Specific heat and heavy-fermionic behavior in $Ce_8Pd_{24}M$ (M=Ga, In, Sn, Sb, Pb, and Bi)

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Specific-heat measurements have been carried out to further elucidate the electronic and magnetic properties of $Ce_8Pd_{24}M$ (M=Ga, In, Sn, Sb, Pb, and Bi). The measurements show that a slight change in the chemical and electronic structure of $CePd_3$, by forming the ordered ternary $Ce_8Pd_{24}M$-type compounds, induces an enhanced low-temperature electronic specific heat γ. This indicates the emergence of heavy-fermionic behavior with the loss of the intermediate valence nature of $CePd_3$. All of the $Ce_8Pd_{24}M$-type compounds studied here exhibit a sharp peak in the specific heat at low temperature due to antiferromagnetic (AF) transitions. Below the transition temperature, the specific heat shows the heavy-fermionic behavior, i.e., $\gamma(T\rightarrow 0)$ equal to a finite value, rather than $\gamma(T\rightarrow 0)=0$ in the paramagnetic AF ground state. [S0163-1829(98)03815-6]

I. INTRODUCTION

A variety of physical ground states are found in some Ce containing intermetallic compounds due to the strong hybridization of the $4f$ electrons with the conduction electrons.[1,2] Of particular importance are the intermediate-valence and heavy-fermion states with paramagnetic, magnetically ordered, and superconducting ground states. Current theoretical models of these materials suggest that the f electron hybridizes with the conduction electrons. The particular state that results is determined by the strength of the hybridization and by the energy difference between the f level and the Fermi level. The ground-state properties are believed to be a delicate function of the chemical and electronic surroundings around the $4f$ element. Thus it is believed that the physical properties of a compound can be changed dramatically by only a slight modification of the atomic arrangement, chemical identity, or electronic structure around the Ce atom.

One of the most interesting Ce intermetallics is $CePd_3$, a known intermediate valence material.[3] Recently, a series of related ordered ternary compounds, $Ce_8Pd_{24}M$ type, were discovered and the temperature-dependent magnetic susceptibility $\chi(T)$ and electrical resistivity $\rho(T)$ were studied.[4,5] The structure of $Ce_8Pd_{24}M$ is easily derived from that of $CePd_3$. $CePd_3$is cubic with Ce at cube corners and Pd on the cube faces (Cu_3Au structure). In $Ce_8Pd_{24}M$, eight unit cells of $CePd_3$ form a cube with an a axis double that of $CePd_3$. One of the eight Pd_6 octahedra is centered by M in an ordered fashion to keep the overall cubic symmetry. Such centering enlarges that Pd_6 octahedra and compresses the six Pd_6 octahedra that join the centered Pd_6 unit. The remaining Pd_6 unit is virtually undistorted. Magnetically, the $\chi(T)$ data indicate that doping the p-block elements into $CePd_3$ causes the loss of the intermediate-valence state and the formation of trivalent Ce atoms that undergo antiferromagentic (AF) ordering at low temperatures. Electronically, the $\rho(T)$data show typical metallic behavior for M=Sn, Sb, Pb, and Bi, whereas for M=Ga and In the data reveal an anomaly attributed to Kondo lattice behavior. In this paper we report the specific-heat data for $Ce_8Pd_{24}M$ (M=Ga, In, Sn, Sb, Pb, and Bi).

II. EXPERIMENT

The samples were prepared by arc melting the elements (>99.9% purity) in the desired ratios using small excess (<2%) of the more volatile elements Pb, Sb, and Bi as described in detail elsewhere.[4,5] Each sample was turned over several times and remelted to help ensure homogeneous ingots. The mass loss after arc melting was always less than 1% of the desired stoichiometric value. The samples were then placed in sections of tantalum tubing, sealed in evacuated quartz tubes, and annealed at 900 °C for two weeks. The single-phase nature of each sample was confirmed using powder diffraction on a SCINTAG θ-2θ diffractometer with Cu $K\alpha_1$ radiation. The specific-heat measurements down to 1.2 K were made on small-mass ($\approx$5 mg) pieces of each sample using a time constant method (relaxation method) technically described in detail elsewhere.[6]

III. RESULTS AND ANALYSIS

A. $Ce_8Pd_{24}In$

The specific heat (C_p) for $Ce_8Pd_{24}In$ is plotted versus temperature (T)in Fig. 1 together with the data for the isostructural, nonmagnetic compound $La_8Pd_{24}In$. By fitting the data of $La_8Pd_{24}In$ to the equation

$$C_p(T)=\gamma T+\beta T^3, \tag{1}$$

where the linear and the cubic terms correspond to the electronic and lattice contributions to the specific heat, respectively, it is found that γ=2.21(14) mJ/mol K^2 and β=5.60(4) mJ/mol K^4, yielding the Debye temperature $\theta_D\approx$225 K from the relation of $\theta_D\propto(n/\beta)^{1/3}$, where n is the number of atoms in a formula unit. In usual metal, the equation (1) for the specific heat is valid for temperatures below θ_D/50 (and de-

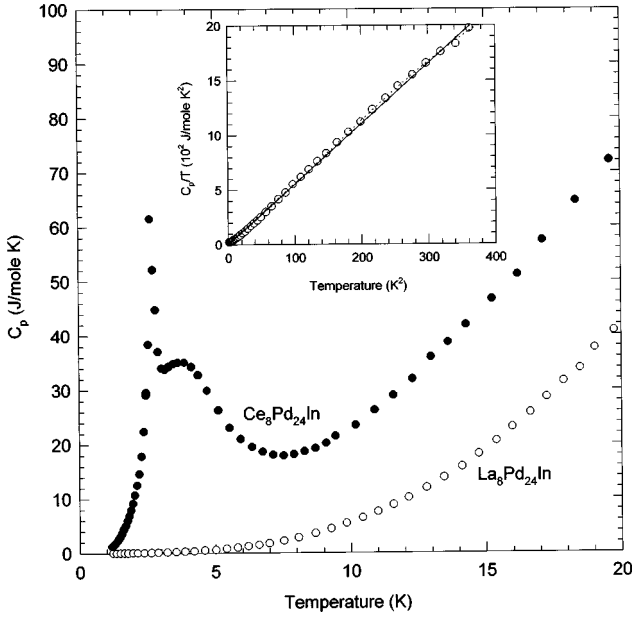

FIG. 1. Specific heat (C_p) versus temperature (T). Closed circles, $Ce_8Pd_{24}In$; open circles, $La_8Pd_{24}In$. Inset: C_p/T of $La_8Pd_{24}In$ versus T^2. The data were fitted to the equations of $C_p = \gamma T + \beta T^3$ (solid line) and $C_p = \gamma T + \beta T^3 + \eta T^7$ (broken line).

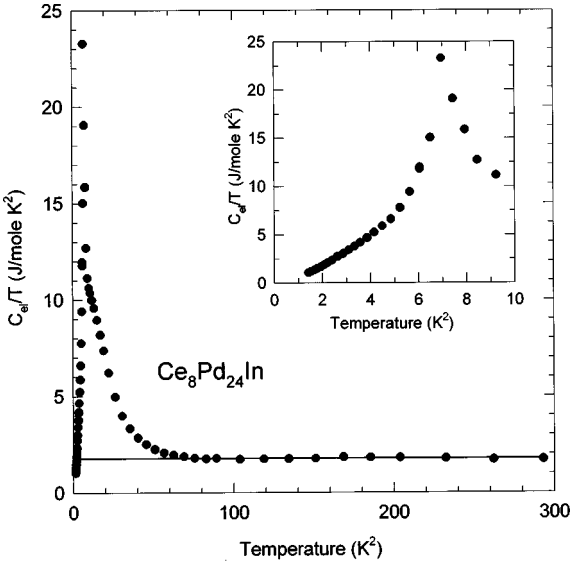

FIG. 2. Electronic specific heat divided by temperature (C_{el}/T) versus T^2. Inset: expanded plot at low temperature. The line indicates the γ_p extrapolated from high temperature.

pending on the material even up to $\theta_D/10$). In order to check the validity of Eq. (1) up to $T=20$ K, higher-order terms including T^5 and T^7 terms are used for evaluating of θ_D again. The fitting results are shown in the inset of Fig. 1 for comparison. This results in the value of $\theta_D \approx 232$ K, which means that Eq. (1) can be used up to $T \approx \theta_D/10$ within an error of a few percent ($\approx 2\%$) in $La_8Pd_{24}In$. Clear evidence of a phase transition in $Ce_8Pd_{24}In$ is given by the sharp peak at $T=2.6$, but a broad maximum at $T \approx 4$ K is also observed. Magnetic susceptibility measurements show a gradual deviation from Curie-Weiss behavior near $T=4$ K and a sharp decrease near $T=2.5$ K indicating an AF transition.[7] Thus, the peak in the specific heat at $T=2.6$ K is attributed to an AF transition and the broad shoulder above this peak corresponds to the other change in $\chi(T)$.

In order to study the electronic specific heat (C_{el}) of $Ce_8Pd_{24}In$, the lattice contribution from $La_8Pd_{24}In$ was subtracted using

$$C_{el}/T = [C^{tot}(Ce) - C^{latt}(La)]/T. \quad (2)$$

Figure 2 shows a plot of $C_{el}(T)/T$ versus T^2 for $Ce_8Pd_{24}In$. Above the transition, C_{el} clearly varies linearly with temperature and the γ_p parameter, denoting the electronic specific-heat parameter in the paramagnetic region, is large: $\gamma_p = 1720$ mJ/mol K^2 or 215 mJ/mol Ce K^2. This is much larger than the values for both $La_8Pd_{24}In$ and $CePd_3$, $\gamma(CePd_3) \approx 35$ mJ/mol K^2.[8] Below the phase transition, the $\gamma_o(T \to 0)$ value looks to be below $\gamma(T=1.2\ \text{K}) = 1000$ mJ/mol K^2 and is evidently smaller than the γ_p derived as above. The inset of Fig. 2 shows the specific heat below the phase transition. In the mean-field approximation, a T^3 behavior of the heat capacity is expected in an AF state. Although a short region at low temperature of the C/T vs T^2 plot has a slope ≈ 1.5 J/mol K^4, the extrapolation to $T=0$ does not intersect the origin, resulting in the negative γ_o, which makes no physical sense. This means that the temperature is not low enough to show the T^3 behavior at the low-temperature limit in the AF ground state in the mean-field approximation.

B. $Ce_8Pd_{24}Sb$

Figure 3(a) shows the as-measured specific heat of $Ce_8Pd_{24}Sb$ versus T. The sharp peak at $T=5.1$ K indicates a phase transition and the shoulder above the peak is similar to the one seen in Fig. 1. Two distinct changes in the $\chi(T)$ of $Ce_8Pd_{24}Sb$ at $T=5.1$ and 6.4 K are reported elsewhere.[5] At $T=5.1$ K there is a sudden drop of $\chi(T)$ with decreasing temperature that resembles an AF transition and between $T=6.4$ and 5.1 K the $\chi(T)$ shows temperature-independent behavior. In addition, a sharp slope change in $\rho(T)$ was found near $T=6.4$ K.[5] Thus the peak in Fig. 3(a) at $T=5.1$ K is again a transition of AF nature and the shoulder seems to correspond to the transition of unknown origin seen at $T=6.4$ K in both $\chi(T)$ and $\rho(T)$.

Subtracting the lattice specific-heat contribution of $La_8Pd_{24}In$ as described above gives the electronic specific heat of $Ce_8Pd_{24}Sb$, plotted as $C_{el}(T)/T$ versus T in Fig. 3(b). The γ_p above the transition is slightly temperature dependent, increasing with decreasing temperature, and is also large: $\gamma_p(T=10\ \text{K}) \approx 1200$ mJ/mol K^2. This value is similar to the $Ce_8Pd_{24}In$ case. The γ_o at $T=1.2$ K below the transition is close to ≈ 400 mJ/mol K^2, which is much smaller than the extrapolated $\gamma_p(T \to 0) \approx 1600$ mJ/mol K^2. As shown in the inset of Fig. 3(b) the specific-heat data below the transition approaches zero as temperature goes to zero. The temperature dependence of specific heat below the transition is quite different from in $Ce_8Pd_{24}In$ above, showing the T^3 behavior near below transition temperature, which must not be caused by the pure AF spin wave because the temperature is close to the transition temperature.

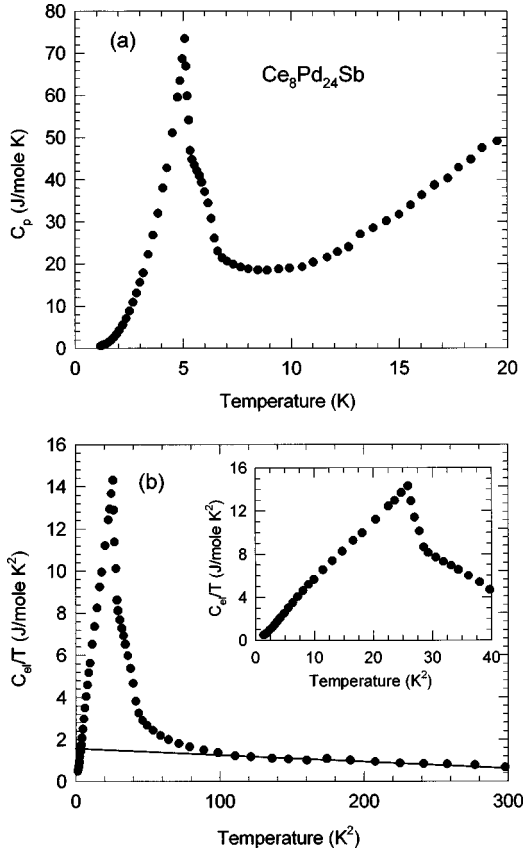

FIG. 3. (a) Specific heat (C_p) of $Ce_8Pd_{24}Sb$ versus T. (b) Electronic specific heat of $Ce_8Pd_{24}Sb$ divided by temperature (C_{el}/T)versus T^2. Inset: expanded plot at low temperature. The line indicates the γ_p extrapolated from high temperature.

C. $Ce_8Pd_{24}M$ (M=Ga, Sn, Pb, and Bi)

The electronic specific heat for the M=Ga, Sn, Pb, and Bi compounds are shown in Figs. 4(a), 4(b), 4(c), and 4(d), respectively. Again, the lattice contribution of $La_8Pd_{24}In$ was subtracted as described above. For M=Ga, the peak near T=3.1 K is smaller and broader compared to other compounds. However, considering the $\chi(T)$ behavior in Ref. 7, it is still considered to result from an AF phase transition. The γ_p is increasing with decreasing temperature with $\gamma_p(T=10\ \mathrm{K})\approx 2720$ mJ/mol K^2 and the extrapolated $\gamma_p(T\rightarrow 0)\approx 3300$ mJ/mol K^2. The magnitude and temperature dependence of γ_p for M=Ga is largest among the series of compounds studied here. The $\gamma_0(T\rightarrow 0)$ seems to be smaller than the extrapolated γ_p at T=0 K, as shown in the inset of Fig. 4(a).

For M=Sn, the peak at T=5.7 K is again of AF origin based upon the $\chi(T)$ and $\rho(T)$.[5] A temperature-independent $\gamma_p\approx 1030$ mJ/mol K^2 is found. One peculiar fact for $Ce_8Pd_{24}Sn$ is that the $\gamma_o(T=1.2\ \mathrm{K})\approx 1900$ mJ/mol K^2 is much larger than the γ_p. The $\gamma_o(T\rightarrow 0)$ is not likely to be much lower than the $\gamma_o(T=1.2\ \mathrm{K})$ because for temperatures below T=2.5 K the $C_{el}(T)/T$ data start to level off rather than continually decreasing as shown in the inset of Fig. 4(b). Thus, for M=Sn the low-temperature region is different from all the other compounds investigated. This is also unusual among the known heavy-fermion compounds such as $NpSn_3$ and U_2Zn_{17}.[9,10] However, since another transition, at low temperatures, could be developing, it is necessary to study the properties at the zero-temperature limit.

For M=Pb and Bi, the peaks at 5.8 and 4.6 K, respectively, are again assigned to be AF in nature, consistent with the $\chi(T)$ and $\rho(T)$.[5] In both cases the γ_p is temperature independent and is $\approx$1100 mJ/mol K^2 for Pb and $\approx$1300 mJ/mol K^2 for Bi. The $\gamma_o(T\rightarrow 0)$ is a little smaller than the γ_p for Pb and is close to the γ_p for Bi. The low-temperature region is similar to the one for M=Sn.

IV. DISCUSSION

Most of the physical properties discussed above are summarized in Table I, including the magnetic transition temperatures (T_M) determined from the $\chi(T)$ and $\rho(T)$ in Refs. 5 and 7. The plot of C/T versus T^2 of raw measured data of $Ce_8Pd_{24}Ga$ shows a slightly negative slope, resulting in almost zero β, which is manifested itself in Fig. 4(a) with the slope being temperature dependent. These indicate that the simple application of Eq. (1) to the as-measured data of these heavy-fermion compounds is not appropriate. For the γ_p that are temperature dependent, the values at T=10 K are listed as indicated parentheses in Table I. Shoulders appear in the specific heat of M=In and Sb and are marked with an s in Table I.

Even though the physical origins of these shoulders are not yet clear, there is good evidence that they are an intrinsic property of the samples. First, the values of the specific heat at these shoulders are significantly large and comparable to the AF transition peaks. The shoulders also occur at different temperatures for the two samples and match features found in their respective $\chi(T)$. This makes it unlikely that the shoulders are due to a common impurity phase that should show a feature at a single temperature. In addition, close examination of the specific heat of each compounds reveals a slight additional temperature dependence of the AF transition peak on the high-temperature side. This may indicate that the shoulder, seen clearly in the In and Sb compounds, is simply screened by the AF transition in the other compounds.

There is a significant increase in γ_p for the $Ce_8Pd_{24}M$-type compounds compared to that of $CePd_3$. This indicates that the slight change of structure and electronic state around the Ce 4f site from the p-block elements induces heavy-fermionic behavior at the cost of the intermediate-valence properties. The γ_p for M=In, Sn, Pb, and Bi is almost temperature independent above the AF transition (for 10 K$<T<$20 K). Similar fermionic behavior was found in the $NpSn_3$ and U_2Zn_{17} compounds in terms of an AF transition, a temperature-independent and high γ_p above the AF transition, and a reduced but still large γ_o.[9,10] In those compounds, the AF transition is due to the itinerant heavy-fermionic electrons in analogy to a BCS-type transition. The net entropy change below the AF transition can be close to zero, even negative, and the decrease of γ_o is considered a result of the opening of a gap in the electronic state at the Fermi level.

For some known heavy-fermion compounds such as $Ce\ Ga_2$ and U_2Zn_{17}, it has been noticed that the crystalline electric field (CEF) contribution to the heat capacity cannot be ignored and that the high value of γ may not be an indication of heavy-fermion behavior.[11] Even though the CEF

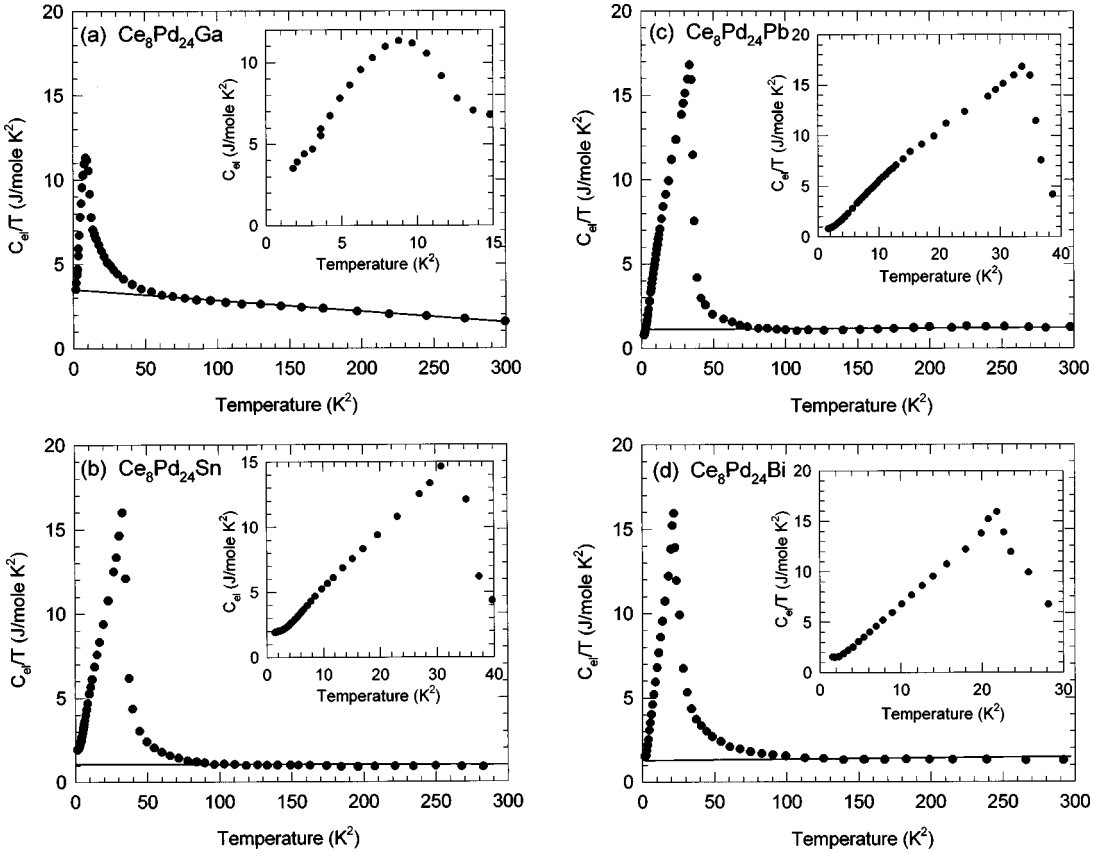

FIG. 4. Electronic specific heat divided by temperature (C_{el}/T) versus T^2: (a) $Ce_8Pd_{24}Ga$, (b) $Ce_8Pd_{24}Sn$, (c) $Ce_8Pd_{24}Pb$, and (d) $Ce_8Pd_{24}Bi$. Inset: expanded plot at low temperature. The lines indicate the γ_p extrapolated from high temperature.

excitation levels of $Ce_8Pd_{24}M$ are unknown, we can expect that the CEF excitations would be similar to those of $CePd_3$ because the local environment around the Ce sites are almost identical for both compounds. The specific-heat data of $CePd_3$ follow the behavior predicted in Eq. (1) and give $\gamma \approx 35$ mJ/mol K^2 in the temperature range where the γ_p values are extrapolated.[8] Thus it is likely that the CEF levels will not influence significantly the extrapolated γ_p.

Although the evaluation of the entropy associated with the transition is obscured due to the broadened transition peak and especially due to the shoulders in $Ce_8Pd_{24}In$ and $Ce_8Pd_{24}Sb$, it be useful to estimate the net entropy change associated with the transition. This is defined as

$$\Delta S = \int C_{el}/T\, dT - \int C_{el}^{extrap}/T\, dT, \qquad (3)$$

where C_{el}^{extrap} is the extrapolation of the higher-temperature data above the transition to $T \rightarrow 0$. When the data above $T = 10$ K are extrapolated, the ΔSvalues calculated from Eq. (3) range between about 16 and 34 J/mol K, which are listed in Table I. These significantly large positive values of ΔS, in contrast to $\Delta S \leqslant 0$ for $NpSn_3$ and U_2Zn_{17},[9,10] indicate that the transition is of localized electronic behavior. However, all of the ΔS values are clearly smaller than $8R$ ln2, where R is gas constant. The reduced entropy can be accounted for by the fact that either the itinerant electrons with heavy effective

TABLE I. Physical parameters of $CePd_3$, $Ce_8Pd_{24}M$(M=Ga, In, Sn, Sb, Pb, and Bi), and $La_8Pd_{24}In$. T_M is the transition temperature from the magnetic susceptibility and electric resistivity data from Refs. 5 and 7. The data for $CdPd_3$ are taken from Refs. 8. *s* denotes shoulder.

Compound	C_{max} (K)	$T_M^{\chi,\rho}$ (K)	γ_p (mJ/mol K^2)	γ_o (T=1.2 K) (mJ/mol K^2)	θ_D (K)	ΔS (J/mol K)
$CePd_3$			35	35	299	
$Ce_8Pd_{24}Ga$	3.1	3.1	2720 (T=10 K)	3300		16.4
$Ce_8Pd_{24}In$	2.6,*s*	2.6,4.0	1720	1000		28.2
$Ce_8Pd_{24}Sn$	5.7	7.5	1030	1900		34.0
$Ce_8Pd_{24}Sb$	5.1,*s*	5.1,6.4	1200 (T=10 K)	400		27.6
$Ce_8Pd_{24}Pb$	5.8	6.0	1160	790		33.4
$Ce_8Pd_{24}Bi$	4.7	5.0	1360	1400		28.8
$La_8Pd_{24}In$			2.21			

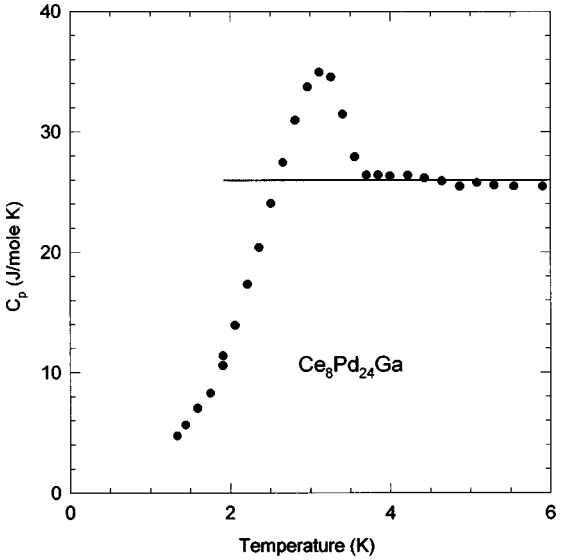

FIG. 5. Specific heat (C_p) versus T for $Ce_8Pd_{24}Ga$.

mass are partly involved in the transition or the magnetic ordering is due to an induced moment, as in Pr_3Tl.[12] The reduced γ_o after the AF transition, seen for all samples but $Ce_8Pd_{24}Sn$, is consistent with the former interpretation.

The high-temperature tail of the phase transition is believed to be due to both a sign of precursor, or short-range order, and sample inhomogeneity. However, there is an indication that the tail in $Ce_8Pd_{24}Ga$ is induced partly by the typical heavy-fermionic nature, which manifests itself as the rapid increase of γ_p as in the typical heavy-fermion materials such as $CeCu_2Si_2$, UBe_{13}, Cd Al_3, and $CeCu_6$.[1] This is represented in the data of $Ce_8Pd_{24}Ga$, as shown in Fig. 5. The $C_p(T)$curve has a broad peak of AF origin near 3.1 K, and is nearly constant between $T=3.7$ and 6 K at a value ≈ 26 J/mol K. Within the given temperature range, the γ_p can be approximated to be temperature independent because the slope of γ_p in Fig. 4(a) is very small compared to the extrapolated γ_p. If the constant $C_p(T)$ between $T=3.7$ and 6 K is attributed to the presence of AF transition, then the AF contribution to C_p would have to decrease linearly with T from 3.7 to 6 K so that the sum of the AF contribution and heavy-fermionic electron contribution $\gamma_p T$ is constant in this temperature range. Hence it is likely that the increase of γ_p, shown in Fig. 4(a), is in part an indication of heavy-fermionic electron behavior. The sharp transition at $T=3.7$ K supports the hypothesis of the heavy-fermionic behavior. The small, broad peak associated with the AF transition in $Ce_8Pd_{24}Ga$ may be a representation of the interplay between the localized magnetic electrons and itinerant electrons with high effective mass.

In this sense, the shoulders in $Ce_8Pd_{24}In$ and $Ce_8Pd_{24}Sb$ remind us of the large Kondo-like anomalies seen in both $CeCu_2Si_2$ (Ref. 13) and UBe_{13}.[14] These compounds have broad maxima in the specific heat near $T=3.5$ and 2.5 K, respectively, above their superconducting transitions. Thus the anomaly in $Ce_8Pd_{24}In$ and $Ce_8Pd_{24}Sb$ may be attributed to Kondo lattice behavior, resulting in the rapid increase in γ_p at low temperatures. Remember that indications of Kondo-like behavior were found in the $\rho(T)$for $Ce_8Pd_{24}In$ and $Ce_8Pd_{24}Ga$.[5] On the other hand, Kim and Stewart found that the anomaly in UBe_{13} fits very well with a Schottky temperature dependence if only a small fraction of the U ions are involved in the low-lying levels, regardless of what induces the low-lying levels.[15] The low-lying levels involved can be the ground-state doublet of Ce, which is split by the hybridization of the magnetic f electrons and the conduction electrons so that the anomaly is Schottky-like behavior for this splitting at high temperature, which changes to AF ordering as T is lowered so that cooperative interactions can dominate. It is also possible that another of the Ce doublets lies sufficiently near the ground-state doublet that population changes in the two could produce a Schottky specific heat that accounts for the shoulder but gets distorted as AF ordering takes place out of the ground-state doublet. In either scenario, the anomaly in the specific heat is not yet well understood.

V. CONCLUSION

The $Ce_8Pd_{24}M$ (M=Ga, In, Sn, Pb, Sb, and Bi) compounds with a modified structure of Cd Pd_3, show enhanced electronic specific heat (γ_p) above an AF transition, indicating heavy-fermionic behavior rather than the intermediate valence of $CePd_3$. This system allows for the study of both the evolution of the heavy-fermionic state from the intermediate-valence state and the effects of subtle changes in the electronic and structural environment around Ce on the properties of strongly correlated electrons with high effective mass. The specific-heat data below the AF transition temperature for most compounds studied here show finite $\gamma_o(T\rightarrow 0)$ behavior, which often appears in the AF state of heavy-fermion compounds, such as $NpSn_3$ (Ref. 9) and U_2Zn_{17},[10] rather than T^3 behavior of the paramagnetic AF ground state in the mean-field approximation. Measurements at lower temperature to see whether there are other transitions will help in the understanding of the magnetic transitions and the electronic properties of the heavy-fermionic state. Since the $Ce_8Pd_{24}Ga$ has the largest and most temperature-dependent γ_p among the compounds studied here, the doping of smaller p-block elements (i.e., M=Ge, Si, and Al) will be very interesting.

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