Journal of Alloys and Compounds 509 (2011) 1436–1439

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: www.elsevier.com/locate/jallcom

Antiferromagnetic ordering below 1.7 K in $PrIr_2Ge_2$

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ARTICLE INFO

Article history:
Received 30 May 2010
Received in revised form
10 November 2010
Accepted 15 November 2010
Available online 23 November 2010

PACS:
75.50.Ee
65.40.Ba
71.70.Ch

Keywords:
Intermetallic compound
Antiferromagnetic ordering
Magnetization
Specific heat
Crystal field effect

ABSTRACT

We present our investigations on magnetic and transport properties of polycrystalline $PrIr_2Ge_2$ which forms in $CaBe_2Ge_2$-type primitive tetragonal structure (space group *P4/nmm*). The ac magnetic susceptibility data exhibit two well pronounced peaks at 2.08 K and 0.76 K due to the onset of magnetic order. The specific heat also exhibits a sharp λ-type anomaly at 1.7 K confirming the onset of bulk antiferromagnetic order. The temperature dependence of magnetic part of entropy suggests a quasi-triplet ground state in this compound. The onset of magnetic order is also confirmed by the electrical resistivity data.

1. Introduction

Ternary rare earth intermetallic compounds RT_2X_2 (R = rare earth, T = transition metal and X = Si, Ge) are well known for their diverse magnetic and superconducting properties, and correlated electron phenomena like heavy-fermion behaviour, unconventional superconductivity, non-Fermi-liquid behaviour, quantum criticality, etc. [1–8]. While the majority of RT_2X_2 compounds crystallize with the $ThCr_2Si_2$-type body centered tetragonal structure (space group *I4/mmm*), there are few which form in a lower symmetry $CaBe_2Ge_2$-type primitive tetragonal structure (space group *P4/nmm*). Both the structures are the ternary derivatives of the $BaAl_4$-type structure and differ from each other in the distribution of layers of T and X atoms [9]. The RT_2X_2 compounds crystallizing with the body centered $ThCr_2Si_2$ structure have more symmetry compared to the primitive tetragonal $CaBe_2Ge_2$ structure which lacks in a mirror symmetry about the R-plane along the *c*-axis. The local symmetry at the R-sites is considered very important for the magnetic and superconducting properties of compounds. Some of the RT_2X_2 compounds have even been found to adopt both the structures depending on the temperature and/or the conditions of synthesis. For example, the compounds like RIr_2Si_2 (R = La, Ce, Pr) [10–14], RNi_2As_2 (R = La, Ce, Pr, Nd, Sm) [15] and UCo_2Ge_2 [16] have $CaBe_2Ge_2$-type structure at high-temperature and transforms to $ThCr_2Si_2$-type structure at low-temperature. The heavy fermion compound $YbIr_2Si_2$, on the other hand, is found to crystallize in both $ThCr_2Si_2$-type and $CaBe_2Ge_2$-type structure depending on the starting stoichiometry and synthesis conditions [7].

In our effort to search for novel Pr-based compounds we have synthesized and investigated the magnetic and transport properties of a compound from the RT_2X_2 series, namely $PrIr_2Ge_2$, which is reported to crystallize with the $CaBe_2Ge_2$-type structure [17]. Pr-compounds have attracted the attention of condensed matter physicists by presenting the novel ground states which are determined by the crystal electric field (CEF) effect as in heavy fermion superconductor $PrOs_4Sb_{12}$ [18,19] and nonmagnetic heavy fermion $PrRh_2B_2C$ [20]. Recently we have reported our results of investigations on PrT_2Si_2 (T = Rh, Pd, Pt) [21–23]. While $PrRh_2Si_2$ and $PrPd_2Si_2$ (with $ThCr_2Si_2$-type structure) both order antiferromagnetically [21,22], $PrPt_2Si_2$ (with $CaBe_2Ge_2$-type structure) exhibits charge density wave (CDW) behaviour without any magnetic order down to 2 K [22,23]. Further, the transition temperature T_N of 68 K in $PrRh_2Si_2$ is found to be anomalously high in the family of RRh_2Si_2 (R = rare earths) compounds [24–31]. In this paper we present our results on $PrIr_2Ge_2$ based on dc magnetization, ac susceptibility,

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doi:10.1016/j.jallcom.2010.11.095

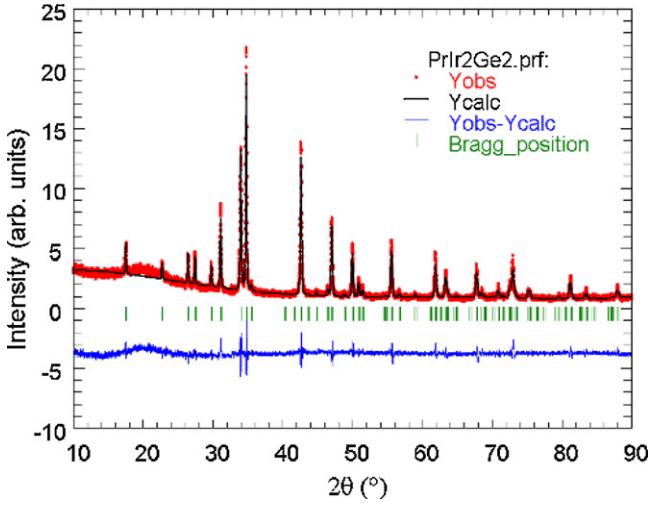

Fig. 1. Powder X-ray diffraction pattern of $PrIr_2Ge_2$ recorded at room temperature. The solid line through the experimental points is the Rietveld refinement profile calculated for $CaBe_2Ge_2$-type tetragonal P4/*nmm* structural model. The vertical short columns indicate the Bragg positions. The lowermost curve represents the difference between the experimental and model results.

specific heat and electrical resistivity measurements. Its Ce-analog, $CeIr_2Ge_2$ which also forms in $CaBe_2Ge_2$-type structure, is a Kondo lattice system that exhibits heavy fermion and non-Fermi liquid behaviour [32,33], and the homologous compound $PrIr_2Si_2$ orders antiferromagnetically below 47 K [34]. Recent investigations on single crystal samples show that $PrIr_2Si_2$ exhibits polymorphism, and orders below 45.5 K with another transition at 23.7 K in $ThCr_2Si_2$-type structure, and remains paramagnetic down to 2 K in $CaBe_2Ge_2$-type structure [13,14].

2. Experimental

We prepared polycrystalline samples of $PrIr_2Ge_2$ by arc melting the high purity elements (99.9% and above) in stoichiometric composition on a water cooled copper hearth under argon atmosphere. To improve the homogeneity and reaction among the constituent elements the samples were flipped and remelted several times during the arc melting process. The overall weight loss after five melts was less than 0.2%. The as-obtained samples were sealed inside the quartz tube under vacuum and annealed at 1000 °C for a week. The phase purity was checked by X-ray diffraction and high resolution scanning electron microscope (SEM) images. The stoichiometry was checked by EDAX composition analysis. A commercial SQUID magnetometer was used for magnetic susceptibility measurement. The specific heat was measured by relaxation method in a physical properties measurement system (PPMS, Quantum-design). Electrical resistivity was measured by the conventional four probe ac technique. The millikelvin temperature was reached by means of an adiabatic demagnetization refrigerator (Cambridge Magnetic Refrigeration). We also prepared the polycrystalline nonmagnetic analog $LaIr_2Ge_2$ by the same procedure as described above.

3. Results and discussion

Fig. 1 shows the X-ray diffraction pattern together with the Rietveld refinement profile obtained on the powdered $PrIr_2Ge_2$ sample. The X-ray diffraction pattern is well represented by the $CaBe_2Ge_2$-type primitive tetragonal structure (space group *P4/nmm*). The best fit of Rietveld refinement using least squares had the χ^2 value of 2.35. The crystallographic parameters are tabulated in Table 1. The lattice parameters so obtained are $a = 4.238(1)$ Å and $c = 10.101(1)$ Å, and the unit cell volume = 181.40(2) Å^3. The nonmagnetic analog $LaIr_2Ge_2$ also forms in the same $CaBe_2Ge_2$-type primitive tetragonal structure (space group *P4/nmm*) with lattice parameters $a = 4.269(2)$ Å and $c = 10.145(4)$ Å, and the unit cell volume = 184.89(7) Å^3. The structure and lattice parameters obtained for $PrIr_2Ge_2$ and $LaIr_2Ge_2$ are consistent with the reported values [17]. No impurity phase was detected in XRD or SEM images.

Table 1
The refined atomic coordinates for $PrIr_2Ge_2$.

Atom	Wyck	*x*	*y*	*z*
Pr	2c	0.25000	0.25000	0.74737
Ir1	2c	0.25000	0.25000	0.37315
Ir2	2a	0.75000	0.25000	0.00000
Ge1	2c	0.25000	0.25000	0.13110
Ge2	2b	0.75000	0.25000	0.50000

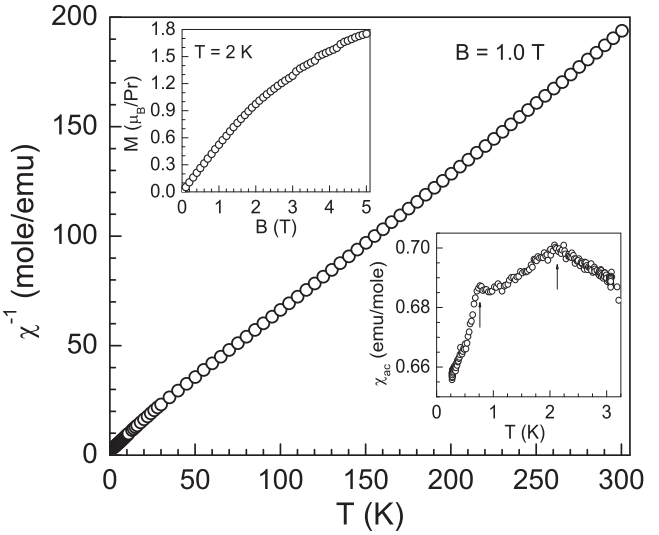

Fig. 2. Temperature dependence of dc magnetic susceptibility of $PrIr_2Ge_2$ plotted as χ^{-1} (T) in the temperature range 2–300 K measured in a field of 1.0 T. The lower inset shows the low temperature ac magnetic susceptibility data measured by using an adiabatic demagnetization refrigerator (CMR), and the upper inset shows the dc isothermal magnetization $M(B)$ as a function of magnetic field at a constant temperature of 2 K.

EDAX composition analysis confirmed the desired stoichiometry of 1:2:2.

Fig. 2 shows the results of magnetization measurements on $PrIr_2Ge_2$. The low temperature ac susceptibility data show two sharp anomalies at 2.08 K and 0.76 K due to the onset of magnetic order (lower inset of Fig. 2). In the paramagnetic state the magnetic susceptibility data is consistent with Curie–Weiss behaviour. A fit to the inverse magnetic susceptibility data above 50 K gives an effective moment $\mu_{eff} = 3.56\ \mu_B$ and Curie–Weiss temperature $\theta_p = -4.82$ K. The effective moment so obtained is very close to the theoretically expected value of 3.58 μ_B for Pr^{3+} ions. The upper inset of Fig. 2 shows the magnetic field dependence of isothermal magnetization $M(B)$ measured at 2 K. Isothermal magnetization at 2 K exhibits only slight nonlinearity suggesting the presence of short range order at this temperature.

Fig. 3 shows the results of specific heat measurements on $PrIr_2Ge_2$. The specific heat data show a sharp λ-type anomaly at 1.7 K confirming the intrinsic nature of bulk antiferromagnetic order in this compound. We also observe an upturn in specific heat below 0.6 K likely related to the second transition as observed in ac magnetic susceptibility. We could not investigate the behaviour below 0.4 K due to the limitation of measurement facility available. The magnetic contribution to the specific heat of $PrIr_2Ge_2$ which was obtained by subtracting the lattice contribution assuming it to be roughly equal to the specific heat of $LaIr_2Ge_2$ shows a rapid increase in C_{mag} above 12 K due to the influence of crystal field effect. From the temperature dependence of magnetic entropy we find that S_{mag} attains a value of $R\ln(3)$ at 3.9 K (inset of Fig. 3) suggesting the CEF-split ground state of $PrIr_2Ge_2$ to be a quasi-triplet. Estimation of Sommerfeld coefficient γ is difficult due to the influ-

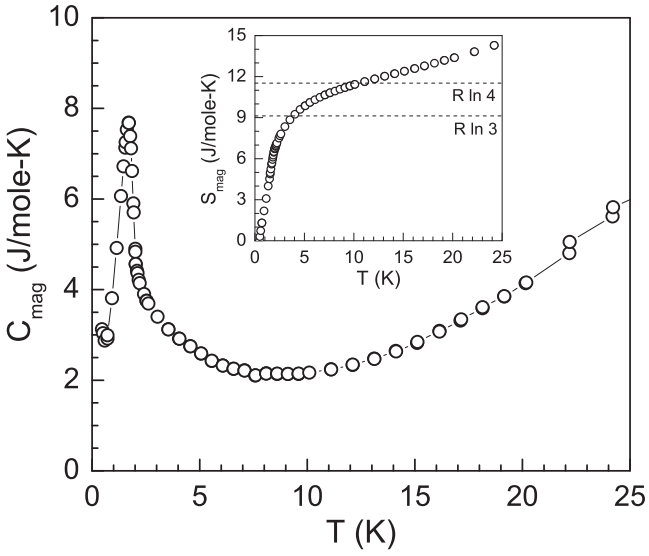

Fig. 3. Temperature dependence of magnetic contribution to the specific heat of $PrIr_2Ge_2$ measured in the temperature range 0.4–25 K. The inset shows the magnetic entropy as a function of temperature.

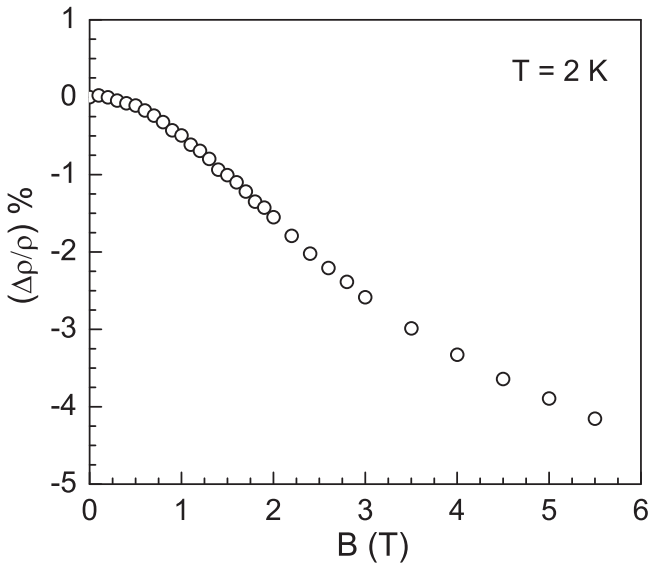

Fig. 5. Magnetic field dependence of electrical resistivity at 2 K normalized as magnetoresistance.

ence of magnetic order below the transition and the presence of crystal field effect above the transition.

Fig. 4 shows the electrical resistivity data of $PrIr_2Ge_2$. The resistivity data also evidence the onset of magnetic order at low temperature (upper inset of Fig. 4). At higher temperature (in the paramagnetic state) the resistivity data present the metallic behaviour. The resistivity is seen to decrease almost linearly with temperature down to 15 K below which it starts deviating, resulting in a broad hump near 5 K (lower inset of Fig. 4); below 2.3 K it becomes almost constant that persists down to 1.5 K where the sample undergoes a magnetic phase transition leading to a sharp decrease in resistivity due to the reduction in spin disorder scattering. The reason for the broad hump near 5 K is not very clear, however, we think that it is likely related to the crystal field effect; the presence of closely situated excited crystal field levels is clearly revealed from the temperature dependence

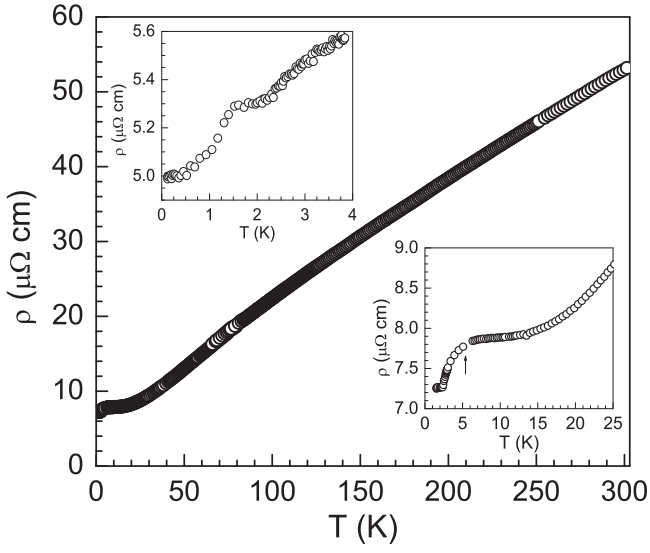

Fig. 4. Temperature dependence of electrical resistivity ρ (T) of $PrIr_2Ge_2$ measured in zero magnetic field. The lower inset shows the expanded view of low temperature resistivity and the upper inset shows the low temperature resistivity measured by using an adiabatic demagnetization refrigerator (CMR).

of entropy (the inset of Fig. 3). The low value of residual resistivity (~7 μΩ-cm at 1.7 K) with a residual resistivity ratio ~7 indicates our sample to be of good quality. We also measured the electrical resistivity as a function of magnetic field which is shown in the lower inset of Fig. 5. The data are plotted as magnetoresistance ($\Delta\rho/\rho(0) = [\rho(B) - \rho(0)]/\rho(0)$, where $\rho(B)$ is the resistivity measured at a magnetic field B). The magnetoresistance at 2 K is negative as expected in the paramagnetic state, and attains a value of ~4% at 5 T.

We thus see that magnetic susceptibility, specific heat and electrical resistivity present clear evidence of antiferromagnetic order in $PrIr_2Ge_2$. The reproducibility of the results discussed in this paper has been checked by performing the similar measurements on second batch of sample.

4. Conclusion

To conclude, we have investigated the magnetic and transport properties of a ternary intermetallic compound $PrIr_2Ge_2$. From the Rietveld refinement on powder X-ray diffraction data we confirmed the crystal structure to be $CaBe_2Ge_2$-type primitive tetragonal structure (space group *P4/nmm*). Two sharp anomalies observed in magnetic susceptibility data reveal the onset of magnetic order. While the first anomaly at 2.08 K is related to the antiferromagnetic order, the second anomaly at 0.76 K occurs due to spin reorientation. The specific heat and electrical resistivity data indicate the onset of bulk antiferromagnetic order below 1.7 K with a quasi-triplet ground state. Strong influence of crystal field effect is also observed in magnetic part of specific heat as well as electrical resistivity data.

Acknowledgment

We would like to thank R.S. Sannabhadti and Anil Kumar for their technical assistance during measurements with adiabatic demagnetization refrigerator (Cambridge Magnetic Refrigeration).

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