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Long-range magnetic ordering in the Kondo lattice $CeCuGa_3$

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Abstract

We present magnetic, calorimetric and transport properties of $CeCuGa_3$, which characterise $CeCuGa_3$ as a magnetically ordered Kondo lattice system. The Kondo temperature T_K is estimated to be between 5.6 and 6.9 K. For the first time this system is shown to exhibit long-range magnetic order at 1.9 K. The system shows negative magnetoresistance in fields of up to 8 T.

Keywords: $CeCuGa_3$; Cerium; Intermetallic alloys; Kondo lattices; Magnetic ordering

1. Introduction

Cerium-based intermetallic compounds exhibit a wide range of fascinating phenomena. In particular, compounds with the $ThCr_2Si_2$ derivative of the $BaAl_4$ superlattice have been investigated in great depth ever since the discovery of heavy-fermion superconductivity in $CeCu_2Si_2$. There are other cerium compounds which possess a more primitive $BaAl_4$ structure and show a wide range of interesting magnetic properties. For instance, $CePtGa_3$ has a spin-glass ground state [1], because of the randomisation of the local exchange coupling caused by atomic site-disorder, $CeCuAl_3$ shows Kondo lattice behaviour with the possible onset of antiferromagnetism at 2.8 K [2]. In Refs. [3,4], resistance, ac susceptibility and specific heat measurements indicated the possible competition between dense Kondo behaviour and ferromagnetism in the series $CeNi_xGa_{4-x}$ for $x = 0.75$, 0.875 and 1.1. This was also suggested by resistance measurements of compounds in the series $CeCu_xGa_{4-x}$ [3] and later by magnetoresistance measurements [5]. In an earlier investigation of the low-temperature properties of $CeCuGa_3$ [6], ac susceptibility measurements suggested that this compound remained paramagnetic down to 0.4 K, and that a rise in the specific heat between 5 and 1.4 K could be explained by a low-lying crystal field level. Recent published results [7] show the same behaviour as the earlier investigation [6], a rise in specific heat between 5 and 2 K, but still do not show conclusively the occurrence of magnetic ordering.

In this paper we present resistivity, specific heat, and ac susceptibility results which show that $CeCuGa_3$ exhibits long-range magnetic order with Kondo lattice behaviour. From these results it was anticipated that the ordering would be antiferromag-

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netic in nature. However, recent neutron diffraction measurements [8], while confirming long-range magnetic ordering, have shown that the ordering is incommensurate.

2. Experimental details

Polycrystalline samples were prepared by arc-melting of the constituent elements (Ce, La 99.9%; Cu, Ga 99.99%) in a titanium gettered argon atmosphere. The samples were then wrapped in tantalum foil and annealed in quartz tubes for two weeks at 600°C.

X-ray powder diffraction showed the samples to be single phase. This was supported by EDS measurements. Both $CeCuGa_3$ and $LaCuGa_3$ have been found to crystallise in the primitive tetragonal $BaNiSn_3$-type structure (space group I4/mm) [9]. From an analysis of the X-ray patterns the lattice parameters of $CeCuGa_3$ and $LaCuGa_3$ were calculated to be $a = 4.24$ Å and $c = 10.44$ Å and $a = 4.26$ Å and $c = 10.46$ Å, respectively. These values are in good agreement with previously reported values [9].

Resistance measurements were performed from 0.3 to 275 K using a conventional four-probe method. Susceptibility measurements were made using a mutual inductance technique in the temperature range 1.4–150 K. The specific heat being measured by a fully automated Nernst calorimeter from 1.5 up to 100 K. Magnetoresistance measurements were performed in magnetic fields of up to 8 T in the temperature range 1.9–250 K.

3. Results

Fig. 1 shows the resistivity versus temperature data for $CeCuGa_3$. The inset shows more detail of the low-temperature behaviour. A broad hump is seen around 100 K, which is typically caused by the interaction between crystal fields and the Kondo effect [10]. With decreasing temperature a shallow minimum is observed at 10 K which is characteristic of Kondo scattering in the crystal field ground state. A sharp drop is seen at 4.8 K which continues down to the limit of the measurements at 0.3 K. This sharp drop is indicative of coherence effects between the

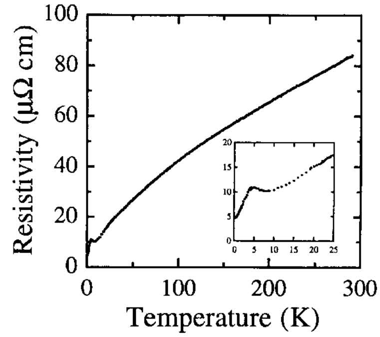

Fig. 1. Temperature dependence of the resistivity ρ of $CeCuGa_3$ between 0.3 and 275 K. The inset shows more detail of the low-temperature behaviour.

scattered electrons seen in Kondo lattice systems. The presence of microcracks in this sample leads to a reduced cross-section and produces the high residual resistivity observed. However, several samples of $CeCuGa_3$ prepared for this study show the same resistive behaviour, but with differing residual resistivities and therefore absolute resistivities. $CeCuGa_3$ can be conclusively shown to be a Kondo lattice if the magnetic contribution to the resistivity shows $-\ln T$ behaviour over a significant temperature range. The magnetic contribution to the resistivity is obtained by subtraction of the isostructural non-magnetic analogue $LaCuGa_3$. Unfortunately, the presence of microcracks in both $CeCuGa_3$ and $LaCuGa_3$ makes this impossible. However, a sensible fit to the experimental data can be achieved using an equation that includes, a constant residual resistivity term, a term which is proportional to T for the phonon contribution, and a $-\ln T$ term for the Kondo effect. This fit can only be made over two temperature ranges: 5–35 and 210–250 K. The resistivity cannot be fitted over the whole temperature range because of the influence of crystal field effects around 100 K and coherence effects below 5 K which cannot be incorporated into this simple model.

Fig. 2 shows the temperature dependence of the magnetic susceptibility χ from 1.4 up to 20 K. For higher temperatures (not shown) $\chi(T)$ exhibits a Curie–Weiss behaviour which gives an effective moment of about 2.6 μ_B close to that of the free Ce^{3+} ion. A peak in $\chi(T)$ is seen at 1.8 K, indicating a transition to a magnetically ordered state.

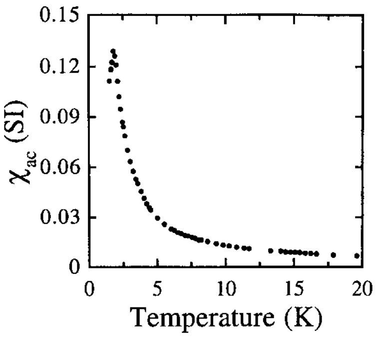

Fig. 2. Temperature dependence of the ac susceptibility χ of $CeCuGa_3$ between 1.4 and 20 K.

The specific heat of $CeCuGa_3$ plotted as C_p/T versus T^2 is shown in Fig. 3. A peak in the data is observed at 1.9 K, confirming the onset of long-range magnetic ordering. From the shape of the peak the ordering is probably antiferromagnetic. Extrapolation of the C/T versus T^2 data between 20 and 10 K to 0 K, yields a value for the electronic contribution to the specific heat $\gamma(T=0)$ of about 150 mJ mol^{-1} K^{-2}. This value is similar to that reported for magnetically ordered Kondo lattice systems, such as $CeCu_2$ and $CeAl_2$, in which the enhancement of the value of γ is attributed to Kondo compensation of the local magnetic moment by the 4f conduction electrons and consequent increase in the density of states at the Fermi level. The magnetic contribution to the specific heat, C_m is shown in the inset of Fig. 3. C_m is obtained by subtraction of the specific heat of paramagnetic $LaCuGa_3$ from C_p. A Schottky anomaly is shown for comparison, the best fit being achieved with a crystal field splitting between the ground state of about 47 K. This may describe part of the low-temperature specific heat data, but no feature is observed in the resistivity at this temperature, as would be expected. Integrating between 0 K and T_N of C_m/T gives a total magnetic entropy, S_m, at T_N of about 2.6 J mol^{-1} K^{-1}. This is small in comparison with the value expected for ordering in the ground state doublet ($R \ln 2$). This may be caused by short-range order, but is probably due to the influence of the Kondo effect lifting the degeneracy of the ground state doublet. The magnetic entropy at the transition can be used to estimate the Kondo temperature, using the simplified model of Mori et al. [11], which is based on the theoretical work of Desgranges et al. [12]. From this, T_K is estimated to be 6.9 K.

An alternative phenomenological model which incorporates the Kondo effect by an additional Lorentzian density of states at the Fermi energy (with $\Delta = k_B T_K$) and long-range magnetic ordering in the scope of the mean field approximation gives a qualitative description of the temperature dependence of the specific heat [13]. The parameters of this model are the Kondo temperature, T_K, and the mean field coupling constant, J. Applying this model to the experimental data (Fig. 4) gives a reasonable fit for the peak position in specific heat with $T_K \approx 5.8$ K and $J \approx 14.1$ K. The moment reduction due to the Kondo effect based on these values of T_K and J amounts to approximately 35% of the ordering mo-

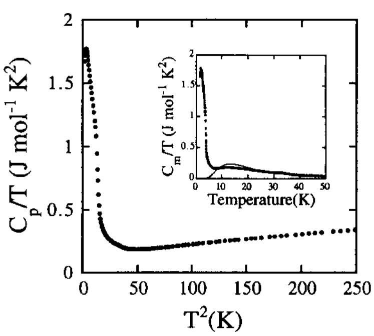

Fig. 3. Specific heat C_p of $CeCuGa_3$ shown as C_p/T versus T^2. The inset shows the magnetic subtraction to the specific heat between 0 and 50 K, with a Schottky anomaly shown for comparison.

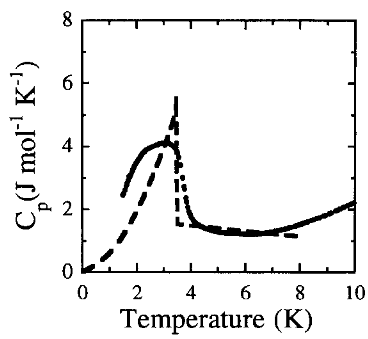

Fig. 4. Temperature dependence of the specific heat C_p of $CeCuGa_3$. The dotted line shows a fit to the data, incorporating long-range magnetic order and the Kondo effect.

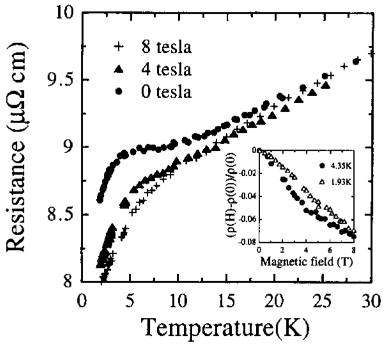

Fig. 5. The resistivity of $CeCuGa_3$ in magnetic fields of 0, 4 and 8 T. The inset shows the magnetoresistivity $[(\rho(H) - \rho(0))/\rho(0)]$ as a function of the applied field H at 1.93 and 4.35 K.

ment without the Kondo interaction. In this hypothetical case an ordering temperature of about 7 K is expected within the mean field approximation.

Previous measurements [6] on samples which contained an impurity phase (< 2 vol%) showed no magnetic order down to 0.4 K. The sensitivity of the properties of $CeCu_xGa_{4-x}$ to stoichiometry may explain this difference.

Fig. 5 shows the magnetoresistance in fields of 0, 4 and 8 T. The magnetoresistance is negative, as expected from suppression of the Kondo effect. The field dependence of the magnetoresistance $[(\rho(H) - \rho(0))/\rho(0)]$ at 1.93 and 4.35 K is shown in the inset to Fig. 5. It should be noted that there is a change in the field dependence between these two temperatures. At 1.93 K there appears to be a positive curvature of the magnetoresistance with respect to the magnetic field axis, whilst a negative curvature is seen at 4.35 K. The change in the nature of the field dependence is similar to that seen in $CeCu_{0.5}Ga_{3.5}$ between 10 and 5 K [5]. This change may be associated with the magnetic ordering in these compounds, but as no ordering temperature is given for $CeCu_{0.5}Ga_{3.5}$ a direct comparison cannot be made. From these measurements it appears that the ordering is antiferromagnetic in $CeCuGa_3$.

4. Conclusion

We have presented measurements on the properties of $CeCuGa_3$. A low-temperature minimum seen in the temperature dependence of the resistivity, and a sharp drop in the resistance at lower temperatures both support the suggestion that $CeCuGa_3$ is a Kondo lattice. The influence of Kondo scattering in the higher crystal field states can be seen from a broad feature in the resistivity around 100 K. The $-\ln T$ behaviour, which is conclusive evidence of Kondo behaviour, could not be reliably extracted due to the microcracks in the $CeCuGa_3$ and the non-magnetic analogue $LaCuGa_3$. The deficiency in the magnetic entropy at the ordering temperature and high γ in the specific heat confirms the occurrence of the Kondo effect. Specific heat and ac susceptibility measurements show the occurrence of long-range magnetic ordering at 1.9 K, which appears to be antiferromagnetic. Neutron diffraction measurements have confirmed the onset of long-range magnetic ordering, but show it to be incommensurate.

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