Specific Heat of $Ce_{0.8}La_{0.2}Al_3$ in Magnetic Fields: A Test of the Anisotropic Kondo Picture

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The specific heat C of $Ce_{0.8}La_{0.2}Al_3$ has been measured as a function of temperature T in magnetic fields up to 14 T. A large peak in C at 2.3 K has recently been ascribed to an anisotropic Kondo effect in this compound. A 14-T field depresses the temperature of the peak by only 0.2 K, but strongly reduces its height. The corresponding peak in C/T shifts from 2.1 K at zero field to 1.7 K at 14 T. The extrapolated specific heat coefficient $\gamma = \lim_{T\to 0} C/T$ increases with field over the range studied. We show that these trends are inconsistent with the anisotropic Kondo model.

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$CeAl_3$ occupies an important position in the history of correlated electron systems. The first report of its unusual low-temperature specific heat [1] sparked enormous interest in materials containing $4f$ or $5f$ elements. For years, $CeAl_3$ was considered the prototypical nonmagnetic heavy fermion system [2], in which Kondo screening of f moments prevails over magnetic ordering to produce a heavy Fermi-liquid ground state. The low-temperature specific heat $C(T)$ is 3 orders of magnitude greater than that of a conventional metal, with a linear coefficient $\gamma = \lim_{T\to 0} C/T \approx 1250$ mJ/K^2 Ce mol [3]. The resistivity is described very accurately above 10 K by a theory for a Kondo impurity in crystalline electric fields [4], and below 300 mK by the Fermi-liquid form $\rho = \rho_0 + AT^2$ with a strongly enhanced A coefficient. The ratio A/γ^2 is about $10^{-5}\ \Omega\,\mathrm{cm\,K^2\,mol^2\,J^{-2}}$, a value close to that theoretically predicted for nonmagnetic Kondo lattices [5], and observed [6] in many other heavy fermion compounds.

The picture of $CeAl_3$ as a nonmagnetic Kondo lattice has been challenged by muon spin rotation [7] and nuclear magnetic resonance [8] measurements, which indicate the presence of short-range magnetic correlations below 2 K. In addition, the specific heat has an unexplained feature: a maximum in C/T near 0.4 K. A similar maximum is found around the same temperature in another heavy fermion system, $CeCu_2Si_2$. In both compounds, this feature was initially attributed to coherence in the Kondo lattice [9]. However, extensive studies of $CeCu_2Si_2$ gave rise to an alternative explanation based on weak magnetic ordering of heavy quasiparticles [10].

An alloying study of $CeAl_3$ has similarly pointed to a magnetic origin for the 0.4 K anomaly [11]. When La is partially substituted for Ce in $Ce_{1-x}La_xAl_3$, this weak feature, observable in C/T but not in C for $x = 0$, grows for $x \geq 0.05$ into a large peak in both C and C/T. The highest-La-content alloy investigated in [11], $Ce_{0.8}La_{0.2}Al_3$, has a pronounced maximum in C near 2.3 K and a corresponding peak in the susceptibility at 2.5 K, reminiscent of an antiferromagnetic transition. The smooth evolution with x suggests that the anomaly has a common origin in the pure and La-doped compounds, and hence that alloying can provide insight into the physics of $CeAl_3$, with the advantages that the features of interest are stronger and are located at higher temperatures. (Since La expands the lattice, presumably decreasing hybridization between $4f$ and ligand states, the apparent increase of magnetic character with doping is consistent with the Kondo necklace model of heavy fermions [12]. However, an interpretation based on this simple model is somewhat undermined by the fact that anomalies in C, similarly pronounced to those produced by La substitution, can be induced by replacing Al atoms with either larger or smaller atoms [13].)

A recent study of the spin dynamics of $Ce_{0.8}La_{0.2}Al_3$ [14] revealed a divergence in the muon spin relaxation (μSR) rate below 3 K, the temperature around which upturns develop in thermodynamic properties. This divergence was attributed to the development of (quasi)static magnetic correlations. Since neutron diffraction showed no evidence for magnetic Bragg peaks, and placed an upper limit of $0.05\mu_B$ on any ordered moment, the μSR data may point either to short-range magnetic order or to long-range ordering of very small moments, as seen in other heavy fermion systems [15]. A change in the neutron scattering from quasielastic to inelastic below 3 K was ascribed not to magnetic ordering but rather to the onset of weakly dissipative local dynamics, consistent with a description of $Ce_{0.8}La_{0.2}Al_3$ and $CeAl_3$ in terms of the anisotropic Kondo model (AKM) [16].

That anisotropy plays a crucial role in $CeAl_3$ is a novel and intriguing idea which merits further investigation. Here we report the effects of magnetic fields up to 14 T on the specific heat of $Ce_{0.8}La_{0.2}Al_3$. We focus on the field dependence of the linear coefficient γ and the temperature positions T_M and T_m of the maxima in C and C/T, respectively. The experimental data show qualitatively different trends from numerical results calculated for the AKM, thereby casting considerable doubt on the validity of this theoretical description of $Ce_{0.8}La_{0.2}Al_3$ and its parent compound, $CeAl_3$. The field dependences leave open the possibility of magnetic ordering, probably short range in character.

Our measurements used a polycrystal of $Ce_{0.8}La_{0.2}Al_3$ from a previous alloying study [11]. The sample was

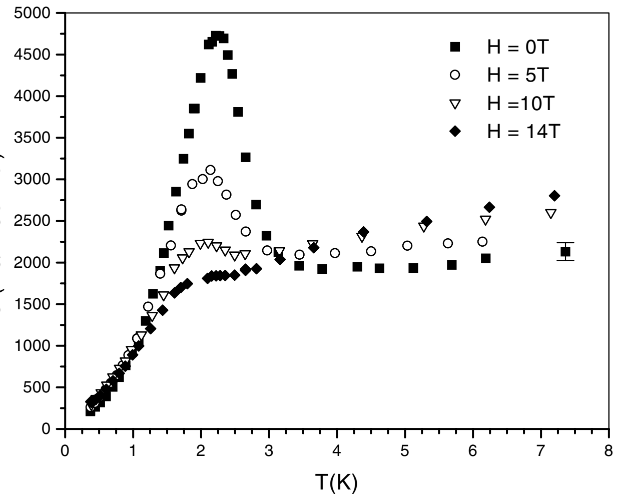

FIG. 1. C vs T for $Ce_{0.8}La_{0.2}Al_3$ at $H = 0$, 5, 10, and 14 T. The experimental uncertainty is shown for a representative zero-field point.

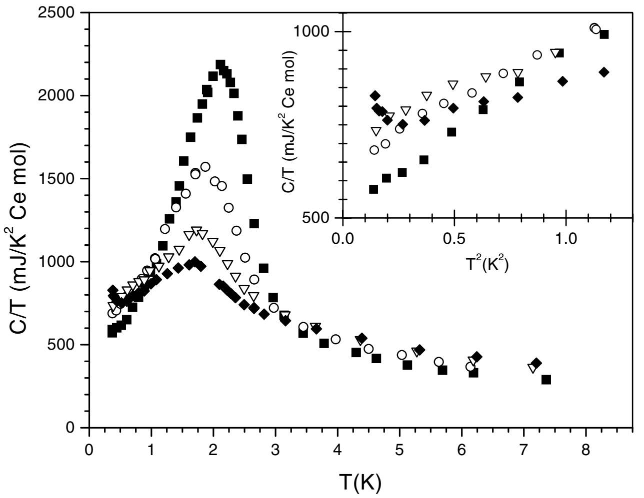

FIG. 2. C/T vs T for $Ce_{0.8}La_{0.2}Al_3$ at $H = 0$, 5, 10, and 14 T. Inset: C/T vs T^2 for $T < 1.1$ K. See the legend of Fig. 1 for an explanation of the symbols.

prepared in an arc furnace under an argon atmosphere, and annealed at 830 °C for three weeks. Magnetic susceptibility and x-ray diffraction measurements revealed no sign of the secondary phases $CeAl_2$ and Ce_3Al_{11}.

Figure 1 shows the specific heat of this alloy in fields $H = 0$, 5, 10, and 14 T. The phonon contribution has been subtracted using the specific heat of $LaAl_3$ [17], and the remainder has been normalized to a mole of Ce. The experimental uncertainty is about 5%. The same data are plotted as C/T vs T in Fig. 2. The main effect of the field is a strong reduction in the magnitude of the anomalies in C and C/T. Also striking is the very weak field dependence of the temperature positions of the anomalies. A pronounced peak in C at $T_M \simeq 2.3$ K for $H = 0$ is replaced by a shoulder near 2.1 K for $H = 14$ T. Likewise, the peak in C/T at T_m shifts slowly with field, from 2.1 K at 0 T to 1.7 K at 14 T. For later reference, we note that the variation of T_m is sublinear in H (see Fig. 3). The difference between T_M and T_m grows with the applied field. A difference of the same order has been observed in zero field for $Ce_{1-x}La_xAl_3$ alloys with $x < 0.2$, where $T_M - T_m$ grows as x becomes smaller [11]. In this respect, an increase in H has a similar effect to a decrease in x.

We also note an increase with field of C/T values at low temperatures (see inset of Fig. 2), signaling a partial restoration of the heavy fermion state present in pure $CeAl_3$. It may be that the large nuclear moments of Al contribute to the enhancement of C/T at the lowest temperatures and the largest fields. Indeed, the 14-T C/T data display a low-temperature tail which might be due to a nuclear hyperfine contribution $\Delta C/T \propto 1/T^3$. None of the curves at lower fields shows a similar upturn. For $H = 0$, 5, and 10 T, the linear specific heat coefficient γ was extracted from C/T vs T^2 curves at the lowest temperatures, even though C/T is not strictly proportional to T^2, particularly for $H \geq 10$ T. For $H = 14$ T, γ was determined from the slope of CT^2 vs T^3 below 1 K. These γ values are shown in the lower panel of Fig. 3, together with error bars which combine experimental and regression uncertainties.

We now attempt to analyze our data in terms of the anisotropic Kondo model for a single magnetic impurity. The model assumes an exchange interaction $J_z S_z s_z + J_\perp (S_x s_x + S_y s_y)$ between the impurity spin $\mathbf{S}$ and the net conduction-electron spin $\mathbf{s}$ at the impurity site. Goremychkin *et al.* [14] proposed the AKM as a description of both $Ce_{0.8}La_{0.2}Al_3$ and $CeAl_3$. The Ising-like crystal-field ground state [18] and the anisotropic susceptibility [19] of $CeAl_3$ point to highly anisotropic exchange constants $J_z \gg J_\perp > 0$, with the magnetic z direction being the crystallographic c axis.

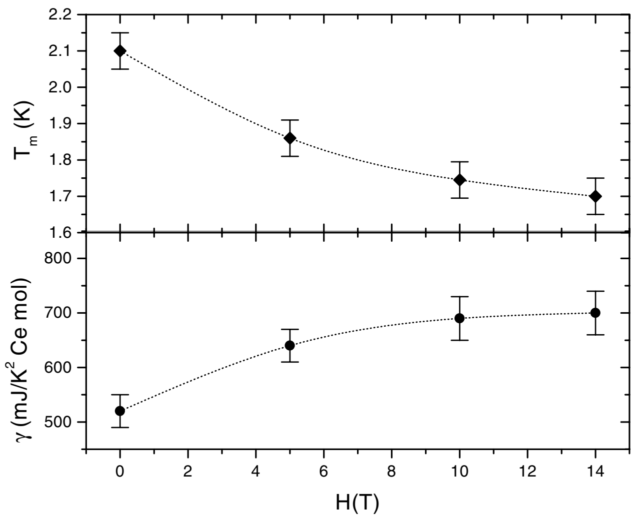

FIG. 3. T_m vs H and γ vs H for $Ce_{0.8}La_{0.2}Al_3$, where T_m is the temperature of the maximum in C/T. The dotted lines are guides to the eye.

The AKM is known to be equivalent in the limit of low energies to a number of other models. In recent years a mapping [20] onto the spin-boson model with Ohmic dissipation has been exploited [16,21] to deduce physical properties of dissipative two-level systems [22] from numerical calculations performed for the AKM. These studies have shown that, under certain conditions, the impurity contribution to the zero-field heat capacity of the AKM exhibits peaks both in C and C/T, qualitatively similar to the data plotted in Figs. 1 and 2. Such peaks are found only for $\varrho_0 J_z \gtrsim 1$, where ϱ_0 is the conduction-band density of states at the Fermi energy. For a given value of $\varrho_0 J_z$, the specific heat for all $J_\perp \ll J_z$ can be collapsed onto a universal scaling curve $C/\gamma T$ vs T/T_m [21]. Moreover, the temperature of the peak in C/T is given by $T_m = \alpha^* R/\gamma$ [3], where R is the gas constant and α^* is a function of $\varrho_0 J_z$ only [23].

From the observed peak position $T_m = 2.1$ K and a value $\gamma = 520$ J/K^2 Ce mol extracted as described above, we deduce $\alpha^* = \gamma T_m/R = 0.13$ for $\mathrm{Ce}_{0.8}\mathrm{La}_{0.2}\mathrm{Al}_3$ in zero field, in agreement [23] with the estimate of [14]. We have used α^* and T_m as inputs for a numerical renormalization-group calculation [24] of the specific heat of the AKM. Figure 4 shows the effect of applying a uniform magnetic field along the magnetic z axis, under the assumption that the impurity and the conduction electrons have g factors $g_i = g_e = 2$. (Changing the g factors multiplies the field scale by an overall factor, but does not otherwise affect the results [25].)

The numerical data exhibit three main trends with increasing magnetic field H: (1) The anomaly in C/T becomes broader and lower. (2) The peak shifts markedly to higher temperatures. (3) C/T decreases at all temperatures below the zero-field value of T_m; the fractional change in γ is greater than that in the peak height, so that $\alpha^*(H) = \gamma(H)T_m(H)/R$ decreases monotonically with increasing H, as shown in the legend of Fig. 4.

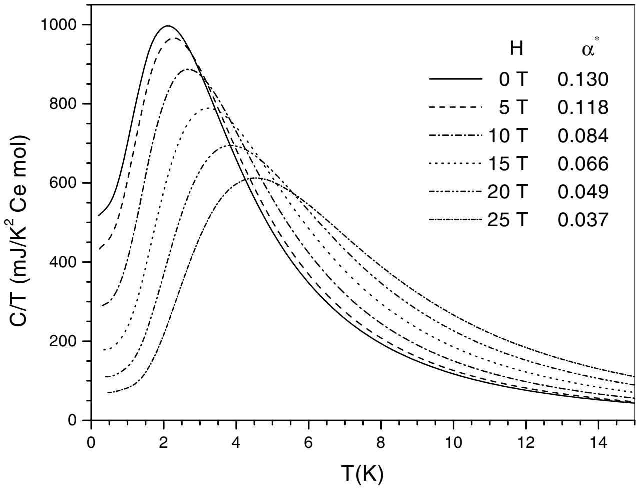

FIG. 4. C/T vs T calculated for the anisotropic Kondo model in various magnetic fields H, with model parameters chosen so that $\alpha^* = 0.130$ for $H = 0$. See the text for details.

The results presented in Fig. 4 are directly applicable only to single-crystal $\mathrm{Ce}_{0.8}\mathrm{La}_{0.2}\mathrm{Al}_3$ with a magnetic field along the c axis. For comparison with our polycrystalline data, one must average over all possible field orientations. The Ising-like crystal-field ground state of Ce^{3+} in CeAl_3 [18] implies that $g_i = 0$ for the basal-plane components of the magnetic field and, hence, that the specific heat of a polycrystal in field H is an equally weighted average of the single-crystal results for all fields between zero and H. This averaging process preserves trends (1)–(3) above.

Trend (1) accords well with our measurements, but (2) and (3) both run counter to experiment. In $\mathrm{Ce}_{0.8}\mathrm{La}_{0.2}\mathrm{Al}_3$, T_m does not rise with increasing field, but instead is weakly depressed, while C/T undergoes a small increase at temperatures much below T_m. In particular, γ rises sufficiently fast that α^* remains essentially constant up to a 14-T field (see Table I), in contrast to the prediction of the AKM (Fig. 4).

Similar discrepancies between experiment and the AKM can be seen in other members of the $\mathrm{Ce}_{1-x}\mathrm{La}_x\mathrm{Al}_3$ series. Preliminary measurements on samples at higher La doping (up to $x = 0.7$) confirm a decrease of T_m with increasing field. It should also be noted that specific heat measurements in fields of 0, 2, and 4 T have been reported for pure CeAl_3 [9]. It is harder to identify unambiguous trends in this compound because of the weakness of the anomaly in C/T and the low temperature at which it occurs. However, it appears that γ rises with increasing H while T_m is depressed, consistent with the tendencies seen more clearly in $\mathrm{Ce}_{0.8}\mathrm{La}_{0.2}\mathrm{Al}_3$.

The preceding comparisons point to significant shortcomings of the AKM as a description of pure and La-doped CeAl_3, perhaps due to its neglect of the magnetic correlations identified in [14]. As noted at the beginning of this paper, these correlations could signify long-range order of very small moments. To examine the implications of our results for this alternative interpretation, it is useful to review the field dependences of heavy fermion systems that are known to order antiferromagnetically. $\mathrm{URu}_2\mathrm{Si}_2$, for example, has an ordered moment of $(0.04 \pm 0.01)\mu_B$ per U atom [15]. In an applied field, the Néel temperature T_N is depressed quadratically in H, while the associated peak in C/T becomes slightly sharper and higher [26]. In CePb_3, which has a larger ordered moment of $(0.55 \pm 0.1)\mu_B$

TABLE I. Values of the specific heat coefficient γ, the peak temperature T_m, and $\alpha^* = \gamma T_m/R$ (where R is the gas constant) for $\mathrm{Ce}_{0.8}\mathrm{La}_{0.2}\mathrm{Al}_3$ in different magnetic fields H.

H (T)	γ (mJ/K^2 Ce mol)	T_m (K)	α^*
0	520 ± 30	2.13 ± 0.05	0.133 ± 0.008
5	640 ± 30	1.86 ± 0.05	0.143 ± 0.008
10	690 ± 40	1.75 ± 0.05	0.145 ± 0.009
14	700 ± 40	1.70 ± 0.05	0.143 ± 0.009

[27], $T_N(0) - T_N(H) \propto H^{\nu}$ for small fields, with $\nu > 2$ [28]. The superlinear field dependences of T_N and the absence of peak broadening in URu_2Si_2 contrast with our findings for $Ce_{0.8}La_{0.2}Al_3$, and weigh somewhat against a small-moment, long-range-ordering scenario for the latter material.

Another explanation of the results reported in [14] might be that the magnetic correlations inferred from μSR are sufficiently short ranged that they cannot be detected via neutron diffraction. Additional μSR measurements are planned to test this possibility.

In summary, we have measured the heat capacity of $Ce_{0.8}La_{0.2}Al_3$ as a function of temperature in magnetic fields up to 14 T. The field strongly diminishes the peaks found around 2 K in both C and C/T, but only weakly depresses the peak temperatures. The linear specific heat coefficient increases with field in the direction of the value for pure $CeAl_3$. We have analyzed our data in terms of the anisotropic Kondo model. The model predicts a shift of the peak in C/T to higher temperatures with increasing field, accompanied by a significant reduction in C/T at low temperatures. These two trends are at odds with experiment. Details of the field response do not fit patterns seen in other heavy fermion systems that exhibit small-moment magnetism, but our data do not rule out a theoretical picture based on short-range magnetic order.

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[25] A magnetic field H enters the partition function of the AKM in the combination $[1 - (2g_e/\pi g_i) \times \tan^{-1}(\pi\rho_0 J_z/4)]g_i \mu_B H$; see P.B. Vigmann and A.M. Finkel'stein, Zh. Eksp. Teor. Fiz. **75**, 204 (1978) [Sov. Phys. JETP **48**, 102 (1978)]. For fixed $\rho_0 J_z$, any change in the g factors can be treated as an effective rescaling of H. In fact, the curves in Fig. 4 were calculated for $g_i = 2$ and $g_e = 0$; then the field scale was converted to that for $g_i = g_e = 2$.

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