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LETTER TO THE EDITOR

Competition between magnetism and heavy fermion behaviour in $Ce_{1-x}La_xCu_3Ga_2$

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Abstract. The results of magnetic susceptibility and electrical resistance measurements on the pseudoternary solid solution $Ce_{1-x}La_xCu_3Ga_2$ are presented. The results, besides showing that $CeCu_3Ga_2$ is an incoherent Kondo lattice, indicate the existence of a competition between magnetism and non-magnetism in this system.

The subject of competition between magnetism and Kondo effect in Ce systems has gained considerable attention recently (see various articles in [1]). In this respect, the compound $CeCu_5$, crystallising in the hexagonal $CaCu_5$ structure, is of special interest. This compound has been identified to be a magnetically ordered Kondo compound ($T_N = 4$ K, $T_K = 5.5$ K) [2, 3]. It is reported that the replacement of Cu by Al or Ga (M) tends to depress magnetic ordering [3–8], though the unit cell volume, V, increases with x in $CeCu_{5-x}M_x$ ($x \leq 2$). This is the opposite of what is expected on the basis of the well known inverse relationship between V and T_K, the Kondo temperature characterising non-magnetism [9]. The observed reduction of T_N with x was, therefore, explained in terms of the changes in the conduction-electron density following the chemical substitution [4]. A similar interpretation has been offered earlier in connection with the competition between Kondo effect and magnetic ordering in $CeRh_{2-x}T_xSi_2$ (T = transition metal [10].

Among the pseudo-ternary alloys based on $CeCu_5$, the compound $CeCu_3Ga_2$ is of interest for the present investigation. This compound has been recently reported [11] to be a heavy-fermion compound, with an electronic contribution, γ, to the specific heat of the order of 730 mJ mol^{-1} K^{-2}; also, no magnetic ordering could be found down to 1.45 K. We considered it worthwhile to investigate the series $La_xCe_{1-x}Cu_3Ga_2$. As there is no change in the number of conduction electrons due to the replacement of Ce by La (unlike what happens in the $CeCu_{5-x}M_x$ series), the effect of the unit-cell volume alone on the magnetic behaviour of Ce could easily be tracked. Though this chemical substitution involves a change in the number of Ce ions, such an investigation might enable us to understand the competition between Kondo effect and magnetic ordering, as in the case of $Ce_{1-x}La_xCu_2Si_2$ [12]. In this Letter, we report the results of magnetic susceptibility, χ, and electrical resistance, R, measurements in the temperature range 4.2–300 K on the alloys $Ce_{1-x}La_xCu_3Ga_2$. The results clearly show that $CeCu_3Ga_2$ is an incoherent Kondo lattice (a compound with a periodicity of Kondo impurities without any evidence for coherence effects at low temperatures). Interestingly, there is a con-

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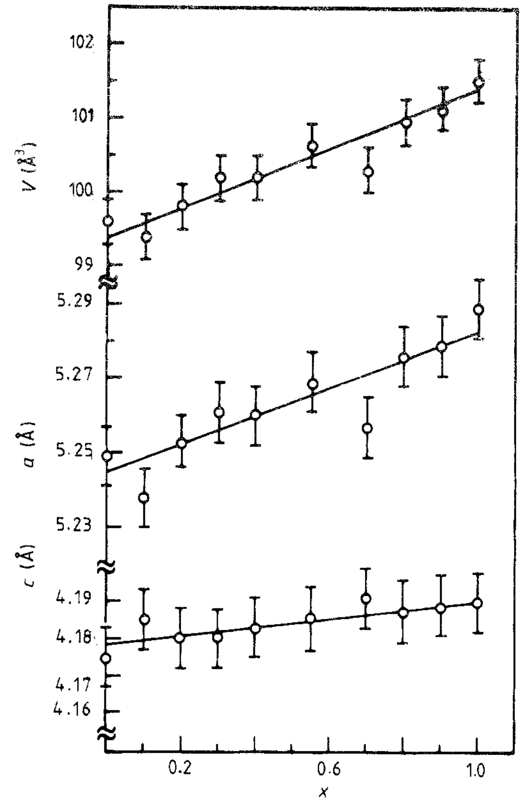

Figure 1. The lattice constants a, c and V (unit cell volume) as a function of x in $Ce_{1-x}La_xCu_3Ga_2$. The full lines through the data points are guides to the eye.

siderable enhancement of the low-temperature susceptibility for initial replacement of Ce by La, indicating the presence of magnetic ordering or higher γ-value for such specimens. This observation might demonstrate the competition between the Kondo effect and magnetism in this series of compounds.

The samples, $Ce_{1-x}La_xCu_3Ga_2$ (x = 0, 0.1, 0.2, 0.3, 0.4, 0.55, 0.7, 0.8, 0.9 and 1.0) were prepared from the master alloys $LaCu_3Ga_2$ and $CeCu_3Ga_2$ by arc melting. They were characterised by x-ray diffraction. Magnetic susceptibility measurements were carried out in a field of 4 kOe by the Faraday method. Resistance measurements were performed using a conventional four-probe method.

The results of the lattice-constant measurements are shown in figure 1. As expected, the lattice parameters, a, c and V, increase almost linearly with x. Therefore, the strength of the 4f conduction-electron coupling constant, J, must decrease with increasing unit-cell volume [9]. In figure 2, the results of measurements for selected concentrations of La are shown. Except for $x = 0.1$, the value of χ at any given temperature decreases with increasing x. For $x = 0.1$, χ below 20 K is much larger than for $CeCu_3Ga_2$. We believe that this observation is real, and not due to any impurity effect, since an identical batch of master alloys was used to prepare all the specimens. The anomalous enhancement of low-temperature susceptibility for $x = 0.1$ might imply that γ for this composition is probably much higher than that for $CeCu_3Ga_2$, as it is well known that $\chi(T \to 0) \propto \gamma$ (see for instance [13]). Alternatively, the increase in χ as $T \to 0$ might signal the onset of magnetic ordering at even lower temperatures. Further addition of La does not seem to favour this highly correlated state, as shown by the low-temperature susceptibility

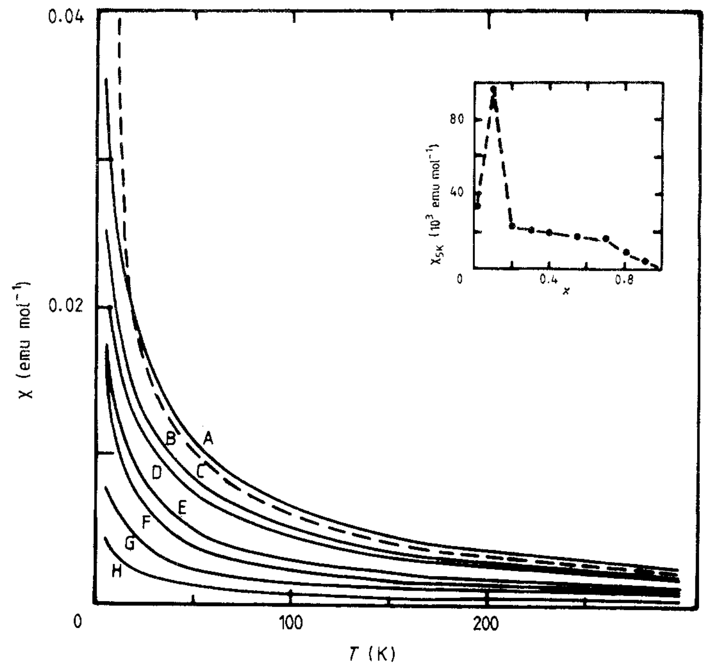

Figure 2. Magnetic susceptibility, χ, as a function of temperature for selected values of x in $Ce_{1-x}La_xCu_3Ga_2$: A, $x = 0$; B, $x = 0.1$; C, $x = 0.2$; D, $x = 0.3$; E, $x = 0.55$; F, $x = 0.7$; G, $x = 0.8$; H, $x = 0.9$. In the inset, χ at 5 K is shown as a function of x.

data. Considering that $\chi(0)$ and γ are inversely related to T_K [14], the data in the inset of figure 2 suggest that T_K as a function of x is minimal at $x = 0.1$.

Further evidence for the above conclusion comes from the high-temperature χ data. The plot of χ^{-1} against T is linear above 80 K (not shown in the figure) and the effective moment per Ce obtained from the slope of this plot is the same as that expected for trivalent Ce ion (2.54 μ_B) for all values of x. The paramagnetic Curie temperature (θ_p) is the same (−30 K) for all Ce-containing specimens, except for $x = 0.1$. As there is no evidence for magnetic ordering for $CeCu_3Ga_2$, θ_p is related to the Kondo temperature and hence this compound is a Kondo lattice, as suggested earlier [11]. Interestingly, the value of θ_p decreases to −20 K for $x = 0.1$. As is known [15], $T_K = m|\theta_p|$ and hence for this concentration alone, T_K decreases relative to that for $CeCu_3Ga_2$, as inferred above from the low-temperature data. Thus a competition between Kondo effect and magnetism exists in this series of compounds. Such a situation can be understood in the light of Doniach's Kondo necklace model [16]. According to this model, for a given density of conduction electrons, there is a critical value of J (which could be varied either by applying external pressure or by alloying), at which the magnetic ordering temperature (T_N) is a maximum; for higher values of J, non-magnetism dominates and for lower values of J, T_N can sharply decrease. The present results, therefore, indicate that the alloy $Ce_{0.9}La_{0.1}Cu_3Ga_2$ might lie somewhere near the peak in the Doniach's magnetic phase diagram. Further studies at very low temperatures, particularly for more concentrations below $x = 0.2$, would be helpful to clarify the nature of the ground state.

The results of electrical resistance measurements, normalised to their respective room-temperature values, are shown in figure 3 for selected specimens. Due to the

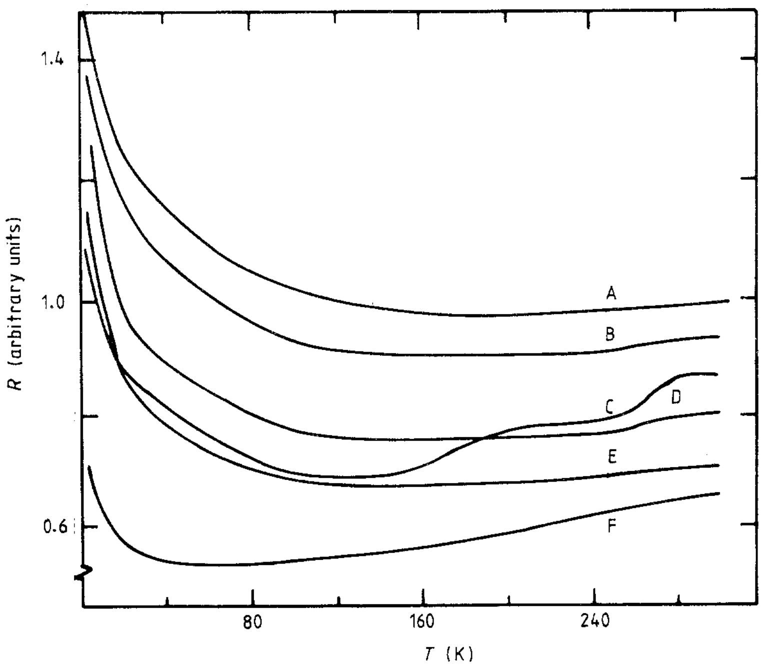

Figure 3. Resistance normalised to the respective 300 K value as a function of temperature in $Ce_{1-x}La_xCu_3Ga_2$ for selected values of x: A, $x = 0$; B, $x = 0.1$; C, $x = 0.2$; D, $x = 0.3$; E, $x = 0.4$; F, $x = 0.7$. For the sake of clarity, for $\chi > 0$, the curves are shifted downwards.

irregular shape of the specimens, it was rather difficult to obtain the absolute value of the resistivity. As noted earlier [11], for $CeCu_3Ga_2$, there is a monotonous logarithmic increase in R with decreasing temperature. This behaviour is caused by the scattering of conduction electrons by Kondo centres. There is no evidence for any decrease of R in the low-temperature region (below 10 K), such as is normally observed in other Ce-based Kondo lattices due to the onset of coherence [15]. Thus, $CeCu_3Ga_2$ is termed an incoherent Kondo lattice. Incoherence is possibly due to atomic disorder created by the random distribution of Ga atoms between 3g and 2c sites. The observation that the temperature-dependent resistive behaviour for the La-substituted specimens, even for high values of x, is similar to that of $CeCu_3Ga_2$ lends credence to the above views. For $x = 0.2$ there is a maximum in the R data around 160 K and this is attributed to crystal-field splitting [17]. It is at present not clear why a similar maximum is not seen for other concentrations.

Finally, it appears that the nature of the ground state of compounds of the series $CeCu_{5-y}Ga_y$ is not fully understood. For instance, there are conflicting reports on the interpretation of the available results for $CeCu_4Ga$. On the one hand, Willis and co-workers [18] claim that $CeCu_4Ga$ orders at 0.7 K from a high state, whereas Kohlmann and co-workers [6] report it to be a non-magnetic heavy-fermion compound. The need to carry out AC χ and specific-heat measurements under an applied field to clarify the situation has been stressed recently by Dhar and Gschneidner [19]. It is at present not clear whether similar discrepancies exist for $CeCu_3Ga_2$ on the question of a non-magnetic ground state [11]. However, such a situation would not alter our basic conclusion that initial substitution of La enhances a highly correlated state—whether it is a magnetic or a high-γ state.

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References

[1] *J. Magn. Magn. Mater.* 1988 **76–77**
[2] Bauer E, Gratz E and Schmitzer C 1987 *J. Magn. Magn. Mater.* **63 + 64** 37
[3] Bauer E, Gignoux D, Schmitt D and Winzer K 1987 *J. Magn. Magn. Mater.* **69** 158
[4] Bauer E, Pillmayr N, Gratz E, Gignoux D, Schmitt D, Winzer K and Kohlmann J K 1988 *J. Magn. Magn. Mater.* **71** 311
[5] Bauer E, Gratz E, Pillmayr N, Gignoux D, Schmitt D, Winzer K and Kohlmann J 1988 *J. Magn. Magn. Mater.* **76–77** 125
[6] Kohlmann J, Winzer K and Bauer E 1988 *Europhys. Lett.* **5** 541
[7] Bauer E, Gratz E and Pillmayr N 1987 *Solid State Commun.* **62** 271
[8] Bauer E, Pillmayr N, Gratz E, Gignoux D and Schmitt D 1987 *J. Magn. Magn. Mater.* **67** L-143
[9] Allen W and Martin R M 1982 *Phys. Rev. Lett.* **49** 1106
[10] Sampathkumaran E V, Dhar S K and Malik S K 1987 *J. Phys. C: Solid State Phys.* **20** 53
[11] Bauer E, Pillmayr N, Gratz E, Gignoux D and Schmitt D 1987 *Phys. Lett.* **124A** 445
[12] Aliev F G, Brandt N B, Moshchalkov Y Y and Chudinov S M 1984 *J. Low Temp. Phys.* **57** 61
[13] Newns D M and Hewson A C 1980 *J. Phys. F: Met. Phys.* **10** 2429
[14] Andrei N, Furuya K and Lowenstein H 1983 *Rev. Mod. Phys.* **55** 331
[15] Brandt N and Moshchalkov V V 1984 *Adv. Phys.* **33** 373
[16] Doniach S 1977 *Physica* B **91** 231
[17] Bhattacharjee A and Coqblin B 1976 *Phys. Rev.* B **13** 3441
[18] Willis J O, Aiken R H, Fisk Z, Zirngiebl E, Thompson J D, Ott H R and Batlogg B 1987 *Theoretical and Experimental Aspects of Valence Fluctuations and Heavy Fermions* ed. L C Gupta and S K Malik (New York: Plenum) p 57
[19] Dhar S K and Gschneidner K A Jr 1989 *J. Magn. Magn. Mater.* at press