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 a particularly important position in the history of heavy fermions. The first report of its unusual low-temperature specific heat [4] a quarter of a century ago led to enormous interest in this and similar systems based on $4f$ or $5f$ elements. For years, CeAl$_3$ was considered a canonical heavy fermion system, and it greatly influenced theoretical work in the field. Indeed, CeAl$_3$ was viewed as a realization of a “standard model” based on the Kondo effect and Fermi-liquid theory. The hallmark properties of this compound are its specific heat and its electrical resistivity. The low-temperature specific heat $C(T)$ is greatly enhanced over that for a conventional metal, with a linear coefficient $\gamma = \lim_{T \to 0} C/T \approx 1250 \text{ mJ/K}^2\text{Ce mol}$. The resistivity above 10 K is described to very high very accuracy by a theory for a Kondo impurity in crystalline electric fields [3]. Below 300 mK the resistivity has a Fermi-liquid form $\rho = \rho_0 + AT^2$, with a strongly enhanced $A$ coefficient. The ratio $A/\gamma^2$ is about $10^{-5}\Omega\text{ cm K}^{-2}\text{ mol}^{-2}\text{ J}^{-2}$, a value close to that theoretically predicted for nonmagnetic Kondo lattices [4], and experimentally observed [4] in many other heavy fermion compounds.

The description of CeAl$_3$ in terms of a nonmagnetic Kondo lattice and a heavy Fermi liquid has been challenged by microscopic measurements such as muon spin resonance ($\mu$SR) [3] and nuclear magnetic resonance (NMR) [7]. According to these measurements, either short-range magnetic order or strong antiferromagnetic correlations exist below 2 K. In addition, the specific heat itself has an unexplained feature: a maximum in $C/T$ near 0.4 K. A similar maximum is found in another heavy fermion system, CeCu$_2$Si$_2$, around the same temperature. In both compounds, this feature was initially attributed to coherence in the Kondo lattice. However, extensive studies of CeCu$_2$Si$_2$ gave rise to an alternative explanation based on weak magnetic ordering of heavy quasiparticles [8]. A previous alloying study of CeAl$_3$ has similarly pointed to a magnetic origin for the 0.4 K anomaly [4]. When La is partially substituted for Ce in Ce$_{1-x}$La$_x$Al$_3$, this weak feature, observable in $C/T$ but not in $C$ for $x = 0$, gradually evolves for $x \geq 0.05$ into a large peak in both $C$ and $C/T$. The highest-La-content alloy investigated in Ref. 8, 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 and monotonic increase with $x$ of the temperature position and magnitude of the anomaly suggests that this feature has a common origin in pure and La-doped CeAl$_3$. The apparent enhancement of the magnetic character of the anomaly upon La doping is consistent with Doniach’s Kondo necklace model [4], since doping increases the lattice constants, and therefore decreases the hybridization between $f$ and ligand states. However, an interpretation based on this 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 [4].

Recent neutron scattering and $\mu$SR studies by Goremychkin et al. [12] on Ce$_{0.8}$La$_{0.2}$Al$_3$ revealed the absence of magnetic Bragg peaks, and estimated the upper limit of any possible ordered moment to be 0.05$\mu_B$. The response function deduced from time-of-flight measurements changes from a quasi-elastic form to an inelastic form around 3 K, the temperature range where features develop in the specific heat and the magnetic susceptibility. This result was attributed to weakly dissipative dynamics consistent with the anisotropic Kondo model (AKM) [13]. $\mu$SR spectra showed Lorentzian damping, with a temperature-dependent damping rate that diverges also around 3 K. The divergence was attributed to the development of static magnetic correlations, indicating the possibility of magnetic order of small moments, as seen in other heavy fermion systems [4].

In order to investigate further the applicability of the AKM to Ce$_{0.8}$La$_{0.2}$Al$_3$, and to search for any contribu-
of the anomalies in $C$ and $C/T$. Also striking is the very weak field dependence of the temperature position of the anomalies. A pronounced peak in $C$ located at $T_M \approx 2.3\,\text{K}$ for $H = 0$ is replaced by a shoulder near $2.1\,\text{K}$ for $H = 14\,\text{T}$. The peak in $C/T$ also shifts slowly with field, $T_m$ decreasing from $2.1\,\text{K}$ at $0\,\text{T}$ to $1.7\,\text{K}$ at $14\,\text{T}$ (see Fig. 4). Note that the difference between $T_M$ and $T_m$ grows with applied field. A difference of the same order has been observed in zero field for Ce$_{1-x}$La$_x$Al$_3$ alloys with $x < 0.2$, where $T_{m} - T_{m}$ grows as $x$ becomes smaller [10]. In this respect, an increase in the magnetic field has a similar effect to a decrease in $x$.

Another important result is an increase with field of $C/T$ values at low temperatures (below $1\,\text{K}$), 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 enhance $C/T$ at the lowest temperatures and the largest fields. Indeed, the 14-tesla $C/T$ 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 show a similar upturn. Therefore, the linear specific heat coefficient $\gamma$ was extracted from a linear fit to $C/T$ vs $T^2$ below $1\,\text{K}$, except for the $14\,\text{T}$ data, where $\gamma$ was determined from the slope of $CT^2$ vs $T^3$ below $1\,\text{K}$. As may be seen in Fig. 3, $\gamma$ seems to saturate in the range $H \lesssim 10\,\text{T}$. (The error bars for $\gamma$ combine experimental and regression uncertainties.)

It is worth noting that $C/T$ for Ce$_{0.8}$La$_{0.2}$Al$_3$ at $14\,\text{T}$ and $C/T$ for CeAl$_3$ in zero field coincide above $4\,\text{K}$ to within the accuracy of the measurement. This is demonstrated in Fig. 4, which also includes the corresponding curve for Ce$_{0.8}$La$_{0.2}$Al$_3$ at $H = 0$. Since $C/T$ for the pure compound is only weakly field dependent above $4\,\text{K}$ (for fields $\sim 10\,\text{T}$) [11], we can claim that the high-field ($H \sim 14\,\text{T}$) specific heats for these two alloys converge in this temperature regime.
We now attempt to analyze our magnetic-field data in terms of the anisotropic Kondo model (AKM) for a single magnetic impurity. The model assumes an exchange interaction $J_z S_z s_z + J_\perp (S_z s_z + S_y s_y)$ between the impurity spin $S$ and the net conduction-electron spin $s$ at the impurity site. Goremychkin et al. [12] have proposed the AKM as a description for the thermodynamic properties of both Ce$_{0.8}$La$_{0.2}$Al$_3$ and CeAl$_3$. A strong dependence on field orientation in the magnetic susceptibility of CeAl$_3$ single crystals [13] is suggestive of anisotropic behavior corresponding to $J_z \gg J_\perp > 0$, with the magnetic $z$ direction being the crystallographic $c$ axis.

The AKM is known to be equivalent in the limit of low magnetic field $H = 0$, and for $H = 14$ T, and for CeAl$_3$ at $H = 0$.

We have used a renormalization-group calculation [21] of the specific heat and for CeAl$_3$ for the AKM model in various magnetic fields $H$, with model parameters chosen so that $\alpha = 0.130$ for $H = 0$. See text for details.

Under the assumption that the impurity and the conduction electrons have $g$ factors $g_i = g_z = 2$. (Changing the $g$ factors multiplies the field scale by an overall factor, but does not otherwise affect the results [22].)

The numerical data exhibit three main trends with increasing field: (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 $\gamma$ is greater than that in the peak height, so that $\alpha = \gamma T_m/R$ decreases monotonically with increasing magnetic field, as shown in the legend of Fig. 5.

These numerical results are directly applicable only to single-crystal Ce$_{0.8}$La$_{0.2}$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$_3$ [23] 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 Ce$_{0.8}$La$_{0.2}$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, $\gamma$ rises sufficiently fast that $\alpha$ remains essentially constant up to a 14 T field (see Table I), in contrast to the prediction of the AKM.

The preceding comparisons seem to indicate significant shortcomings in the AKM as a description of Ce$_{0.8}$La$_{0.2}$Al$_3$ in magnetic fields. One reason for the inadequacy of the AKM may be the neglect of magnetic correlations around the temperature of the maximum, as identified in the $\mu$SR studies of Ref. [12]. It was noted above that the specific heat anomaly is reminiscent of an anti-
TABLE I. Values of the specific heat coefficient $\gamma$, the peak temperature $T_m$, and $\alpha = \gamma T_m / R$ (where $R$ is the gas constant) for Ce$_{0.8}$La$_{0.2}$Al$_3$ in different magnetic fields $H$.

| $H$ (T) | $\gamma$ (mJ/K$^2$Ce mol) | $T_m$ (K) | $\alpha$ |
|---------|-------------------------|-----------|---------|
| 0       | 520 ± 20                | 2.13 ± 0.02 | 0.133 ± 0.005 |
| 5       | 640 ± 20                | 1.86 ± 0.02 | 0.143 ± 0.005 |
| 10      | 690 ± 30                | 1.75 ± 0.02 | 0.145 ± 0.006 |
| 14      | 700 ± 40                | 1.70 ± 0.02 | 0.143 ± 0.008 |

The entropy under the peak in $C/T$ is a large fraction (≈50%) of $R \ln 2$, and the linear specific heat coefficient $\gamma = 520$ mJ/K$^2$Ce mol is less than half that of pure CeAl$_3$ ($\gamma = 1250$ mJ/K$^2$Ce mol). However, our field data suggest that any magnetic transition associated with the anomaly is rather unusual. We find that $T_M$ and $T_m$ are depressed in an applied field at a much lower rate than is the Neél temperature in Ce-based heavy fermion systems that order antiferromagnetically. In CeCu$_{5.2}$Au$_{0.8}$, for example, $T_N$ is reduced from 0.7 K to 0 K in a field of about 2.5 T [2]. In CePb$_3$, which exhibits unconventional small-moment ordering at 1.1 K, a field of order 10 T depresses $T_N$ to zero [22].

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$_3$, implying partial restoration of the heavy fermion state suppressed by La doping. 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. Our results do not rule out an alternative theoretical picture based on small-moment magnetism. However, the field-insensitivity of the temperature of the heat-capacity peak remains to be understood within this scenario.

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