ELSEVIER

Physica B 223&224 (1996) 316–318

PHYSICA B

Magnetic behaviour of new Ce compounds

E.V. Sampathkumaran*, R. Mallik

Tata Institute of Fundamental Research, Homi Bhabha Road, Bombay 5, India

Abstract

We report initial results of our investigation on the magnetic behaviour of some new Ce compounds. The compounds, $CeIr_2B_2C$ and $CeIr_2Ge_2$, do not appear to exhibit bulk magnetic ordering down to 2 K. The alloys, Ce_2Pd_2In and Ce_2Cu_2In, order magnetically below 4 and 6 K, respectively, and a marginal change in the Pd(Cu) /In composition does not significantly influence the ordering temperatures.

Considering current interest on the physical properties of Ce based compounds, we report here the initial results of our investigation on the compounds $CeIr_2B_2C$, $CeIr_2Ge_2$, $CePd_{2+x}In_{1-x}$ and $Ce_2Cu_{2+x}In_{1-x}$ ($x = 0.0$ and 0.4). Crystallographic and preliminary magnetic investigations on $CeIr_2Ge_2$ [1], Ce_2Pd_2In (ferromagnetic, $T_c = 4$ K [2]) and Ce_2Cu_2In (antiferromagnetic, $T_N = 6$ K [2]) have been reported in the recent literature.

All the alloys were prepared by arc melting stoichiometric amounts of constituent elements and the molten ingots (except in the case of the Cu alloys) were annealed in evacuated sealed quartz tubes ($CeIr_2B_2C$ at 900°C for 18 days; $CeIr_2Ge_2$ and $Ce_2Pd_{2+x}In_{1-x}$ at 800°C for 7 days). The samples were characterized by X-ray diffraction. We find that $CeIr_2B_2C$ crystallizes in the $BaAl_4$-derived tetragonal structure, unlike $CeIr_2B_2$ [3] which forms in the $CaRh_2B_2$-type orthorhombic structure (see Fig. 1). Most of the X-ray diffraction lines in the Pd and Cu alloys could be assigned to Mo_2FeB_2-type tetragonal structure. A few weak extra lines were present even in the samples of Hulliger and Xue [2] and we notice that the intensity of these extra lines gets reduced considerably for $x = 0.4$. Magnetic susceptibility (χ), isothermal magnetization (M) (employing a superconducting quantum interference device), heat capacity (C) (by a semi-adiabatic heat-pulse method) and electrical resistivity (ρ) measurements were performed selectively on these samples. We do not attach much significance to the absolute values of ρ due to several cracks in the samples due to brittleness.

The results of C, M, χ and ρ measurements are plotted in Fig. 2 for $CeIr_2B_2C$. There is no prominent λ-type anomaly in the plot of C versus T which signals the absence of bulk magnetic ordering above 2 K. Two very weak peaks at about 3.5 and 5.8 K might arise from traces of Ce ordering magnetically due to local inhomogeneities. The value of C/T extrapolated to absolute zero from the data in the temperature range 10–20 K turns out to be about 40 mJ/mol K^2. With respect to the M behaviour, at 2 and 5 K, M does not saturate till 55 kOe, although the plots of M versus H are non-linear presumably from a small fraction of magnetic Ce ions. The plot of inverse χ versus T is not linear in the temperature range of investigation due to the combined influence of the crystal field and Kondo effects. The effective moment (μ_{eff}), if obtained from the data in the temperature interval 200–300 K, is larger (about $3.1\mu_B$) than that of free Ce^{3+} ion, which can be attributed to the polarization of the conduction band. The corresponding value of the paramagnetic Curie temperature (θ_p) is

* Corresponding author.

PII S0921-4526(96)00110-X

about − 350 K, as if this alloy is characterized by a large Kondo temperature (T_K) . It is at present not clear whether the apparently large values of μ_{eff} and θ_p are due to an artefact of crystal-field effects on χ. With respect to

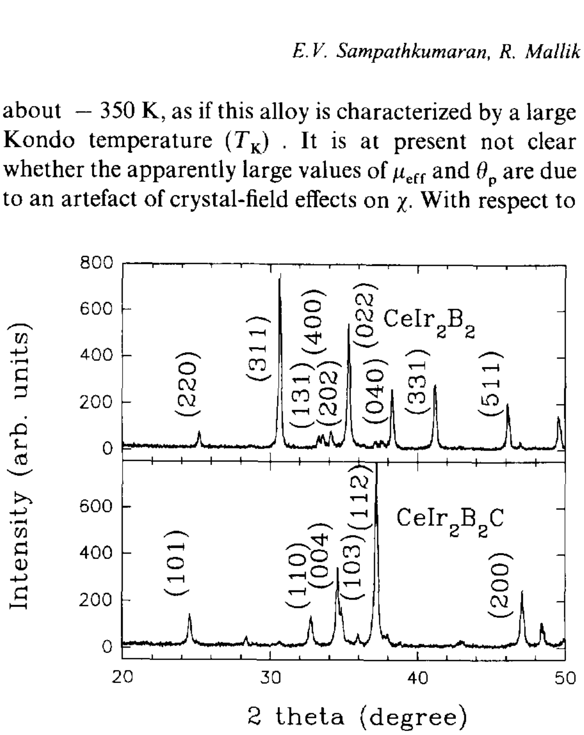

Fig. 1. X-ray diffraction patterns (Cu–K_α) of $CeIr_2B_2$ and $CeIr_2B_2C$.

the behaviour of resistivity, a Kondo minimum in the plot of ρ versus T appears at about 20 K. A drop in ρ below 6 K is also noted which presumably might arise from Kondo coherence effects or from the presence of traces of magnetic Ce impurities. Finally, it may be recalled that $CeIr_2B_2$ was shown to undergo ferromagnetic ordering at 6 K, with negligible contribution from the Kondo effect [3]. Thus, the main finding we bring out is that the addition of carbon to $CeIr_2B_2$ modifies crystallographic and magnetic behaviour.

The results of χ, M, C and ρ for $CeIr_2Ge_2$ are shown in Fig. 3. The temperature dependent behaviour of χ is quite complex: there is a high temperature Curie–Weiss region (above 180 K, $\mu_{eff} = 2.8\mu_B$ and $\theta_p = -190$ K), a tendency for a broad peak in the temperature interval 60–100 K, an increase below 60 K, then a near-constant value in the range 6–10 K (which was mentioned in Ref. [1] as well) and, finally, an increase at lower temperatures. The behaviour above 60 K (large θ_p and flattening of χ in the interval 60–100 K) suggests that this alloy could be characterized by a rather large spin fluctuation temperature. C does not exhibit any prominent peak above 2 K, which implies that the low temperature

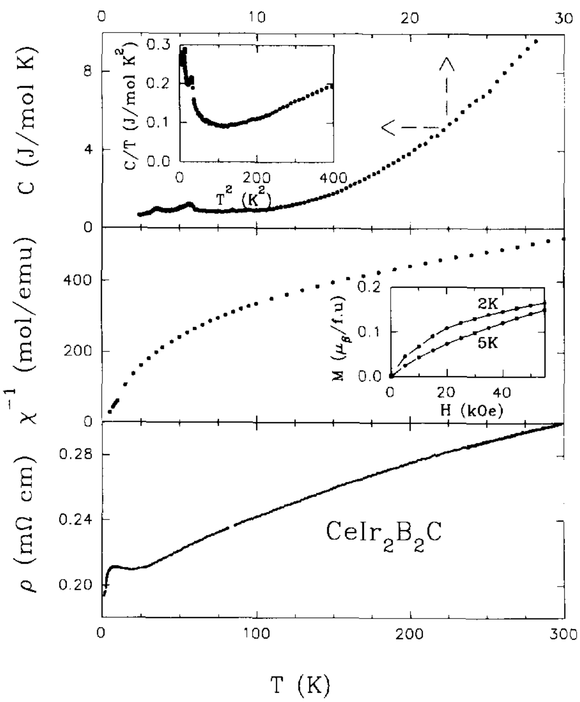

Fig. 2. Heat capacity (C), inverse susceptibility (χ) and electrical resistivity (ρ) as a function of temperature (T), for $CeIr_2B_2C$. C/T versus T^2 and isothermal magnetization are also shown in the insets.

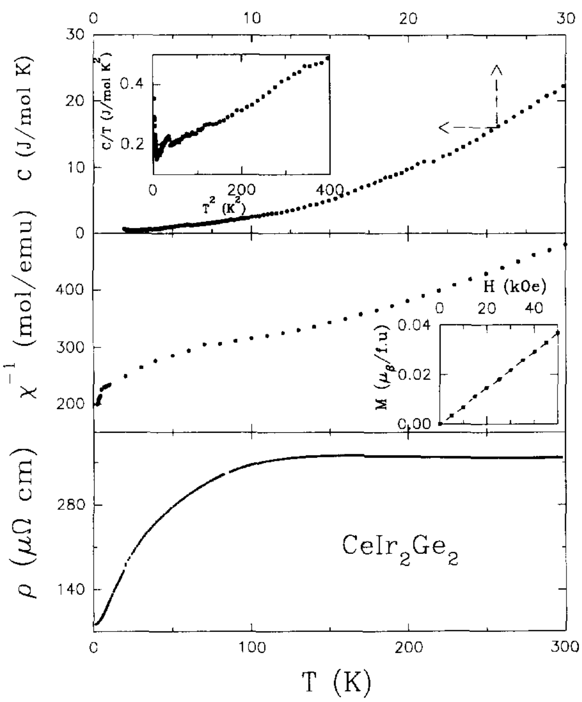

Fig. 3. Magnetic susceptibility (χ), heat capacity (C) and electrical resistivity as a function of temperature (T) for $CeIr_2Ge_2$; C/T versus T^2 and isothermal magnetization at 5 K are shown in the insets.

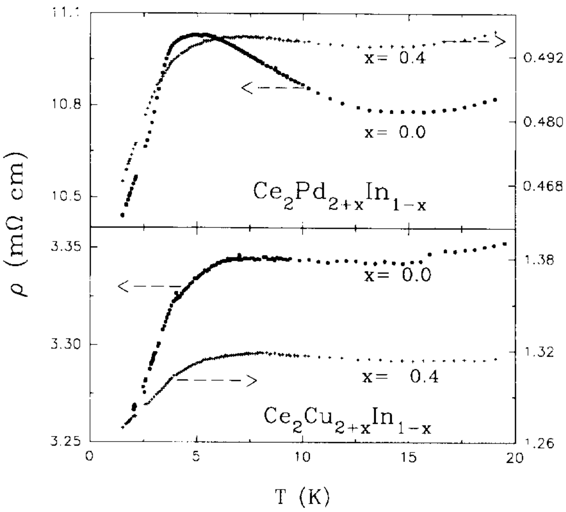

Fig. 4. Temperature dependent electrical resistivity behaviour in the vicinity of magnetic ordering temperature for the alloys $Ce_2Cu_{2+x}(Pd_{2+x})\ In_{1-x}$ (x = 0.0 and 0.4).

anomalies in χ do not arise from magnetic ordering and that this alloy is essentially non-magnetic above 2 K. The plot of C/T versus T^2 is nearly linear below 20 K and the value of extrapolated C/T (to absolute zero) is about 130 mJ/mol K^2, characteristic of (moderate) heavy-fermions. The electrical resistivity appears to vary linearly in the temperature interval 2–6 K and, in this sense, this alloy exhibits non-Fermi liquid behaviour. ρ drops below 90 K continuously, which presumably arises from Kondo coherence effects, and it saturates above 100 K.

On the basis of χ data, Hulliger et al. [2] concluded that the new Ce compounds, Ce_2Cu_2In and Ce_2Pd_2In, crystallizing in the Mo_2FeB_2-type structure, undergo antiferro- and ferromagnetic ordering at 5.6 and 4 K, respectively [2]. The present investigation involving the study of additional compositions is mainly motivated by a recent report [4] that there is a drastic reduction of magnetic ordering temperature from 42 to 13 K as x is increased from 0 to 0.4 in the ternary system, $U_2Pd_{2+x}Sn_{1-x}$. In our alloys, the drop in the resistivity data (Fig. 4) for $x = 0.0$ at low temperatures confirms the existence of a magnetic phase transition as indicated by χ data [2]. The point of emphasis is that the magnetism is not depressed as x is increased to $x = 0.4$ in sharp contrast to the situation in the U isomorphous alloys; indeed, there is a marginal upward shift of the ρ versus T plot in the vicinity of magnetic ordering temperature for $Ce_2Pd_{2.4}In_{0.6}$ relative to that of Ce_2Pd_2In as though T_c has increased for $x = 0.4$. It may also be noted that there is a resistivity minimum around 12 K, confirming the existence of the Kondo effect in these alloys. We have also performed C measurements on one of the alloys, viz. Ce_2Pd_2In, and the results, besides confirming the existence of magnetic ordering at 4 K, yield a C/T value, obtained by linear extrapolation of the data in the range 8–20 K to absolute zero, of about 200 mJ/mol K^2, possibly arising from electronic contribution.

Summarizing, the alloying of $CeIr_2B_2$ with carbon modifies magnetic and crystallographic behaviour significantly. The alloy $CeIr_2Ge_2$ exhibits complex temperature dependent χ behaviour; the results suggest that this compound apparently is a non-magnetic, moderate heavy-fermion (down to 2 K), however, with a non-Fermi liquid behaviour in the low temperature ρ data. Variations in the stoichiometry of the alloys $Ce_2Cu_2(Pd_2)$ In do not depress magnetism in contrast to the situation in isomorphous U alloys. We plan to carry out further characterization of these materials.

References

[1] N.D. Mathur and C.D. Frost, J. Alloys Compounds 215 (1994) 325.

[2] F. Hulliger and B.Z. Xue, J. Alloys Compounds 215 (1994) 215; F. Hulliger, private communication.

[3] E.V. Sampathkumaran and I. Das, Phys. Rev. B 51 (1995) 8628.

[4] F. Mirambet, L. Fournes, B. Chevalier, P. Gravereau and J. Etourneau, J. Magn. Magn. Mater. 138 (1994) 244.