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Menon, L.; Kayzel, F.E.; de Visser, A.; Malik, S.K.

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## Gap suppression and development of antiferromagnetic order in $\text{CeRh}_{1-x}\text{Pd}_x\text{Sb}$ : Heat-capacity studies

Latika Menon

*Tata Institute of Fundamental Research, Homi Bhabha Road, Colaba, Mumbai 400 005, India*

F. E. Kayzel and A. de Visser

*Van der Waals-Zeeman Laboratory, University of Amsterdam, Amsterdam, The Netherlands*

S. K. Malik

*Tata Institute of Fundamental Research, Homi Bhabha Road, Colaba, Mumbai 400 005, India*

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$\text{CeRhSb}$  is a mixed-valent compound in which a gap opens up in the electronic density of states below a temperature of about 10 K. Earlier studies on Pd substitution at the Rh site in this compound showed that the gap is suppressed for around 10% Pd and an antiferromagnetic ground state emerges with higher Pd substitution. Here, we report heat-capacity results on some of these samples, namely,  $\text{CeRh}_{0.8}\text{Pd}_{0.2}\text{Sb}$  and  $\text{CeRh}_{0.7}\text{Pd}_{0.3}\text{Sb}$ . We observe a sharp rise in  $C_{4f}/T$  of  $\text{CeRh}_{0.7}\text{Pd}_{0.3}\text{Sb}$  starting at around 5 K with a peak at about 2.5 K, which confirms the antiferromagnetic ordering in this compound. An entropy change of 2.5 J/mol K is obtained for the ordering. Above the Néel temperature, the value of the electronic specific heat coefficient  $\gamma$  is found to be about 40 mJ/mol K<sup>2</sup>. In  $\text{CeRh}_{0.8}\text{Pd}_{0.2}\text{Sb}$ ,  $C_{4f}/T$  is found to rise below 10 K down to the lowest temperature of measurement, namely, 1.5 K with a  $\gamma$  value of about 40 mJ/mol K<sup>2</sup>. The rise may be due to non-Fermi-liquid behavior or could be due to impending magnetic order in the compound below 1.5 K.

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Intermetallic compounds containing Ce are known to show interesting physical properties, such as mixed-valence, Kondo lattice, heavy fermion behavior, etc.<sup>1,2</sup> A variety of ground states are exhibited by them, namely, magnetic, non-magnetic, superconducting, etc. These phenomena are believed to arise due to hybridization between the Ce-4*f* electrons and the conduction electrons. One such interesting compound is  $\text{CeRhSb}$  (Ref. 3) in which Ce ions are in a mixed-valence state as evidenced by a broad maximum in the susceptibility at around 100 K.<sup>3</sup> Its resistivity is found to show a sharp rise at low temperatures. This is attributed to opening of a gap in the electronic density of states of  $\text{CeRhSb}$ , leading to the resistivity rise and an insulatinglike ground state. A similar behavior is observed in  $\text{CeNiSn}$  (Ref. 4) and  $\text{Ce}_3\text{Bi}_4\text{Pt}_3$ ,<sup>5</sup> etc.<sup>6</sup>

We have been carrying out systematic investigations of the gap formation in  $\text{CeRhSb}$ , by means of substitutional studies at the Ce and Rh sites, as revealed in electronic, transport, and magnetic measurements.<sup>7-9</sup> We have observed that substitution at the Ce site by La leads to gap suppression with about 10% La.<sup>7</sup> Similarly, substitution at the Rh site by Ni also leads to gap suppression with around 10% substitution.<sup>8</sup> Pd substitution at the Rh site leads to very interesting results, namely, the development of antiferromagnetic ordering following gap-suppression<sup>9</sup> as inferred from magnetic susceptibility and electrical resistivity measurements. These results are consistent with some of the theoretical models<sup>10-12</sup> for doping effects in such gap-forming systems. In this paper, we report the heat-capacity results on two representative samples of the  $\text{CeRh}_{1-x}\text{Pd}_x\text{Sb}$  series, namely,  $\text{CeRh}_{0.8}\text{Pd}_{0.2}\text{Sb}$  and  $\text{CeRh}_{0.7}\text{Pd}_{0.3}\text{Sb}$ . In the case of  $\text{CeRh}_{0.7}\text{Pd}_{0.3}\text{Sb}$ , a peak in  $C_{4f}/T$  is also seen at 2.5 K, thus

confirming the antiferromagnetic ordering noted from resistivity and magnetic studies. No such peak is seen for non-magnetic  $\text{CeRh}_{0.8}\text{Pd}_{0.2}\text{Sb}$ , where  $C_{4f}/T$  continues to rise down to 1.5 K. This may be due to non-Fermi-liquid behavior or due to impending magnetic order below 1.5 K.

The samples of  $\text{CeRh}_{1-x}\text{Pd}_x\text{Sb}$  were prepared by the usual arc-melting technique, as discussed in Ref. 9 and are the same on which magnetic and resistivity measurements were reported earlier. At the Rh-rich end ( $0 < x \leq 0.4$ ), the samples are orthorhombic, isostructural to  $\text{CeRhSb}$ , crystallizing in the  $\epsilon$ -TiNiSi-type crystal structure (space group *Pnma*) while at the Pd rich end ( $0.7 \leq x < 1$ ) the samples are hexagonal, isostructural to  $\text{CePdSb}$  (space group *P6<sub>3</sub>/mmc*). In the region  $0.4 < x < 0.7$ , the samples are found to be multiphase. For the hexagonal phase samples, ferromagnetic ordering is observed with Curie temperature  $T_C$  ranging from 8 K in  $\text{CeRh}_{0.3}\text{Pd}_{0.7}\text{Sb}$  to 17 K in  $\text{CePdSb}$ .<sup>13</sup> For the orthorhombic phase samples ( $0 < x \leq 0.4$ ), with increasing Pd substitution, there is a small increase in unit-cell volume which may lead to corresponding decrease in hybridization strength between the conduction electrons and the 4*f* electrons.

For the orthorhombic phase in the  $\text{CeRh}_{1-x}\text{Pd}_x\text{Sb}$  series, for  $x = 0.05$  and 0.1, a rise in resistivity is observed similar to that seen in the  $\text{CeRhSb}$ , though the rise is slower compared to that in  $\text{CeRhSb}$ . For  $x > 0.2$ , no rise in resistivity is observed. In fact, for samples with  $x = 0.3$  and 0.4, a drop in resistivity is seen at around 5 K. Moreover, the susceptibility of these two samples, shows a peak at around 3 K, attributed to antiferromagnetic ordering of the Ce moment. In light of this behavior seen for the orthorhombic samples, heat-

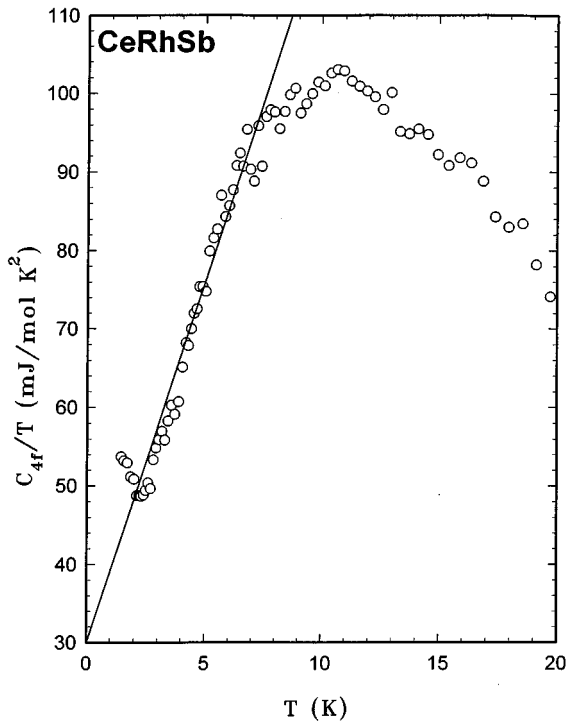


FIG. 1.  $C_{4f}/T$  (where  $C_{4f}$  is the 4*f*-related electron contribution to the heat capacity)  $T$  for CeRhSb (taken from Ref. 14).

capacity measurements for some representative samples in the orthorhombic phase are carried out. Two samples have been chosen, one with the antiferromagnetic ground state, that is, the sample with  $x=0.3$ , and the other nonmagnetic (down to 1.5 K) sample with  $x=0.2$ , where the gap is just suppressed. The heat-capacity measurements have been carried out in the temperature range 1.5–40 K, using the semia-diabatic heat pulse technique.

Before discussing the heat-capacity behavior of the Pd-substituted CeRhSb, we will first recapitulate the heat-capacity behavior of CeRhSb as discussed in Ref. 14. The heat capacity of CeRhSb gradually decreases with decrease in temperature down to 1.8 K.  $C_{4f}/T$  (where  $C_{4f}$  is the

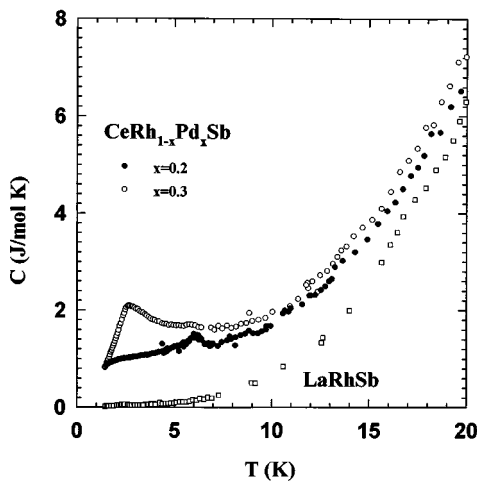


FIG. 2. Heat capacity  $C$  vs temperature  $T$  for the samples of CeRh<sub>0.7</sub>Pd<sub>0.3</sub>Sb, CeRh<sub>0.8</sub>Pd<sub>0.2</sub>Sb, and LaRhSb.

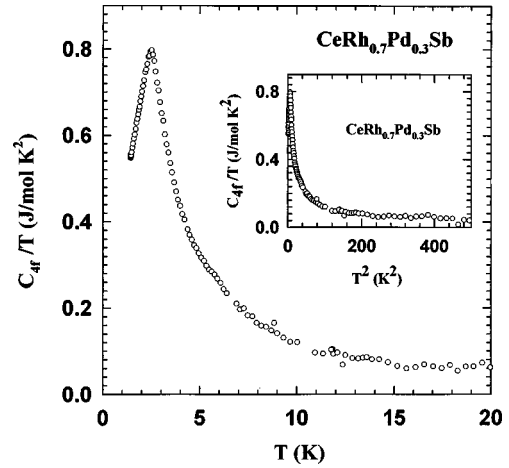


FIG. 3.  $C_{4f}/T$  vs  $T$  plot for the compound CeRh<sub>0.7</sub>Pd<sub>0.3</sub>Sb. The inset shows the plot of  $C_{4f}/T$  vs  $T^2$  for the same.

4*f*-related contribution to the heat capacity) rises with decrease in temperature down to 10 K, below which there is a drop down to 2 K (see Fig. 1). The increase down to 10 K suggests a tendency towards heavy-fermion-like behavior, but due to the development of the gap and a corresponding decrease in the density of states,  $C_{4f}/T$  drops below 10 K.

Figure 2 shows a plot of heat capacity vs temperature for the two Pd-doped samples with  $x=0.2$  and  $x=0.3$ . The heat capacity of LaRhSb (of the type  $C = \gamma T + \beta T^3$  with a  $\gamma$  value of about 7 mJ/mol K<sup>2</sup>) is also shown for comparison. We first discuss the heat-capacity results on CeRh<sub>0.7</sub>Pd<sub>0.3</sub>Sb. Its heat capacity drops gradually on lowering the temperature and shows a peak at about 2.5 K. The temperature of the heat-capacity peak is the same as the temperature of the susceptibility peak and the drop in the resistivity.<sup>9</sup> The bulk antiferromagnetic ordering reported earlier is confirmed by the present measurements taking into account the entropy associated with the peak (see below). This ordering is indeed intrinsic, as microprobe studies<sup>15</sup> do not indicate the presence of any noticeable second phase. The plot of  $C_{4f}/T$  (where  $C_{4f}$  is the 4*f*-related contribution to the heat capac-

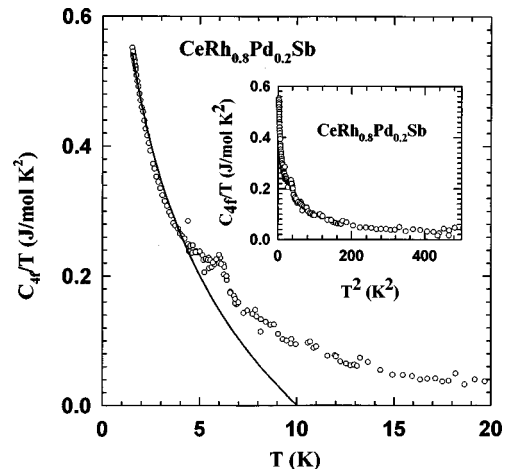


FIG. 4.  $C_{4f}/T$  vs  $T$  plot for the compound CeRh<sub>0.8</sub>Pd<sub>0.2</sub>Sb. The solid line shows the  $\ln T$  fit at low temperatures. The inset shows the plot of  $C_{4f}/T$  vs  $T^2$  for the same.

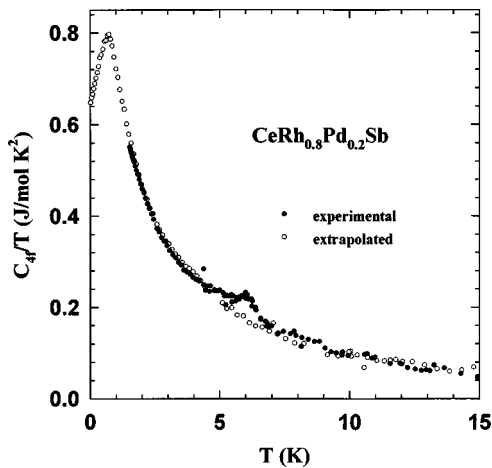


FIG. 5.  $C_{4f}/T$  vs  $T$  plot for the compound  $\text{CeRh}_{0.8}\text{Pd}_{0.2}\text{Sb}$  extrapolated below 1.5 K.

ity) for  $\text{CeRh}_{0.7}\text{Pd}_{0.3}\text{Sb}$  is shown in Fig. 3. A sharp peak is observed at around 3 K. The area under the peak, in the temperature range 0–6 K, gives the entropy change for the transition to be about 2.5 J/mol K. Assuming that the Ce ions in  $\text{CeRh}_{0.7}\text{Pd}_{0.3}\text{Sb}$  are in a doublet ground state, possibly due to crystal field effects, one expects a value of  $R \ln 2$ , that is about 5.75 J/mol K for the entropy change due to the magnetic ordering. The obtained value of 2.5 J/mol K for the entropy change suggests that the full moment of the crystal-field-split ground state doublet of Ce does not take part in the ordering. This is most probably due to the partial quenching of moments on the Ce ions due to hybridization effects mentioned above. In order to get an estimate of electronic specific heat coefficient  $\gamma$  we may plot  $C_{4f}/T$  vs  $T^2$  (see inset of Fig. 3). Above 10 K,  $C_{4f}/T$  is almost constant, which when extrapolated to  $T \rightarrow 0$ , gives a value of  $\gamma$  of about 40 mJ/mol  $\text{K}^2$ , which is the same as that obtained for  $\text{CeRhSb}$ .<sup>14</sup>

The heat capacity of  $\text{CeRh}_{0.8}\text{Pd}_{0.2}\text{Sb}$ , drops gradually with decrease in temperature (Fig. 2). At a temperature of 6 K, a small hump is seen which is most probably due to the presence of some magnetic impurity phases. In fact, for this sample, microprobe studies indicate the presence of about 2% of extra phase.<sup>15</sup> The extra phase may be  $\text{CeSb}$  which orders magnetically at around 6 K. Figure 4 shows a plot of  $C_{4f}/T$  vs  $T$  for this sample. A rise in  $C_{4f}/T$  is observed below 10 K. The rise is similar to that seen for  $\text{CeRh}_{0.7}\text{Pd}_{0.3}\text{Sb}$ . However, unlike  $\text{CeRh}_{0.7}\text{Pd}_{0.3}\text{Sb}$ , there is no signature of magnetic ordering down to 1.5 K, from resistivity and magnetization studies on this sample consistent with the present measurements. One possible reason for the rise may be the non-Fermi-liquid behavior at low temperatures,

as has been observed earlier in systems such as  $\text{CeCu}_{5.9}\text{Au}_{0.1}$ .<sup>16</sup> The non-Fermi-liquid behavior generally manifests in a  $\ln T$  behavior of  $C_{4f}/T$  in contrast to the  $C_{4f} \sim \gamma T$  kind of behavior expected for Fermi-liquid systems. In a system such as  $\text{CeCu}_{6-x}\text{Au}_x$ , going from the long-range ordered state for  $x > 0.1$  as one reduces the Au concentration, the Néel temperature scales to zero signalling a zero temperature quantum phase transition. It is believed that at this point deviations from Fermi-liquid behavior are expected. In the present  $\text{CeRh}_{1-x}\text{Pd}_x\text{Sb}$  system, since antiferromagnetic ordering is observed for  $x \geq 0.3$ , with decreasing Pd concentration, one expects the Néel temperature to scale to zero, with non-Fermi-liquid behavior for a critical range of Pd concentration. For  $\text{CeRh}_{0.8}\text{Pd}_{0.2}\text{Sb}$ , one may try and fit  $C_{4f}/T$ , at low temperatures, to a  $\ln T$  kind of behavior. In Fig. 3, the  $\ln T$  fit is shown by the solid line. A reasonably good fit is obtained below 5 K. However, it is very possible that there may be a magnetic transition below 1.5 K of which the rise below 5 K may be the high-temperature part. In order to check for a possible transition below 1.5 K, one may try and extrapolate the data for  $\text{CeRh}_{0.8}\text{Pd}_{0.2}\text{Sb}$ , below 1.5 K by normalizing  $C_{4f}/T$  data for  $x=0.3$  with respect to  $x=0.2$ . Figure 5 shows a plot of  $C_{4f}/T$  vs  $T$  for  $x=0.2$  extrapolated below  $T=1.5$  K. A peak in  $C_{4f}/T$  is seen at about 0.75 K, which may suggest a possible antiferromagnetic ordering in this compound. Hence, it is expected that with a slightly lower concentration of Pd ( $>20\%$ ), the ordering temperature should be reduced below 0.75 K and for a critical concentration of Pd, non-Fermi-liquid behavior is expected. Considering that the drop in the Néel temperature as one goes from  $x=0.3$  to  $x=0.2$  is rapid, from 2.5 to 0.75 K, it may seem that the critical concentration of Pd is almost close to  $x=0.2$ . The inset of Fig. 5 shows a plot of  $C_{4f}/T$  vs  $T^2$  for  $\text{CeRh}_{0.8}\text{Pd}_{0.2}\text{Sb}$ . Above 10 K,  $C_{4f}/T$  is almost constant and gives a value of  $\gamma$  about 40 mJ/mol  $\text{K}^2$ , which is the same as that obtained for  $\text{CeRhSb}$  and  $\text{CeRh}_{0.7}\text{Pd}_{0.3}\text{Sb}$ . Thus, above 10 K, the heat-capacity behavior seems to be almost unaffected by Pd substitution.

As seen above, the system  $\text{CeRh}_{1-x}\text{Pd}_x\text{Sb}$  presents interesting results. While in  $\text{CeRhSb}$  there is a drop in  $C_{4f}/T$  at around 10 K believed to be due to the gap formation in the electronic density of states, there is no such drop at around 10 K for  $\text{CeRh}_{0.7}\text{Pd}_{0.3}\text{Sb}$  and  $\text{CeRh}_{0.8}\text{Pd}_{0.2}\text{Sb}$ . Instead, there is a rise below 10 K in both these samples. In the case of  $\text{CeRh}_{0.7}\text{Pd}_{0.3}\text{Sb}$ , the rise is followed by a peak at around 2.5 K, consistent with the antiferromagnetic ordering. In the case of  $\text{CeRh}_{0.8}\text{Pd}_{0.2}\text{Sb}$  there does not seem to be any ordering down to 1.5 K. The rise in  $C_{4f}/T$  is most probably due to impending magnetic order at around 0.75 K. A non-Fermi-liquid behavior is expected for Pd substitution smaller than but close to 20%.

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