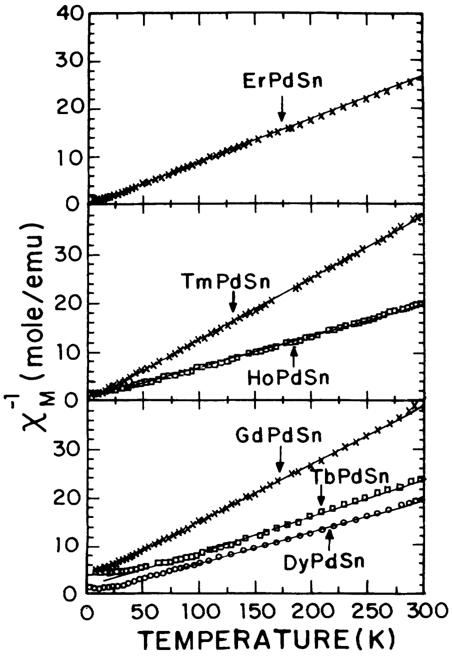

FIG. 2. Inverse magnetic susceptibility of *R*PdSn ($R$ = Gd, Tb, Dy, Ho, Er, and Tm) compounds as a function of temperature.

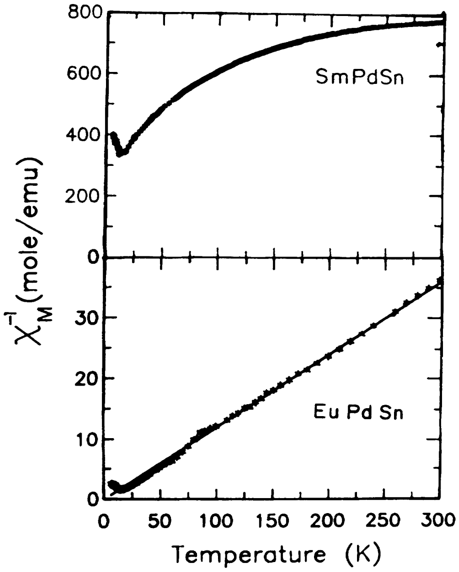

FIG. 4. Inverse magnetic susceptibility of SmPdSn and EuPdSn as a function of temperature.

Figure 5 shows the low-temperature susceptibility of some of the *R*PdSn compounds. A peak in the magnetic susceptibility of Ce, Sm, Eu, Gd, Tb, Dy, and Er compounds is observed at 7.5, 11, 13, 14.5, 23.5, 11.4, and 5.6 K, respectively, which is taken to be due to the antiferromagnetic ordering of the rare-earth moments in these compounds. The antiferromagnetic ordering is also observed in the resistivity measurements as a sharp drop in the spin disorder resistivity due to the ordering of the rare-earth moments as seen in Figs. 6 and 7. The temperature where the resistivity exhibits a sharp drop agrees well with the temperature where the susceptibility shows the peak. The susceptibility of CePdSn obeys Curie-Weiss law between 300 and 40 K, but shows deviation from it below 40 K. This is thought to be due to the effect of crystalline electric fields on the Ce$^{3+}$ ($J=\frac{5}{2}$) ion, which lift the ground-state degeneracy. The value of $\Theta_P$ in CePdSn is large and negative ($-68$ K) implying strong hybridization of the Ce-4*f* electrons with the conduction electrons. Detailed analysis of the susceptibility, resistivity, and heat capacity of CePdSn has been carried out in terms of crystalline-electric-field (CEF) effects.[16] This analysis yields one possible set of the CEF coefficients, namely, $B_2^0=-4.65$ K, $B_4^0=18.6$ K. Here $B_2^0$ and $B_4^0$ represent the strength of second- and fourth-order terms in the crystalline-electric-field Hamiltonian. The uniqueness of these parameters cannot be ascertained at present. The susceptibility of SmPdSn deviates from Curie-Weiss behavior at high temperatures (Fig. 4). This can be understood in terms of the temperature-independent Van Vleck contribution to the susceptibility associated with the second-order Zeeman effect, which becomes important due to the narrow multiplet width in Sm$^{3+}$ ion.[18]

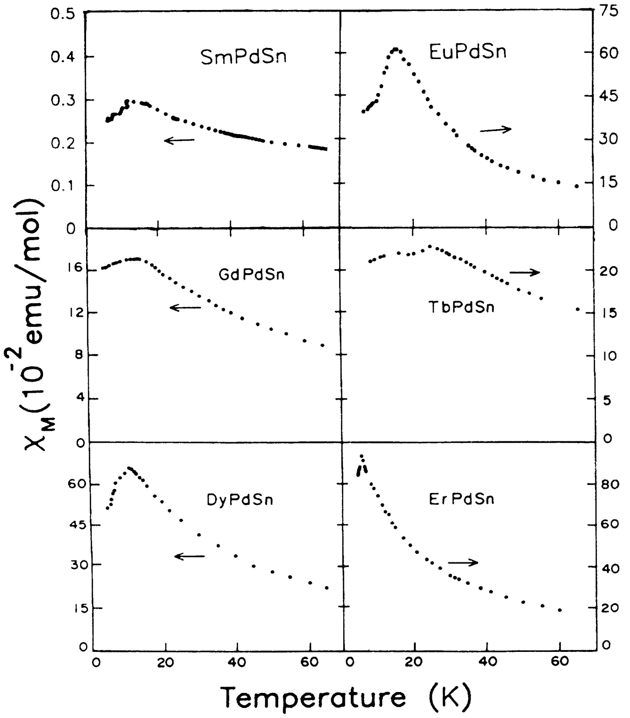

FIG. 5. Magnetic susceptibility of *R*PdSn (Sm, Eu, Gd, Tb, Dy, and Er) compounds at low temperatures showing the peak due to antiferromagnetic ordering.

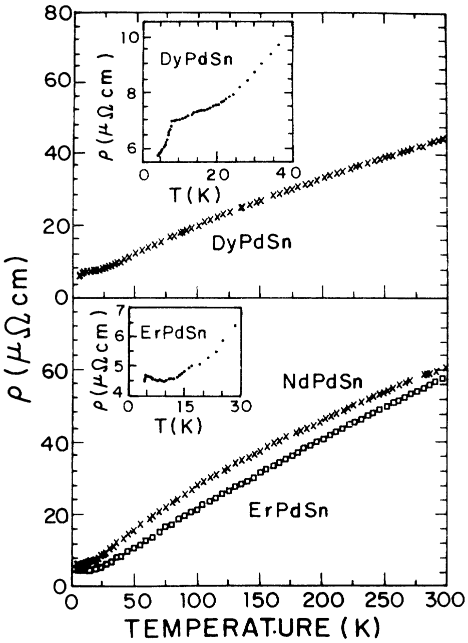

FIG. 6. Electrical resistivity of *R*PdSn ($R$=Nd, Dy, and Er) compounds as a function of temperature. Inset shows the resistivity drop at the Néel temperatures.

Figures 6–8 show the variation of electrical resistivity of some of the *R*PdSn compounds with temperature. All the *R*PdSn compounds exhibit metallic behavior in the temperature range of 4.2–300 K. The resistivity ($\rho$) of YbPdSn at 300 K is 8.6 m$\Omega$ cm, which is somewhat high compared to that observed in other *R*PdSn compounds where $\rho$ is between 50 and 700 $\mu\Omega$ cm. This may be due to the unusual nature of Yb ions in this compound. The

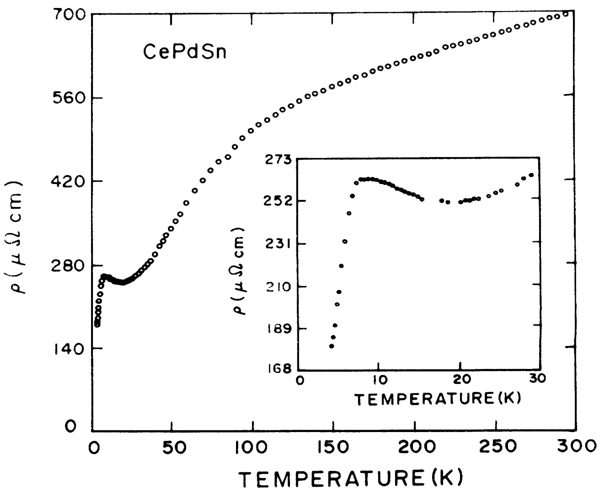

FIG. 7. Electrical resistivity of CePdSn as a function of temperature. Inset shows the variation of resistivity at low temperatures.

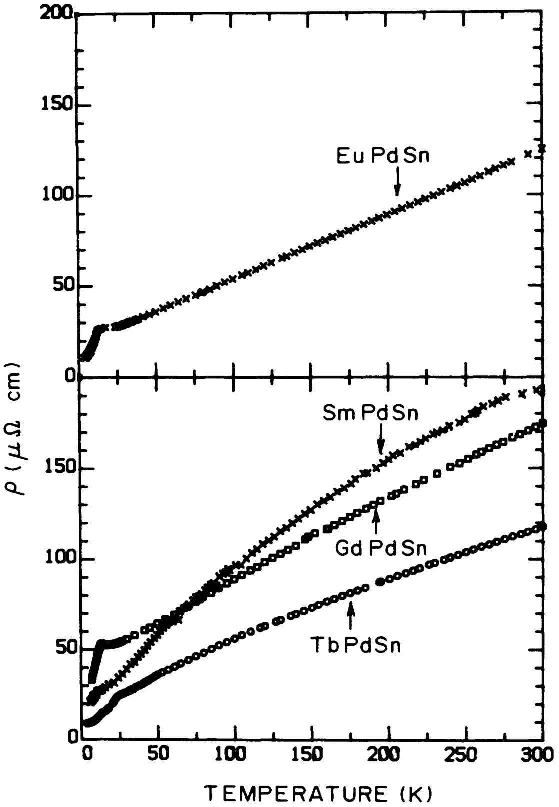

FIG. 8. Electrical resistivity of *R*PdSn ($R$=Eu, Sm, Gd, and Tb) compounds as a function of temperature.

resistivity of CePdSn is of particular interest. It decreases linearly with decreasing temperature from 300 to 70 K below which it shows a curvature. As mentioned above, this has been analyzed on the basis of crystalline-electric-field effects on the 4*f* electrons of $Ce^{3+}$ ion. A minima in the resistivity of CePdSn is observed at 20 K followed by a maxima at 7.5 K. The minima is typical of the Kondo lattice systems and suggests that CePdSn may represent an antiferromagnetically ordered Kondo lattice system. This observation is also consistent with the thermoelectric power measurements on this compound.[19]

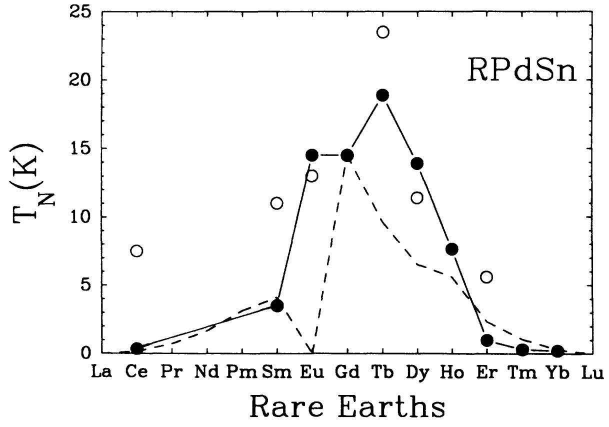

FIG. 9. Magnetic ordering temperatures of *R*PdSn compounds as a function of the rare-earth ion (open circles), prediction of the de Gennes rule (dashed line) and crystal-field model with the $B_2^0$ term only (solid circle and solid line).

### C. Magnetic ordering temperatures in the *R*PdSn series

The magnetic ordering temperatures in a series of isostructural compounds are usually explained on the basis of indirect exchange interaction between the 4*f* moments via conduction electrons proposed by Ruderman, Kittel, Kasuya, and Yosida (the so-called RKKY interaction).[20–22] In this approach, the conduction electron spin, **s**, is polarized by the rare-earth spin, **S**, through an exchange interaction of the type $-2J(0)\mathbf{S}\cdot\mathbf{s}$, where $J(0)$ is the exchange-interaction constant. This polarization is nonuniform in nature and couples different 4*f* spins or moments. The orientation of the local moments will depend on the sign of the conduction-electron polarization at the site and determines the type of magnetic ordering. Extending the perturbation approach with the molecular field approximation, de Gennes has shown[23,24] that the paramagnetic Curie temperature, $\Theta_P$, in the simplified RKKY theory is given by

$$\Theta_P = -\frac{3\pi n^2}{k_B E_F} |J(0)|^2 (g_J - 1)^2 J(J+1) \sum_i F(2k_F r_i) , \qquad (1)$$

where $n$ is the average number of conduction electrons per atom, $k_F$ is the Fermi wave vector, $g_J$ is the Landé $g$ factor, and $J$ the total angular momentum of the rare-earth ion. The function $F(x)$ stands for $x\cos(x) - \sin(x)$ with $x = 2k_F r$, and $\sum F(x)$ represents the lattice sum over the local-moment distances with origin at one of the local-moment sites. It is obvious from Eq. (1) that, all other parameters remaining constant, in an isostructural series of compounds, the $\Theta_P$ (or magnetic ordering temperatures) should be proportional to $G = (g_J - 1)^2 J(J+1)$. This is known as de Gennes scaling.[23,24] For various compounds studied[25] it is observed that the ordering temperatures generally follow the same universal curve when plotted against de Gennes factor $G$.

The magnetic ordering temperatures ($T_N$ or $T_M$) of *R*PdSn compounds are plotted as a function of the rare-earth ion ($R$) in Fig. 9. According to the de Gennes scaling mentioned above, in the isostructural, orthorhombic *R*PdSn series, $T_N$ should be maximum for the $Gd^{3+}$- or the $Eu^{2+}$-containing compounds. However, we find that the maxima, instead, occurs at Tb in this series. It has been observed earlier that $T_M$ of $RRh_4B_4$ series also does not scale with the de Gennes factor.[26] In this case, $T_M$ of $DyRh_4B_4$ is higher than that of $GdRh_4B_4$. Further, on the basis of de Gennes scaling, one would expect the transition temperature of the Ce compound to be 91 times smaller than that of the isostructural Gd compound, which, however, is not the case in the *R*PdSn series, the $T_N$ of CePdSn being approximately half that of GdPdSn. The $T_N$ of SmPdSn is also almost equal to the $T_N$ of GdPdSn. Because of the unstable nature of the 4*f* shell in Ce and Sm, it is likely that the high value of the $T_N$ of Ce and Sm compounds is due to the relatively strong exchange coupling, between the 4*f* electrons and the conduction electrons, caused by the hybridization of the $4f$ states with the conduction-electron states.[27] This is also reflected in the spin disorder resistivity of these compounds [proportional to $J(0)^2$], which is higher than that of the other $R$PdSn compounds.

The shifting of the $T_N$ peak from Gd to Tb can be understood to be a consequence of the crystalline-electric-field (CEF) effects. When crystalline electric fields split the $J_z$ states significantly with respect to $T_M$ or $T_N$, the de Gennes behavior is not to be expected in the isostructural series of compounds. In this case crystalline-electric-field terms should be added to the exchange Hamiltonian. Noakes and Shenoy[28] and Dunlap *et al.*[29] have shown that crystal fields of a suitable sign can enhance the ordering temperatures and shift the maximum away from the Gd compound as has been observed in $R$Rh$_4$B$_4$ compounds. In the presence of crystalline electric fields, the equation for $T_M$ may be written as[22]

$$T_M = 2J_{ff}(g_J-1)^2\langle J_z^2(T_M)\rangle_{\rm CF} , \tag{2}$$

where $\langle J_z^2(T)\rangle_{\rm CF}$ is the expectation value of $J_z^2$ in the presence of crystalline electric fields alone, evaluated at $T=T_M$ along the easy direction of magnetization, and $J_{ff}$ is the exchange constant for the exchange interaction between the rare-earth $(4f)$ ions. Equation (2) is an implicit equation for $T_M$, which can be solved numerically. Since Gd is an $S$-state ion which is not influenced by crystalline electric fields, its $T_M$ can be used to fix the value of the exchange constant $J_{ff}$. As mentioned earlier, the presence of crystalline electric fields in the $R$PdSn series has been inferred from the analysis of the susceptibility, resistivity, and heat-capacity data of CePdSn. A set of crystalline-electric-field parameters which can explain the above results in CePdSn has been obtained, though the uniqueness of this set cannot be ascertained at present. Assuming that the sign and possibly the magnitude of $B_2^0$ is correct in CePdSn, we have scaled this value to that for other rare-earth ions taking into account the expectation value, $\langle r^2\rangle$, of the $4f$ electron radius, Stevens' operator equivalent factor $\alpha_J$ and assuming no shielding correction. Equation (2) has been solved for all the magnetically ordered compounds of the $R$PdSn series and the calculated $T_M$ values are plotted in Fig. 9. It is seen that the CEF model with the simple $B_2^0$ term produces the maxima in $T_M$ for Tb and also nearly its magnitude as observed experimentally. The inclusion of the $B_2^2$ term, projected from that obtained for Ce in CePdSn, however, causes $T_M$ values to reduce. It is possible that either the $B_2^2$ term is not well determined or the other $B_n^m$ crystal-field coefficients, which have not been included, may be required for further analysis.

## IV. CONCLUSIONS

The magnetic and transport studies on ternary equiatomic compounds, $R$PdSn ($R$=Ce–Yb), reveal that the compounds with $R$=Ce, Sm, Eu, Gd, Tb, Dy, and Er order antiferromagnetically with Néel temperature $T_N$=7.5, 11, 13, 14.5, 23.5, 11.4, and 5.6 K, respectively, while compounds with $R$=Pr, Nd, Ho, Tm, and Yb remain paramagnetic down to 4.2 K. The $T_N$ of $R$PdSn series does not follow the de Gennes scaling. The $T_N$ of CePdSn (7.5 K) and SmPdSn (11 K) are of the same order as the $T_N$ of GdPdSn (14.5 K) possibly due to strong hybridization between the $4f$ electrons and the conduction electrons. Further, the $T_N$ of TbPdSn (23.5 K) is higher than that of GdPdSn, although de Gennes scaling predicts maximum $T_N$ for the Gd compound. This behavior can be understood as a consequence of crystalline electric fields acting on the rare-earth ion in the $R$PdSn compounds.

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