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LETTER TO THE EDITOR

Resistivity and thermopower studies of $Ce_{3-x}La_xAl$ alloys

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Abstract. We present resistivity (ρ) and thermopower (S) measurements at temperatures between 3 K and 300 K on $Ce_{3-x}La_xAl$ alloys ($x = 0, 0.15, 0.30, 0.45, 0.6, 0.9, 1.2$). All the alloys showed evidence of Kondo-like scattering (occurrence of ρ_{min} and S_{min} around 30 K). The low-temperature S_{min} crossed over to positive values at $x \geqslant 0.45$, giving rise to a positive thermopower over the entire temperature range. The low-temperature positive peak (around 5–10 K) in S and ρ measurements gave evidence of a coherence effect. This peak was found to vary with La concentration in the thermopower studies. At higher temperatures (50–100 K), alloys with $0 \leqslant x \leqslant 0.6$ showed thermal hysteresis in both types of measurements, indicating the presence of lattice transformation effects.

Recently alloys of the type Ce_3M ($M \equiv$ Sn, In, Al) have been reported which evidenced Kondo-lattice (KL) behaviour from susceptibility and resistivity studies (Thompson *et al* 1987). Of these, Ce_3Al (with a hexagonal Ni_3Sn structure) was found to exhibit hysteresis in the ρ–T curve at zero pressure and studies with varying external pressure indicated that this could be associated with a structural transformation involving no change in crystal symmetry, analogous to the γ–α transition in pure Ce (Lawrence *et al* 1987). Lattice constant measurements indicate that a- and c-values of the hexagonal cell vary by about 0.5% across this transition (Sakurai *et al* 1988). At low temperatures, the resistivity reveals a Kondo-like behaviour ($\partial\rho/\partial T < 0$) with the occurrence of a maximum at 4.7 K, below which ρ drops abruptly. The susceptibility peak at 2.7 K indicates antiferromagnetic ordering (Lawrence *et al* 1987).

In view of the wide variety of electrical and magnetic effects, we have carried out resistivity and thermopower (TEP) studies on $Ce_{3-x}La_xAl$ ($x = 0, 0.15, 0.3, 0.45, 0.6, 0.9, 1.2$) alloys. These alloys (with $x > 0$) also crystallise in the Ni_3Sn structure as investigated by x-ray diffraction using a Siemens diffractometer. All the alloys were prepared by arc melting, under flowing argon conditions, and studied in the as-cast condition, except for the Ce_3Al alloy, which was also studied in the annealed condition.

The resistivity curves exhibit several interesting features. ρ varied linearly with T when cooled from 300 K (see figure 1). Alloys with $x \leqslant 0.6$ (curves A–E) showed a sharp rise where the thermal hysteresis was manifested whereas those with $x > 0.6$ (curves F and G) showed a smooth variation without thermal hysteresis. We note that this hysteresis was not observed in some of the previous studies of Ce_3Al, possibly due to the formation of a small amount of the Cu_3Au phase (Sera *et al* 1987, Thompson *et al* 1987). This hysteresis was found to occur at different temperatures for different values of x.

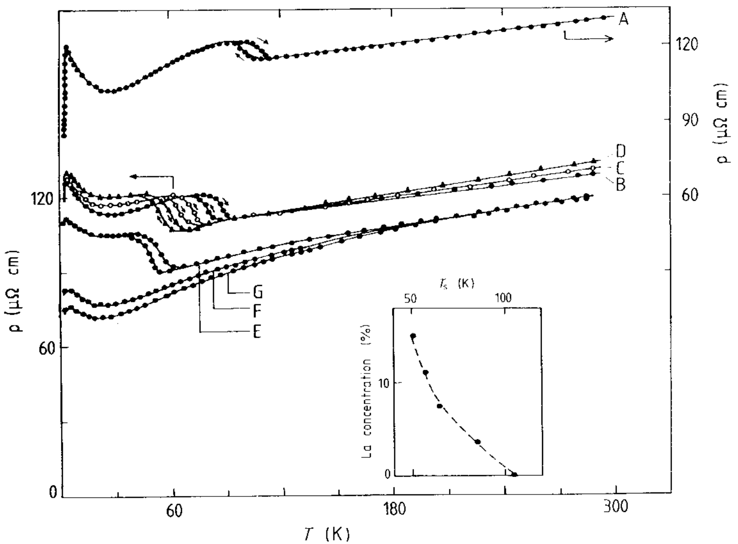

Figure 1. The variation of ρ with T for the system $Ce_{3-x}La_xAl$ from 3 K to 300 K. The curve (A) for Ce_3Al ($x = 0$) has been shifted up for clarity. A, $x = 0$; B, $x = 0.15$; C, $x = 0.3$; D, $x = 0.45$; E, $x = 0.6$; F, $x = 0.9$; G, $x = 1.2$. Inset: variation of T_s with La concentration. The curves are drawn to guide the eye.

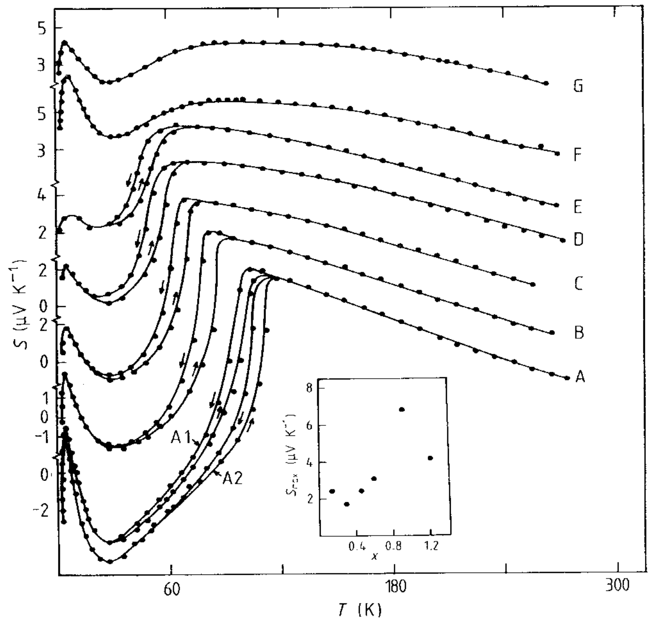

Figure 2. The variation of S with T for the system $Ce_{3-x}La_xAl$ from 3 K to 270 K. The curves are labelled as in figure 1. A1 denotes the as-cast Ce_3Al alloy and A2 the same alloy after annealing at 200 °C for 15 days (Thompson *et al* 1987). The curves are drawn to guide the eye. Inset: variation of S_{max} with x.

The range of temperature (ΔT_r) over which hysteresis occurs varied from 20 K to 35 K and the maximum width (ΔT_w) of the hysteresis loop was about 8–10 K. If we designate the midpoint of the hysteresis zone T_s, it is observed that it decreases appreciably with increase in x (see inset, figure 1). At lower temperatures ($10\ K < T < 50\ K$), all the alloys exhibit minimum resistivity at T_{min}. We find that T_{min} is almost constant at 28 K for $0 \leqslant x \leqslant 0.6$ (curves A–E) and then drops to about 22 K for higher values of x. The constancy of T_{min} over such a large substitution range of Ce by La indicates that this is related to single-ion incoherent Kondo scattering and qualitatively T_{min} may be taken as equal to T_K (the Kondo temperature) to indicate the onset of this effect (Lawrence *et al* 1987). Below T_K, $\partial\rho/\partial T$ was found to be negative, indicating the growth of these Kondo-like fluctuations.

As the temperature is lowered, all the alloys begin to show a maximum at approximately the same temperature $T_{max} \simeq 4$ K (figure 3). Below T_{max}, the drop in ρ down to 3 K is steepest in the case of Ce_3Al (curve A). This maximum signals the onset of coherence effects, taking over from single-ion Kondo behaviour. This maximum becomes broader and less pronounced and the drop in ρ less steep below T_{max} (figure 3) with increasing x, indicating that this coherence state is sensitive to La substitution of Ce ions.

We now look at the temperature variation of the thermopower S. S was found to increase almost linearly with T as the samples were cooled from 300 K (see figure 2). For $0 \leqslant x \leqslant 0.6$, S exhibited a much more pronounced thermal hysteresis than that observed in the resistivity studies (compare figures 1 and 2). For Ce_3Al (curve A), $\Delta T_r =$ 75 K in contrast to 200 K as measured from ρ–T data. However, the value of ΔT_w ($\simeq$ 10 K) is approximately the same as that observed in the resistivity behaviour. As we lower the temperature below the hysteresis zone, Ce_3Al shows a negative minimum at the same temperature T_K where ρ_{min} occurs. This behaviour is conventional to all KL systems studied. Some typical cases are $CeAl_3$ (Steglich *et al* 1985) and $CeCu_2Si_2$ (Moschalkov and Aliev 1985). At still lower temperatures ($T < T_K$), S exhibits a positive peak at T_m, close to T_{max} in the resistivity curve. However, the most interesting feature is the transition to completely positive values of S (over all temperatures) for $x \geqslant 0.45$ (curves D–G) even though the positions (T_K and T_m) of the extrema remain approximately the same. S_{min} at T_K gradually increases and becomes zero around $x = 0.45$ (curve D). S_{max} simultaneously increases from about 1.8 $\mu V\ K^{-1}$ for $x = 0$ to about 4.2 $\mu V\ K^{-1}$ for $x = 1.2$. A marked increase in S_{max} (to about 6.9 $\mu V\ K^{-1}$) was observed for the alloy with $x = 0.9$ (see inset, figure 2). Also, the position of S_{max} was found to be slightly dependent on x, varying from $T_m = 4.5$ K for Ce_3Al ($x = 0$) to about 7–8 K for higher values of x ($\sim$0.9). This again indicates that the coherent phase at low temperatures is sensitive to the amount of substitution of Ce ions by La. We note that this feature of S remaining positive (for KL systems) over the entire temperature range has up to now been observed in only two other systems, namely $CeRu_2Si_2$, having $T_K \sim 20$–30 K (Amato and Sierro 1985, Steglich *et al* 1985) and $CeCu_6$ having $T_K \sim 6$ K (Onuki *et al* 1984). We remark here that our case presents the first example where the transition from the normal TEP behaviour (negative minimum of S at T_K) to a fully positive TEP variation in T takes place as the concentration x of the substituent is varied in a single alloy series, T_K remaining approximately constant. The change of sign of S below T_K from negative to positive is a further clear indication of the occurrence of Kondo resonance at the Fermi level E_F from the coherence-derived quasi-particles. As the temperature is lowered, the resonance level can split into two levels, resulting in a 'pseudo-gap' in the density of states (DOS). The positive value of S (at $T < T_K$) could then arise when E_F intersects a

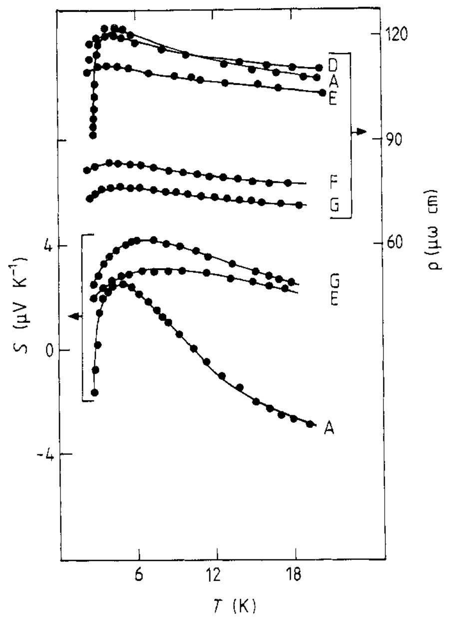

Figure 3. The expanded plot of ρ versus T and S versus T in the temperature range 2.7–20 K for selected values of x. The curves are labelled as in figure 1. Curves are drawn to guide the eye.

hole part in the DOS (Steglich *et al* 1985, Grewe 1984). Figure 3 shows the low-temperature variation of S on an expanded temperature scale. This increase of T_m with x could be related to the highly temperature-dependent DOS (Grewe 1984) and the variation of E_F with the substitution of Ce ions by La (i.e. the variation of x). Such a variation was not observed for T_{max} from our resistivity studies. It is thus noticed that the variation of S with T is more pronounced than that of ρ. This is related to the fact that while ρ is related to the DOS at E_F, S is related to its energy derivative, which is strongly temperature-dependent at low temperatures. The sensitivity of S is also manifested at higher temperatures ($10 < T < T_K$) where both Kondo scattering and coherence-derived fluctuations begin to evolve and compete. Although the positions of the extrema in the resistivity and TEP curves vary slightly, the interplay of the above two processes leads to different temperature-dependent curves for different alloys. We find that $\rho(T_{max}) - \rho(T_K)$ reaches a maximum of 23 $\mu\Omega$ cm for $x = 0$, dropping to 8 $\mu\Omega$ cm for $x > 0.6$. Correspondingly, $S(T_m) - S(T_K)$ is about 7 $\mu V\ K^{-1}$ for Ce_3Al ($x = 0$), dropping to about 1 $\mu V\ K^{-1}$ for $x = 0.6$ (see figures 1 and 2).

TEP measurements were found to be sensitive to annealing as revealed from our as-cast and annealed Ce_3Al system (curves A1 and A2 in figure 2, respectively). The values of S at both low-temperature extrema are found to be sensitive to heat treatment as shown in figure 2. Also dramatic is the large value of $\Delta T_r = 75$ K for both Ce_3Al samples, compared to the ρ data, for which $\Delta T_r = 10$ K. However, $\Delta T_w \sim 10$ K as observed in ρ studies. We note that the large value of ΔT_r reflects the fact that the electron states are very different (where the lattice transformation takes place at about T_s), the effect on the TEP being more pronounced than that on ρ.

To summarise, we find that our resistivity and thermopower studies reveal clearly the presence of Kondo-like scattering over the entire alloy series $Ce_{3-x}La_xAl$, as evidenced through the occurrence of ρ_{min} and S_{min}. The single-ion character of this scattering process is revealed in the relative insensitivity of this extremum point to variations in x.

Secondly, the occurrence of ρ_{max} and S_{max} arises from coherence effects of quasi-particles. The low temperature peak in S is sensitive to variations in x. However, it is not possible from our electrical resistivity studies alone to indicate the exact nature of magnetic interactions in the coherent regime. In the temperature interval $10 < T < T_K$ the interaction of single-ion Kondo scattering and the growth of the coherent state is shown to vary considerably with temperature, from both ρ and S curves. At still higher temperatures (near T_s), resistivity and TEP studies reveal giant hysteresis loops, indicating the presence of lattice transformations.

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