Evidence for low-dimensional magnetic behaviour in CePt$_5$Ge$_3$

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Abstract
We present the results of magnetic susceptibility, magnetization, heat capacity, resistivity and magnetoresistance measurements for the low-dimensional Ce-based intermetallic compounds $CePd_5Ge_3$ and $CePt_5Ge_3$, which crystallize in the YNi_5Si_3-type structure. Magnetic order is manifested by a peak in both the magnetic susceptibility and the specific heat at 1.9 K and 1.1 K for $CePd_5Ge_3$ and $CePt_5Ge_3$, respectively. For $CePt_5Ge_3$ a significant tail in the specific heat from T_N up to 5 K indicates pronounced antiferromagnetic short-range correlation of low-dimensional character above the 3D long-range order. Both compounds exhibit large negative magnetoresistance at low temperature.

1. Introduction

Among the rare-earth compounds, Ce-based intermetallics have been most extensively investigated due to their interesting physical properties. The competition between the Kondo interaction and the RKKY interaction leads to a variety of ground states such as a magnetically ordered Kondo lattice with reduced magnetic moment, heavy-fermion superconductivity, Kondo insulator etc [1]. Most of the heavy fermion (HF) compounds are of 3D nature. Only recently, the HF behaviour has been observed in the quasi-2D system $CeRhIn_5$ in which application of pressure induces a first-order-like transition from a heavy-fermion antiferromagnetic ground state with reduced magnetic moment to a superconducting state [2]. Also, there has been considerable interest to make artificial structures of Kondo compounds with reduced dimension such as small particles, wires and thin films [3–5]. The most dramatic result demonstrating the effect of dimensionality was found in the case of $CeAl_2$ where the bulk material orders magnetically at $T_N = 3.8$ K with the linear coefficient of specific heat, $\gamma = 150$ mJ mol^{-1} K^{-2} well below T_N, whereas for small particles of size 80 Å the magnetic transition is suppressed and γ is highly enhanced to a value of about 7 J mol^{-1} K^{-2} at 0.11 K with an extrapolated value at absolute zero of 9 J mol^{-1} K^{-2}, which is one of the highest values

reported for heavy-fermion systems [3]. Thus, it is of great interest to investigate a Ce-based system with low-dimensional character.

Recently $CePd_5Ge_3$ has been reported to form in the YNi_5Si_3-type orthorhombic structure (space group: *Pnma*), which consists of low dimensional building blocks (chains of Ce along the *b*-axis). In this structure the atoms of cerium and palladium produce trigonal prisms in which germanium atoms are located. Three trigonal prisms combine with each other to produce a star with Ce at the centre. These stars form linear chains along the *b*-axis [6]. With a motivation to identify more Ce-based compounds with this structure, we attempted to synthesize CeM_5Ge_3 (M = Ni, Ru, Rh, Pd, Pt). Here we report on the transport and magnetic properties and specific heat of single-phase samples of $CePd_5Ge_3$ and $CePt_5Ge_3$.

2. Experimental details

Polycrystalline samples of CeM_5Ge_3 (M = Ni, Ru, Rh, Pd and Pt) were prepared by arc melting a stoichiometric amount of high-purity elements under argon atmosphere on a copper hearth. Weight loss during the melting process was negligible. Samples were annealed at $900\,^{\circ}C$ for one week. Powder x-ray diffraction, investigation of the microstructure of a polished surface of the sample with optical microscope and electron probe microanalysis revealed that $CePd_5Ge_3$ and $CePt_5Ge_3$ were single phase, whereas samples with Rh had about 10% secondary phases and Ni and Ru compounds had more than 50% secondary phases. The resistivity and the magnetoresistance in the longitudinal configuration were measured by a standard low-frequency ac four-probe technique in the temperature range 0.4 K to 300 K and in magnetic fields up to 14 T. Magnetic susceptibility measurements were performed using a commercial superconducting quantum interference device magnetometer in the temperature range 1.9–300 K. High-field (up to 14 T) magnetization was measured using the extraction method. Magnetic measurements were performed on a solid piece of the polycrystalline sample or pelletized powder and no demagnetization correction has been applied to the data. Since the magnetization of these samples is small, the demagnetizing field is negligible compared to the externally applied field. An upper limit of the error involved in not considering the demagnetization factor is estimated to be 0.1%. Heat capacity measurements were carried out in a commercial physical property measurement system (Quantum Design) using a relaxation method. Most measurements were performed on two independently prepared samples of each compound. Both samples showed very similar behaviour.

3. Results and discussion

Powder x-ray diffraction revealed that $CePd_5Ge_3$ and $CePt_5Ge_3$ form in the YNi_5Si_3-type orthorhombic structure. The lattice parameters of $CePd_5Ge_3$, $a = 20.274$ Å, $b = 4.130$ Å and $c = 7.253$ Å are in agreement with the literature [6]. The lattice parameters of $CePt_5Ge_3$ are slightly larger, $a = 20.315$ Å, $b = 4.142$ Å and $c = 7.287$ Å. For $CePd_5Ge_3$, the Ce–Ce distance along the chain is 4.13 Å whereas the shortest interchain Ce–Ce distance is 5.97 Å. The corresponding distances for $CePt_5Ge_3$ are 4.14 Å and 5.99 Å.

The susceptibility (χ) data measured in a field of 0.1 T are plotted in figure 1 as χ^{-1} against T. The susceptibility data follow a Curie–Weiss behaviour in the temperature range 70–300 K for $CePd_5Ge_3$ and 50–300 K for $CePt_5Ge_3$ with effective magnetic moment $\sim 2.6\,\mu_B$ for both compounds, which is close to the value expected for a trivalent Ce^{3+} ion. The paramagnetic Curie temperature, θ_p, is -36 K for $CePd_5Ge_3$ and -22 K for $CePt_5Ge_3$. The θ_p values are rather large, similar to those found in many Ce-based Kondo compounds and the value for

CePd$_5$Ge$_3$ agrees with the literature [7]. However, the θ_p values were different on different pieces of samples and they were also very much dependent on the temperature range over which the susceptibility data were fitted. The above-mentioned values are derived from the susceptibility data measured on a pressed pellet of the polycrystalline powder sample. The deviation from the Curie–Weiss behaviour at low temperature is possibly due to the crystal field effect. We wish to mention here that the magnetization is linear up to 0.1 T even at the lowest temperature of the measurement and hence this deviation can not be due to nonlinear M–H behaviour at low temperature. CePd$_5$Ge$_3$ shows a sharp peak in the susceptibility at 1.9 K whereas CePt$_5$Ge$_3$ shows a broad rounded peak at 2.1 K. We do not find a significant difference between the zero-field-cooled and field-cooled susceptibility. Additional ac-susceptibility measurements were carried out down to 0.1 K in CePt$_5$Ge$_3$. We observed a peak in the susceptibility at 1.1 K, which corresponds to the long-range magnetic order as evidenced from the heat capacity data (see below). No signature of superconductivity was found down to 0.1 K.
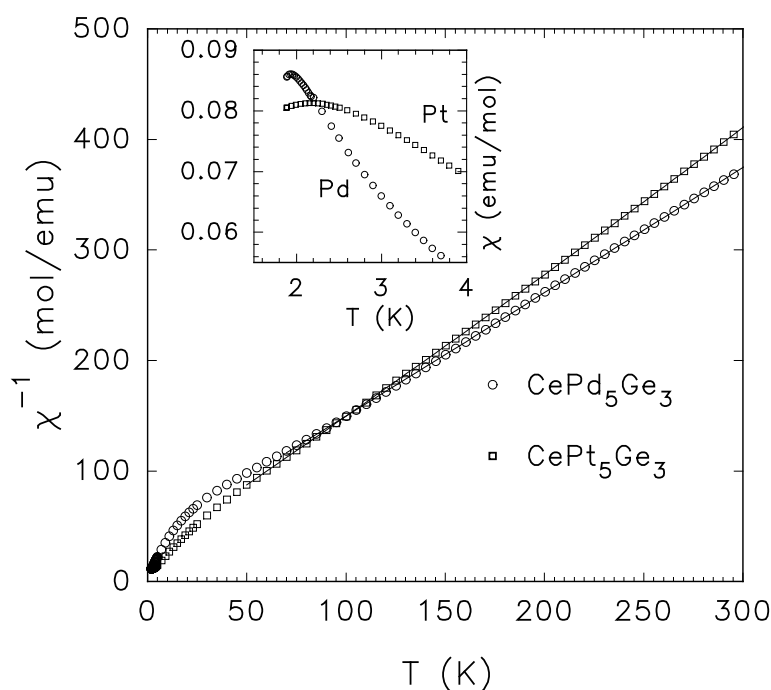
Figure 1. Inverse susceptibility (χ^{-1}) of CePd$_5$Ge$_3$ and CePt$_5$Ge$_3$ as a function of temperature. The inset shows the susceptibility at low temperature.

Magnetization data at 1.8 K up to 14 T are shown in figure 2. The magnetic moments at 1.8 K and 14 T for CePd$_5$Ge$_3$ and CePt$_5$Ge$_3$ are 0.77 and 1.08 μ_B/Ce respectively, which are much less than the saturation moment for the trivalent Ce^{3+} ion. An upturn in the $M(B)$ curve is seen at $\sim$1.2 T for CePd$_5$Ge$_3$ and at $\sim$2.5 T for CePt$_5$Ge$_3$. The upturn is possibly due to field-induced spin reorientation. Unfortunately, at present we do not have the data at lower temperatures and higher fields to determine the magnetic phase diagram of these two compounds.

Since the measurements were carried out on polycrystalline samples, we cannot determine the anisotropy of the magnetic behaviour. However, as polycrystalline samples of these compounds show significant differences in the magnetization when measured along two
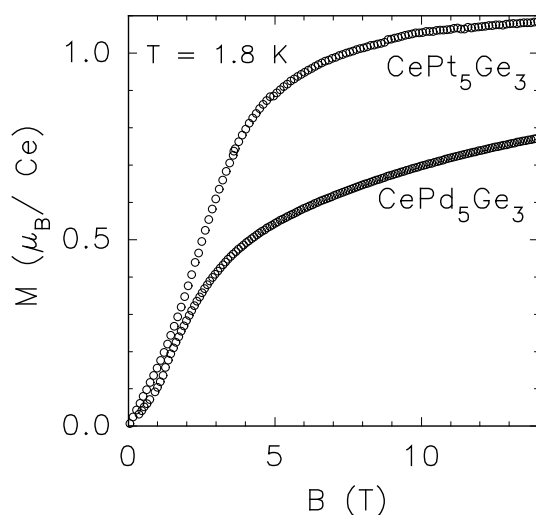
Figure 2. Magnetization as a function of field at 1.8 K.

perpendicular directions, we believe that these compounds show a significant magnetic anisotropy. Measurements on single crystalline samples are required to determine the anisotropic magnetic properties of these compounds.

In figure 3 we have plotted the magnetic part of the specific heat data, which is obtained by subtracting the specific heat of the La homologues. The specific heat data of $CePd_5Ge_3$ show a prominent anomaly with a peak at 1.8 K. The maximum value of the specific heat at the peak is $\sim$11.5 J mol^{-1} K^{-1}, which is close to the mean field value of 12.48 J mol^{-1} K^{-1}, and the magnetic entropy at T_N is $\sim$0.9R ln 2. This indicates a well behaved 3D antiferromagnetic order in $CePd_5Ge_3$. In contrast, the specific heat of $CePt_5Ge_3$ is more complicated. With decreasing temperature it shows a minimum at 6 K, below which it starts to increase till 1.2 K, then a sharp peak is observed at 1.1 K. At $T_N = 1.1$ K, the magnetic entropy is only $\sim$0.3R ln 2. The entropy at 5 K is $\sim$0.95R ln 2 in both compounds indicating that the crystal field ground state doublet is well separated from the first excited state. The magnetic part of the specific heat follows T^3 behaviour below 1.5 K in the case of $CePd_5Ge_3$ and below 1.0 K for $CePt_5Ge_3$, which is consistent with the antiferromagnetic ground state in both compounds.

The temperature dependence of the electrical resistivity for $CePd_5Ge_3$ and $CePt_5Ge_3$ is shown in figure 4. Both data show metallic behaviour with the room-temperature value typical for Ce-based intermetallic compounds. In the case of $CePd_5Ge_3$, below 10 K, a slight increase of the resistivity with decreasing temperature is found above a sudden drop of resistivity due to the onset of magnetic ordering (inset, figure 4). At first sight one would attribute the minimum in resistivity to the presence of the Kondo effect. However, since the magnetic entropy nearly reaches the expected value at T_N, the Kondo effect if at all is very weak in this compound. Therefore, a more likely possibility is the presence of critical scattering above the magnetic ordering. For $CePt_5Ge_3$ the resistivity is essentially temperature independent in the range 4–10 K, which is followed by a progressive decrease below 4 K, probably due to the onset of short-range correlations. A further clear drop in the resistivity is observed at 1.1 K, where the sharp peak in the heat capacity is observed.

Longitudinal magnetoresistance data at constant temperatures are shown in figure 5. At 1.8 K the resistance of $CePd_5Ge_3$ initially increases with increasing field, which is consistent
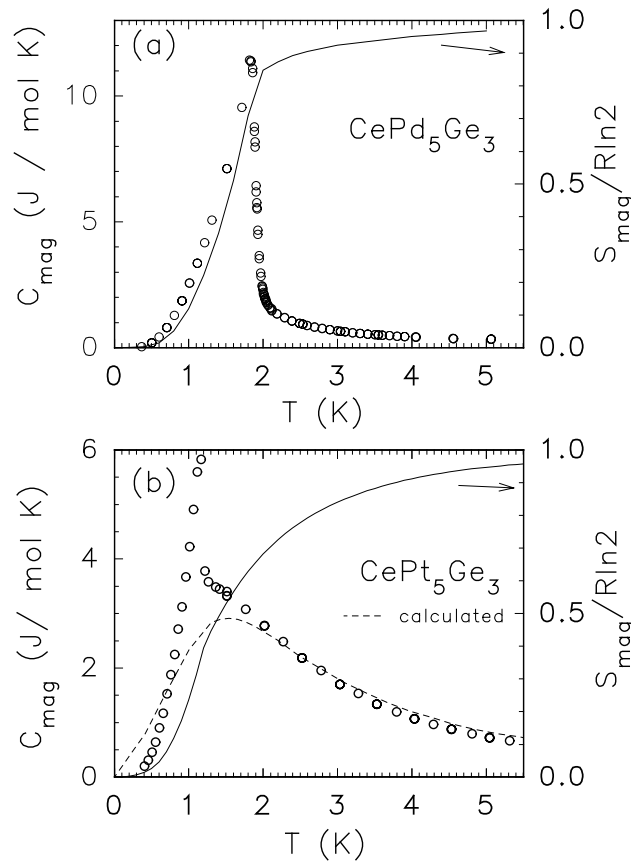
Figure 3. (a) Magnetic part of the specific heat and magnetic entropy of CePd$_5$Ge$_3$. (b) Magnetic part of the specific heat and magnetic entropy of CePt$_5$Ge$_3$. The dotted curve shows the theoretical value of the specific heat of an $S = 1/2$ AF Heisenberg chain with $J/k_B = 3.35$ K [9].

with the anitiferromagnetic nature of the magnetic ordering. At ~ 1.2 T, the resistivity starts decreasing. The resistivity of CePt$_5$Ge$_3$ also shows a large decrease at ~ 2.7 T. It is to be noted that the field at which the resistivity shows a large decrease agrees with the critical field at which an upturn in the $M(B)$ curve occurs. The magnetoresistance values are quite large at 1.8 K and 14 T, being -24% for CePt$_5$Ge$_3$ and -36% for CePd$_5$Ge$_3$.

In the case of CePd$_5$Ge$_3$ a peak in the susceptibility, a linear variation of magnetization and a positive magnetoresistance at low fields point to a long-range three-dimensional AF ordering at 1.8 K. For CePt$_5$Ge$_3$, the long-range magnetic ordering below 1.1 K is also possibly of antiferromagnetic type, since the magnetic part of the heat capacity in the ordered state follows T^3 rather than $T^{3/2}$ dependence. However, additional magnetic measurements in the low-temperature region are required to confirm this. The broad maximum in the susceptibility at 2.1 K, the continuous increase of C and the decrease of $\rho(T)$ below 4 K all indicate the development of AF short-range correlations. The broad rounded peak in susceptibility and the appearance of a heat capacity maximum at $0.7T_{\chi\,max}$ ($T_{\chi\,max}$ = temperature of the susceptibility maximum) is one of the characteristic features of the low-dimensional systems [8]. Considering that the crystal structure consists of Ce chains along the b-axis, we believe that the broad maximum in the susceptibility and the heat capacity tail in CePt$_5$Ge$_3$ are associated
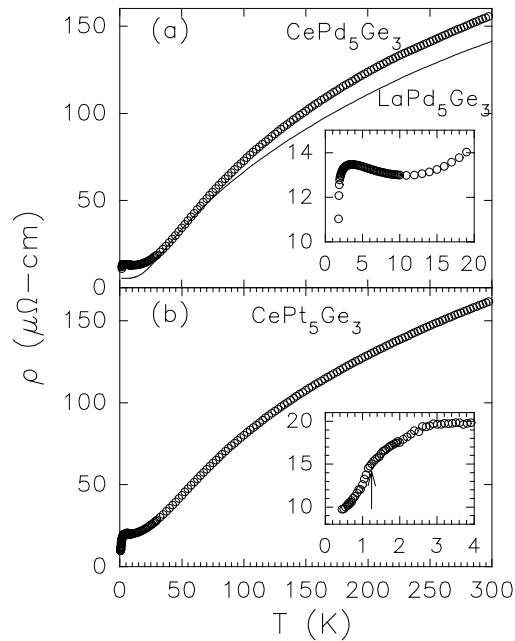
Figure 4. Resistivity of CePd$_5$Ge$_3$ and CePt$_5$Ge$_3$ as a function of temperature. The inset shows an expanded view at low temperature.

with the short-range magnetic correlations due to the low-dimensional character of the material. In the case of the $S = 1/2$ AF uniform Heisenberg chain with exchange constant J, a broad maximum in the susceptibility is observed at $T_{\chi\,max} = 0.6408 J/k_B$ (k_B = Boltzmann's constant) and a broad heat capacity maximum is observed at $T_{Cmax} = 0.4802 J/k_B$ with peak value $C_{max} = 0.349R \sim 2.9$ J mol^{-1} K^{-1} (see [9]). With $T_{\chi\,max} = 2.15$ K we obtain $J/k_B = 3.35$ K. Using this value of J, we plot in figure 3, the theoretical result of the magnetic specific heat for an $S = 1/2$ chain obtained by scaling the numerical results of Klümper [9]. The calculated specific heat agrees reasonably with the experimental value in the temperature interval 2–5 K. This suggests that the large tail in the heat capacity is due to low-dimensional short-range magnetic correlations. The magnetic entropy at T_N is also consistent with the low-dimensional magnetic behaviour. At $k_B T_N/J = 0.328$, the entropy of the $S = 1/2$ Heisenberg chain is estimated to be $0.36R \ln 2$ [9]. Thus an upper limit of the magnetic entropy at T_N is $0.36R \ln 2$, which is close to the experimentally observed value of $0.3R \ln 2$. A low-lying excited crystal-field level can also give rise to such a tail above a broad maximum in the specific heat. But then, one would expect the magnetic entropy at 5 K to be $\sim R \ln 4$ (corresponding to the ground state doublet and the excited doublet), which is much larger than the experimental value. In fact, the magnetic entropy reaches $R \ln 2$ at 10 K and even at 20 K the magnetic entropy is only $0.53R \ln 4$. Thus, this scenario can be excluded. A Kondo contribution can also give rise to an increase of the specific heat with decreasing temperature. However, the absence of Kondo type resistivity behaviour and the magnetic entropy of $0.95R \ln 2$ at 5.5 K suggests that the Kondo effect is negligible in this compound. Hence this possibility can be ruled out too. No difference between the ZFC and FC susceptibility rules out a spin glass type of behaviour. Thus, we believe that the broad maximum in the susceptibility at 2.1 K and the large tail between 2 K and 5 K in the specific heat of CePt$_5$Ge$_3$ are due to short-range magnetic
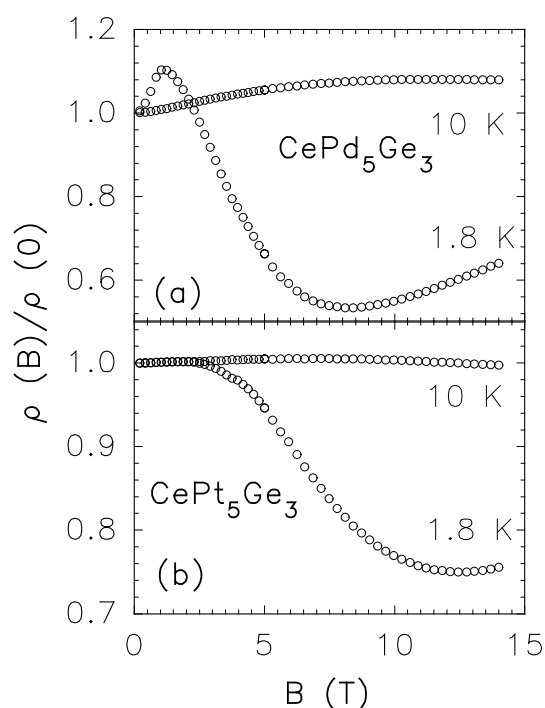
Figure 5. Normalized magnetoresistance as a function of field for CePd$_5$Ge$_3$ and CePt$_5$Ge$_3$.

correlations of low-dimensional character. A short-range order of low-dimensional character above a long-range three-dimensional magnetic order has also been seen in CePdSb [10, 11]. It is not clear at present why the Pt compound shows strong short-range correlations whereas they are absent in the Pd compound. A magnetic neutron scattering experiment would be very helpful for a better understanding of the different behaviours of these compounds.

4. Conclusions

The resistivity, magnetoresistance, magnetization and heat capacity data provide conclusive evidence of the magnetic phase transitions in CePd$_5$Ge$_3$ and CePt$_5$Ge$_3$. The magnetic entropy is nearly $R \ln 2$ at 5 K for both compounds, which implies a crystal field doublet ground state for both. Magnetization isotherms at 1.8 K show an upturn at 1.2 T in CePd$_5$Ge$_3$ and at 2.5 T in CePt$_5$Ge$_3$, which leads to a large negative magnetoresistance. A large shoulder in the heat capacity and a broad maximum in the magnetic susceptibility of CePt$_5$Ge$_3$ above the long-range order at 1.1 K are striking features which deserve further investigations. They suggest pronounced low-dimensional AF short-range correlations.

Acknowledgments

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