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Transition from magnetic ordering to Kondo behavior in $Ce_{3-x}La_xAl_{11}$

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Abstract

Ce_3Al_{11} shows coexistence between magnetic ordering and Kondo behavior. Substituting La for Ce reduces the magnetic ordering temperature and simultaneously the entropy associated with the magnetic order. This results in an enhancement of the low temperature specific heat coefficient γ. Samples with small Ce concentrations, where no magnetic order is found, show a single ion Kondo effect, with C/T rising down to the lowest measured temperatures.

Keywords: Ce compounds; Kondo alloys; Magnetic measurements; Specific heat measurements

1. Introduction

For heavy fermion systems the competition between RKKY interaction and the screening of the magnetic moments by the Kondo effect plays an important role. Substitution experiments with Ce_3Al_{11} were carried out to determine what parameters are important to achieve a heavy fermion groundstate. Ce_3Al_{11} crystallizes in the orthorhombic *oI*28 La_3Al_{11} structure and shows two magnetic transitions at low temperatures: a ferromagnetic one at 6.2 K and an antiferromagnetic one at 3.2 K. These transitions are associated with the two different Ce sites occurring in this structure, with rather different magnetic moments ($m_{Ce_I} = 1.11\ \mu_B$ and $m_{Ce_{II}} = 0.26\mu_B$) [1,2], coexisting with a Kondo type behavior with increased Sommerfeld coefficient ($\gamma = 370\ mJ\ mol^{-1}\ K^{-2}$ in the ferromagnetic HT-phase and $\gamma = 120\ mJ\ mol^{-1}\ K^{-2}$ in the antiferromagnetic LT-phase) [3]. For a recent single crystal study on the rather complex magnetic behavior of Ce_3Al_{11} see Ref. [4]. Since La_3Al_{11} shows no magnetic behavior, La is an ideal dopant. For this reason it was interesting to study the transition from this complex magnetic behavior to a non-magnetic groundstate. The question was, if magnetism is suppressed by substituting La for Ce would this affect the competition between the Kondo and the RKKY interaction and give rise to an enhanced low temperature specific heat γ? Another aim of this work was to show how the two different magnetic transitions of Ce_3Al_{11} are influenced by substituting La for Ce.

2. Experimental

The samples of $Ce_{3-x}La_xAl_{11}$ ($x = 0$, 0.5, 1, 1.5, 2, 2.8, 2.9, 2.95, 2.98) were prepared by melting together stoichiometric amounts of the constituents in an arc-melter in purified Ar atmosphere. The samples were remelted three times to improve the homogeneity. In all the samples no further second phase peaks were visible in the X-ray diffraction patterns, especially no second phases of $CeAl_2$ and $CeAl_3$ could be determined (that would affect the low temperature specific heat dramatically). Susceptibility measurements between 1.8 and 400 K were carried out in a Quantum Design 7T magnetometer, while specific heat measurements between 350 mK and 20 K were made with a thermal relaxation technique as described in the literature [5,6].

3. Results and discussion

The specific heat data of $Ce_{3-x}La_xAl_{11}$ for $x = 0$, 1 and 2 are shown in Fig. 1. For the samples with

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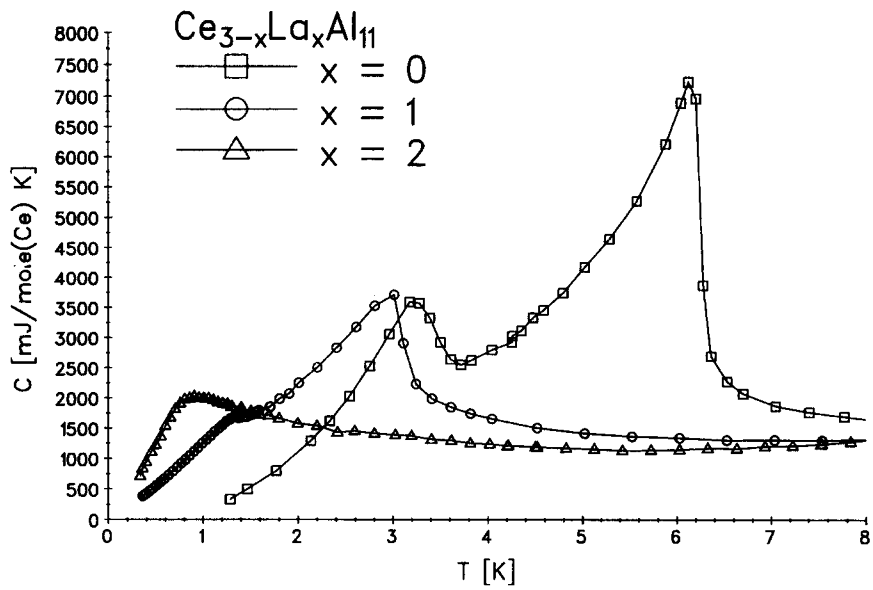

Fig. 1. Specific heat C vs. T for $Ce_{3-x}La_xAl_{11}$ for $x = 0$, 1, 2. Solid lines are a guide to the eye.

$x = 0.5$, 1.5 no specific heat was measured. For $x = 0$ the peak at 6.2 K (ferromagnetic transition) is sharp, while the peak at 3.2 K (antiferromagnetic) is somewhat broad. For $x = 1$ the ferromagnetic transition is shifted down to about 3 K, at 1.5 K a small anomaly in the specific heat is visible. For $x = 2$ the ferromagnetic peak at about 800 mK is quite broad.

The low temperature specific heat divided by temperature for $x = 1$ and $x = 2$ is plotted, expanded, in Fig. 2. For $x = 1$ a broad peak around 1.5 K is visible that may be due to antiferromagnetic order. The ferromagnetic T_C and the antiferromagnetic T_N (for $x = 0, 1$) ordering temperatures are decreased with increasing La concentration. The dependence of the ordering temperatures T_C and T_N vs. the Ce concentrations is plotted in the inset to Fig. 2. The T_C for $x = 0.5$ and 1.5 are extrapolated from the inverse susceptibility (not shown). This figure implies that the ferromagnetic order should be suppressed at a finite Ce concentration. That the order for $x = 1$ and $x = 1.5$ is ferromagnetic was checked by measuring hysteresis effects in the magnetization (not shown), that gave clear indications of ferromagnetism. Also, magnetization measurements at $T = 2$ K showed spontaneous

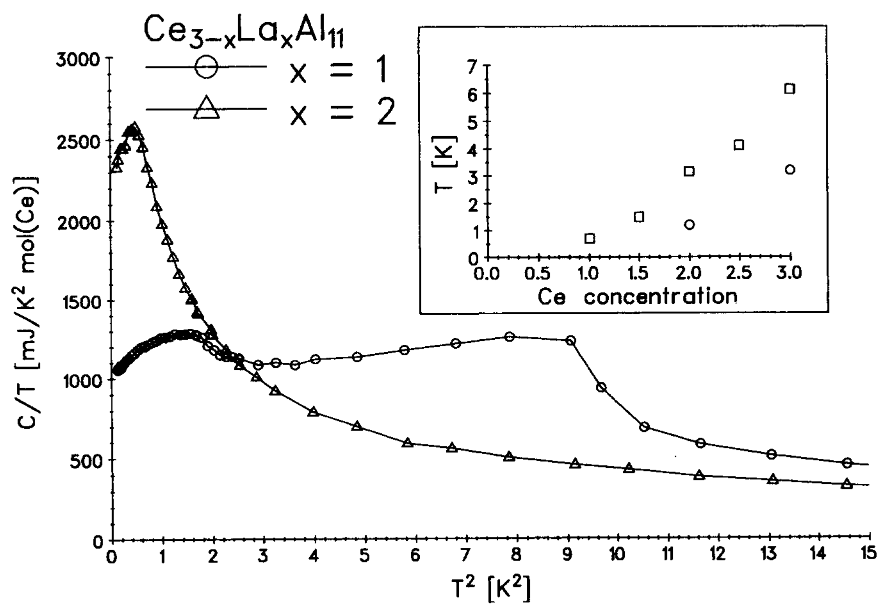

Fig. 2. Low temperature specific heat C/T vs. T^2 for $x = 1$, 2. Solid lines are a guide to the eye. The inset shows the ferromagnetic (□) and antiferromagnetic (○) ordering temperatures vs. Ce concentration. For $x = 0.5$, 1.5 the ordering temperatures are from an extrapolation of the inverse susceptibility, because the specific heat was not measured. Therefore, T_N was not determined for these samples.

magnetization for $x \leq 1.5$ (not shown). The extrapolated γ (defined as $\gamma = C/T(T \rightarrow 0)$) in Fig. 2 amounts to about 1000 mJ mol^{-1} (Ce) K^{-2} for Ce_2LaAl_{11} (for the LT- and HT-phases) and 2200 mJ mol^{-1} (Ce) K^{-2} for $CeLa_2Al_{11}$ within some error bars due to the uncertainty of the extrapolation.

Another interesting feature is contained in Fig. 3 where the entropy for $x = 0, 1, 2$ is shown after subtracting the lattice contribution to the specific heat (the specific heat of La_3Al_{11} was fitted to the following equation: $C = 12.053 + 0.295T^3 + 2.6 \times 10^{-3}T^5 - 2.6 \times 10^{-6}T^7$). The entropy for every sample per mole Ce is nearly the same at $T = 12$ K. Reducing the entropy associated with the magnetic ordering (indicated by the arrows) by substituting La for Ce enhances the entropy of the Kondo interaction. This results in an enhanced low temperature γ shown in Fig. 2. Similar results have been obtained in the dense Kondo systems $Ce_{1-x}La_xGe_2$ [7] and $Ce_{1-x}La_xAl_2$ [8]. For a further check of our results we measured samples with low Ce concentration ($x = 2.8, 2.9, 2.95, 2.98$), where the magnetic order should be even more fully suppressed (see inset to Fig. 2). The results of the susceptibility and specific heat measurements are shown in Figs. 4 and 5 respectively. In both cases the La_3Al_{11} contributions are subtracted, so that only contributions of the electronic Ce part is shown. From these figures it is obvious that the enhanced low

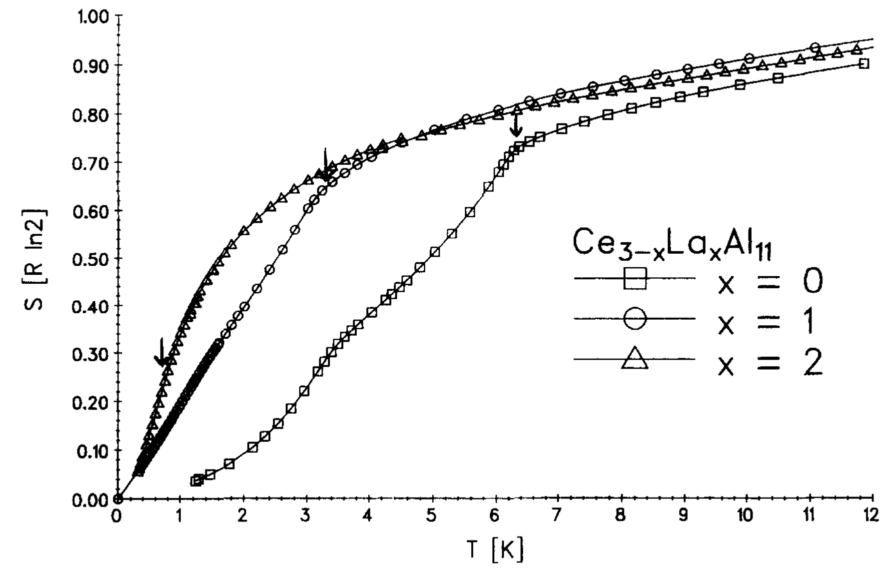

Fig. 3. Entropy S vs. T of the electronic part of $Ce_{3-x}La_xAl_{11}$ for $x = 0, 1, 2$. Solid lines are a guide to the eye.

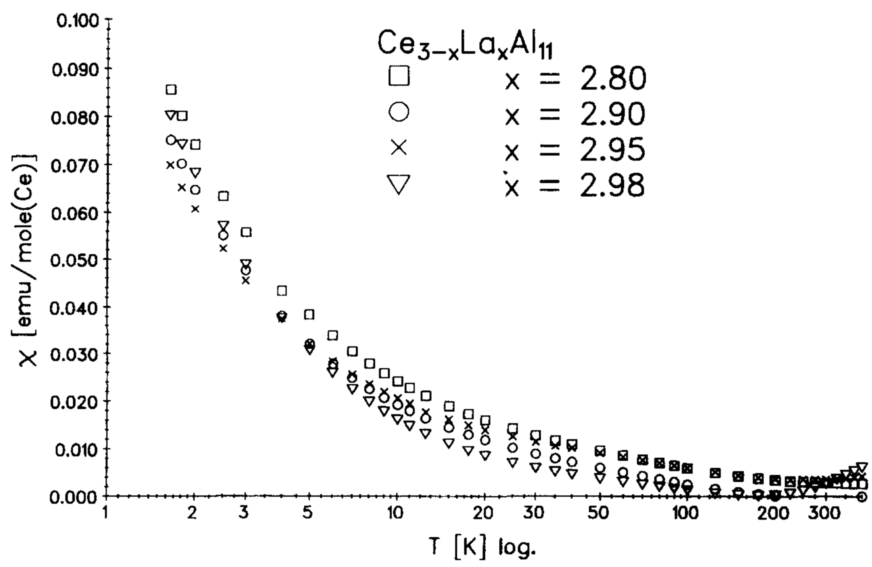

Fig. 4. Susceptibility χ vs. T of $Ce_{3-x}La_xAl_{11}$ for $x = 2.8, 2.9, 2.95, 2.98$. The diamagnetic contribution of La_3Al_{11} was subtracted. Note the logarithmic scale of the x-axis.

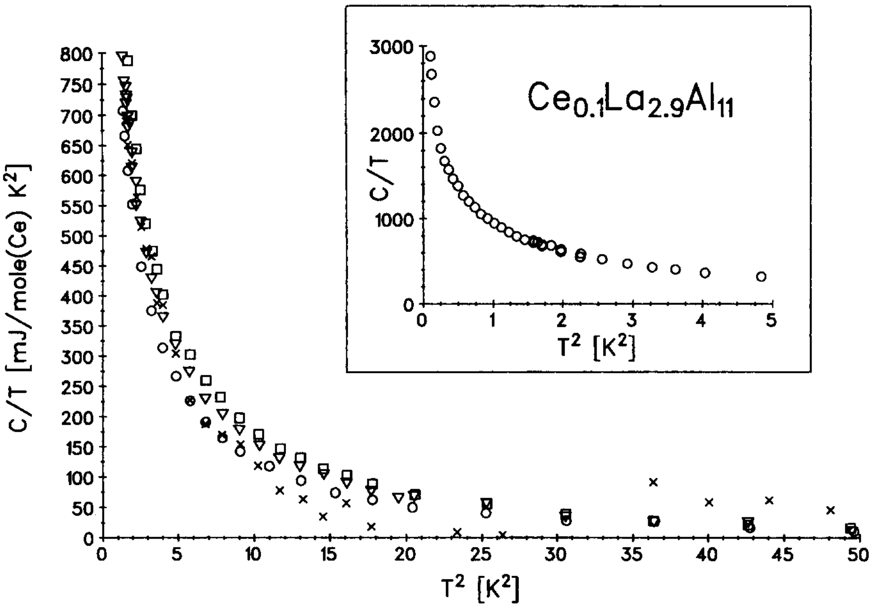

Fig. 5. Specific heat C/T vs. T^2 of $Ce_{3-x}La_xAl_{11}$ for $x = 2.8, 2.9, 2.95, 2.98$ (for symbols see Fig. 4) down to 1.3 K. The contribution of La_3Al_{11} was subtracted. The inset shows the low temperature specific heat down to 0.35 K C/T vs. T^2 for $Ce_{0.1}La_{2.9}Al_{11}$.

temperature specific heat and the susceptibility are constant per mole Ce over one decade of Ce concentration. This may be taken as a strong indication of a single ion Kondo effect. Magnetization measurements at $T = 2$ K (not shown) show a saturation behavior, indicating that the magnetic moments are still not fully screened by the Kondo interaction at this temperature. The rather enhanced γ of $Ce_{0.1}La_{2.9}Al_{11}$ plotted in the inset to Fig. 5 is still rising down to the lowest temperatures ($T = 350$ mK) measured.

4. Conclusion

By substituting La for Ce in $Ce_{3-x}La_xAl_{11}$ we were able to show that the electronic part of the entropy could be shifted from magnetic order to a single ion Kondo interaction, without loss of entropy. This results in an enhanced Sommerfeld coefficient γ.

Acknowledgements

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