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Specific heat of $CeNi_{1-x}Cu_x$ in the 0.2 to 300 K temperature range ☆

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Abstract

We present the specific heat measurement of the $CeNi_{1-x}Cu_x$ series in a large temperature range (0.2–300 K). The high temperature measurements allow us to estimate the crystalline electric field splitting which is proximately the same for all the studied compounds, while in the low temperature range we observe two kinds of magnetic transitions related to long-range order and spin-glass state. The estimate of the γ coefficients yields the highest values for the $CeNi_{0.8}Cu_{0.2}$ compound, while a continuous increase of C/T when $T \to 0$ K is observed for $x = 0.15$, indicating a possible non-Fermi liquid behaviour. © 2002 Elsevier Science B.V. All rights reserved.

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The competition between the magnetic RKKY, Kondo and crystalline electric field (CEF) interactions plays an important role in determining the magnetic properties in the $CeNi_{1-x}Cu_x$ compounds. Long-range antiferro or ferromagnetic order and a spin-glass-like state were found changing the Cu concentration [1]. The microscopic nature of these magnetic states is being studied by μSR spectroscopy [2], whereas in the actual paper we present a macroscopic specific heat study which provides useful information about the energy levels of the Ce^{3+} ions, the density of states at the Fermi level and the 4f-conduction band hybridisation.

Specific heat measurements between 0.2 and 6 K have been carried out at the ICMA on an adiabatic demagnetisation calorimeter in the polycrystalline $x = 0.15, 0.2, 0.3, 0.4, 0.5$ and 0.6 compounds whereas measurements between 2 and 300 K have been performed using a quantum design microcalorimeter at the University of Cantabria in the $x = 0.2, 0.3, 0.4, 0.5, 0.6, 0.7,$ 0.8 and 0.9 compounds.

Fig. 1 shows the specific heat versus temperature for all the studied compounds. In the common temperature range (2–6 K) the matching between both sets of data is excellent.

The anomaly observed for $x = 0.9$ corresponds to the antiferromagnetic transition ($T_N = 2.5$ K) defined as the inflexion point above the maximum of the $C(T)$ curve. For $x = 0.6$, the maximum centred around 2 K corresponds to the spin-glass freezing temperature and the small λ-type anomaly corresponds to the Curie temperature, $T_C = 1.1$ K [3]. The long-range ordering temperatures have been corroborated by neutron diffraction experiments [4]. For the $x = 0.5, 0.4$ and 0.3 compounds, abrupt anomalies appear around 2.5 K. However, they are not due to long-range magnetic ordering but to spin-glass freezing, according to the χ_{ac} and magnetisation (FC and ZFC) measurements performed in the same samples [1]. The shape of these anomalies suggests a kind of spin-glass arrangement which is inhomogeneous better than a canonical one, in

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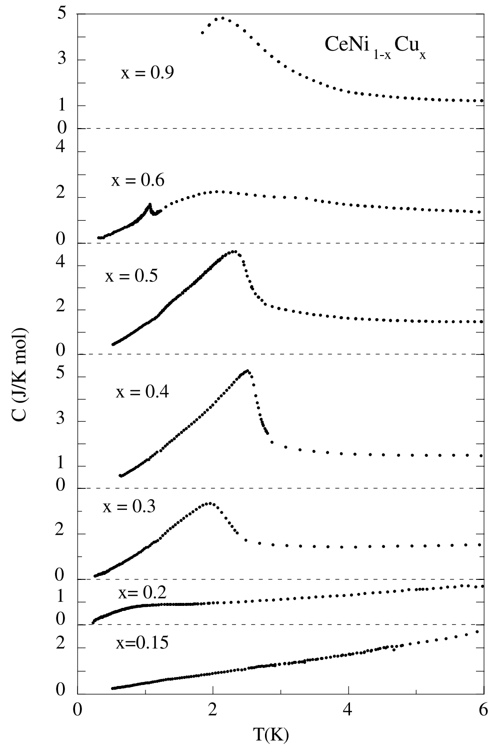

Fig. 1. Low temperature specific heat measured on the $CeNi_{1-x}Cu_x$ series.

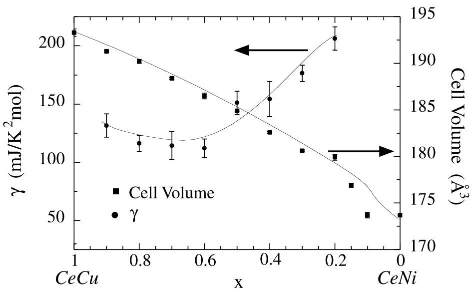

Fig. 2. Electronic specific heat coefficient (γ) and cell volume as a function of composition. Solid lines are guides for the eye.

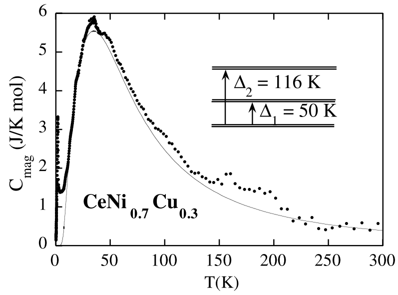

Fig. 3. Magnetic contribution to the specific heat for $CeNi_{0.7}Cu_{0.3}$. The solid line corresponds to a Schottky-type fit with $\Delta_1 = 50$ K, $\Delta_2 = 116$ K.

agreement with the results obtained from μSR spectroscopy [2]. Although expected [1], no other transitions at lower temperatures were detected, probably due to the strong reduction of the magnetic moments because of the enhancement of the Kondo effect. $CeNi_{0.8}Cu_{0.2}$ presents one slight anomaly around 1 K which is consistent with the μSR spectroscopy results [2], indicating a low temperature transition (probably long-range order). No evidence of magnetic anomalies were found for the $x = 0.15$ compound. However, the C/T versus T plot shows a continuous increase as $T \to 0$ K, suggesting the possible existence of a non-Fermi liquid behaviour as the magnetic–nonmagnetic crossover is reached.

The electronic coefficient (γ) is estimated by extrapolation of the linear part of the C/T versus T^2 plot for each alloy. In Fig. 2, we represent both the values of γ and the cell volume as a function of the Cu content. The γ value increases as the cell volume decreases, reaching the maximum value ($\gamma \simeq 200$ mJ/K^2 mol) for the $x = 0.2$ compound. This result reflects the increasing hybridisation effects as the Cu concentration decreases and confirms the close correlation between the increasing hybridisation and the decrease of the cell volume.

The magnetic contribution to the specific heat has been obtained for all the compounds taking the specific heat of the isomorphous non-magnetic compound YNi as the lattice contribution. A Schottky-type anomaly centred around 50 K is clearly observed for all the studied compounds. The magnetic contribution for $x = 0.3$ is presented as an example in Fig. 3. Due to the low-symmetry site occupied by the Ce^{3+} ion, the $J = \frac{5}{2}$ ground multiplet splits into three doublets. The excited states are separated from the ground state by energy gaps Δ_1 and Δ_2. Within this CEF scheme, the best Schottky-type fit to C_{mag} corresponds to $\Delta_1 \approx 50$ K and $\Delta_2 \approx 120$ K, being similar for all the compositions with $x \geqslant 0.2$. This means that the CEF splitting does not change significantly due to the small lattice constant variation induced by the dilution.

Our specific heat measurements confirm the general trends of the phase diagram of the $CeNi_{1-x}Cu_x$ system. Furthermore, they offer valuable information on the nature of the spin-glass arrangement. While the Ni substitution by Cu does not mainly affect the crystalline electric field interaction, it plays a major role in the evolution of the magnetic behaviour leading to the evanescence of the long-range magnetic order.

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