Evidence for Anisotropic Kondo Behavior in Ce$_{0.8}$La$_{0.2}$Al$_3$

E. A. Goremychkin,1 R. Osborn,1 B. D. Rainford,2 and A. P. Murani3

1 Argonne National Laboratory, 9700 South Cass Avenue, Argonne, IL 60439
2 Department of Physics, University of Southampton, Southampton SO17 1BJ, United Kingdom
3 Institut Laue-Langevin, BP 156, F-38042 Grenoble Cédex 9, France

(February 26, 2018)

We have performed an inelastic neutron scattering study of the low energy spin dynamics of the heavy fermion compound Ce$_{0.8}$La$_{0.2}$Al$_3$ as a function of temperature and external pressure up to 5 kbar. At temperatures below 3 K, the magnetic response transforms from a quasi-elastic form, common to many heavy fermion systems, to a single well-defined inelastic peak, which is extremely sensitive to external pressure. The scaling of the spin dynamics and the thermodynamic properties are in agreement with the predictions of the anisotropic Kondo model.

PACS numbers: 71.27.+a, 75.30.Mb, 78.70.Nx, 61.12.-q, 76.75.+i

CeAl$_3$ was the first material to be classified as a heavy fermion system over twenty years ago [1]. At first, it was thought to have a non-magnetic ground state, and anomalous features in the thermodynamic and transport properties at about $T^* \sim 0.5$ K were interpreted as the signature of a transition from a single-ion Kondo regime to a coherent Kondo lattice regime [2]. However, Barth et al. [3] presented evidence from muon spin relaxation of quasi-static internal magnetic fields at temperatures below 0.7 K, which suggested the existence of frustrated short-range magnetic order. Since then, there has been conflicting evidence concerning the existence of magnetic order in CeAl$_3$ [4]. Recently, Andraka et al. [5] reported that the temperature of the anomalies in the specific heat and magnetic susceptibility increases gradually with lanthanum doping, from $T^* \sim 0.5$ K in pure CeAl$_3$ to $T^* \sim 2.2$ K in Ce$_{0.8}$La$_{0.2}$Al$_3$, and interpreted this as evidence of a stabilization of the antiferromagnetic ground state due to the changing balance of Kondo and RKKY interactions.

The aim of our investigation was to clarify the microscopic origin of the low temperature anomalies in Ce$_{1-x}$La$_x$Al$_3$ through measurements of the dynamic magnetic correlations, using neutron scattering and muon spin relaxation ($\mu$SR). We find a dramatic evolution of the dynamic magnetic susceptibility, from a quasi-elastic response at high temperature to an inelastic response below $T^*$. In Ce$_{0.8}$La$_{0.2}$Al$_3$, the inelastic peak is at approximately 0.5 meV at 1.7 K. It is extremely sensitive to applied pressure and the dynamics become purely relaxational above only 2 kbars. Although we observe a sharp increase in the $\mu$SR relaxation rate at $T^*$, suggesting the development of static magnetic correlations, we find no evidence of long-range magnetic ordering from high-intensity neutron diffraction. We argue that the anomalous response of Ce$_{0.8}$La$_{0.2}$Al$_3$ is consistent with the predictions of the anisotropic Kondo model $\mathcal{K}^\perp$ which has been attracting considerable theoretical attention recently because of its relation to statistical models of two-level systems with Ohmic dissipation $\mathcal{K}^\perp$. In this interpretation, the transition at $T^*$ is driven by the onset of weakly-dissipative single-ion dynamics, rather than cooperative magnetic ordering.

The sample of Ce$_{0.8}$La$_{0.2}$Al$_3$ was prepared by arc melting stoichiometric quantities of the constituent elements, followed by annealing at 900°C for about four weeks. Both neutron and x-ray diffraction confirmed that the sample was single phase. The neutron scattering experiments were performed at the Institut Laue-Langevin on the time-of-flight spectrometer IN6, using an incident energy of 3.1 meV, the high flux powder-diffractometer D20, using a wavelength of 2.41 Å, and the polarized diffractometer D7, using a wavelength of 4.8 Å. A continuously loaded helium high-pressure cell, operating up to a maximum pressure of 5 kbar, was inserted into a standard helium cryostat for the IN6 experiments. The zero-field $\mu$SR measurements were performed at the ISIS pulsed muon facility, using the MUSR spectrometer.

An example of low temperature IN6 data, in the form of the scattering law $S(Q, \epsilon)$ is shown in Fig. 1. We have modeled $S(Q, \epsilon)$ using a standard Lorentzian lineshape

$$S(Q, \epsilon) \propto F^2(Q) \left( \frac{\epsilon \chi_0}{1 - \exp(-\epsilon/kT)} \right) \times \frac{1}{2\pi} \left( \frac{\Gamma}{(\epsilon - \Delta)^2 + \Gamma^2} + \frac{\Gamma}{(\epsilon + \Delta)^2 + \Gamma^2} \right)$$

(1)

where $F(Q)$ is the Ce$^{3+}$ magnetic form factor, $\chi_0$ is the static susceptibility, and $\Gamma$ the half width at maximum of the Lorentzians centered at energy transfers $\pm\Delta$. The usual paramagnetic response of heavy fermion compounds, including CeAl$_3$ [10], is purely relaxational (i.e. $\Delta = 0$), typically with a square root temperature dependence of the linewidth: $\Gamma(T) = \Gamma_0 + \beta \sqrt{T}$ [11]. Our analysis of the IN6 data from Ce$_{0.8}$La$_{0.2}$Al$_3$ shows that the low energy spin dynamics follow this behavior from 3.3 K up to 100 K with $\beta = 0.18(1)$ meV$^{-1/2}$ and $\Gamma_0 = 0.34(3)$ meV. However, there is a radical change in the shape of response function below 3.3 K, from a
over momentum transfers from 0.3 to 1.1˚

an incident energy of 3.15meV. The spectra were summed
formed both

the specific heat and bulk susceptibility, so we have per-
proximately coincides in temperature with peaks in both
indicating that interionic exchange interactions are small.

in Ce

zero temperature. The linewidth is 0.42(1) meV.

pendent; we estimate that it increases to 0.54(1) meV at
is at 0.474(7) meV at 1.7 K, is weakly temperature de-

in Ce

9

generally broadening due to a static, or quasi-static, dis-
tribution of fields arising from magnetic ordering of the
4f electrons. There was no evidence, within our limited
time resolution, for a second Kubo-Toyabe function or an
oscillatory component, as seen in the µSR data for CeAl
3
3
[3,14]. At high temperatures, the spectra are mainly
determined by the relaxation of the 27Al nuclear moments.
Below a characteristic temperature, T∗ (~ 3K for x = 0.8 and 0.5, ~ 1.5 K for x = 0.3, and ~ 0.5K for x = 0.1), the Lorentzian damping starts to contribute to G(t) (see Fig. 2), with a sharp increase in the damping rate as the temperature is lowered below T∗. The temperature at which λ diverges corresponds to the maximum in the specific heat for x = 0.8 (see Fig. 3).

In support of our attribution of the Lorentzian damping in the µSR data to inhomogeneous broadening arising from quasi-static fields, we note that the increase in the µSR relaxation rate at T∗ occurs when the response becomes purely inelastic. If the damping rate λ were attributed to dynamical processes, we would expect λ ≈ limₜ→0 T S 3/2 χ′(0)/ɛ ∼ S(0) = 0. If it follows that we would expect λ to decrease below T∗, since S(0), determined from the analysis of the inelastic lineshape, falls dramatically at T∗ (see Fig. 3). On the other hand an increase in λ should give rise to an increase in low-frequency magnetic response measured by neutron scattering, for which there is no evidence in the IN6 data. There is no increase in the elastic peak intensity and therefore no evidence of a transfer of spectral weight to an unidentified low-frequency component.

The most direct method of determining the presence of long-range magnetic ordering is neutron diffraction. We have performed a series of experiments on x = 0.8 and 0.5 samples on the high-intensity powder diffractometer D20. The difference in the diffraction patterns measured below and above T∗ shows no evidence of any magnetic Bragg peaks in both compounds. We estimate the Ce magnetic moment in any magnetically ordered phase of Ce0.8La0.2Al3 to be less than 0.05 μB. Furthermore, measurements on D7 with full polarization analysis gave no sign of either magnetic Bragg peaks or significant short-range magnetic order at 1.5 K. It should be noted that the estimated value of the Ce magnetic moment from the heat capacity anomaly of Ce0.8La0.2Al3 is 0.34 μB [3], which would easily be seen by neutron diffraction. However we argue below that most of this heat capacity anomaly arises from the change in form of the single ion dynamics at T∗, so that this estimate of the ordered mo-

FIG. 1. Inelastic neutron scattering data (open circles) from Ce0.8La0.2Al3 measured on the IN6 spectrometer with an incident energy of 3.15meV. The spectra were summed over momentum transfers from 0.3 to 1.1 Å⁻¹. The dotted line is the elastic nuclear scattering, the dashed line is the profile fit of the Eqn. (1), and the solid line is the sum of both contributions. The dash-dotted line is the best fit at T = 1.7K to a quasielastic lineshape.

quasi-elastic to an inelastic form (i.e. Δ ≠ 0). Figure 1 shows that the fit to a quasi-elastic response gives a poor description of the data at 1.7 K. The peak energy, which is at 0.474(7) meV at 1.7 K, is weakly temperature dependent; we estimate that it increases to 0.54(1) meV at zero temperature. The linewidth is 0.42(1) meV.

We note that there is no possibility that this inelastic peak results from a magnon-like excitation within the ground-state doublet. The crystal field (CF) potential in Ce0.8La0.2Al3 is a polynomial in (Jz)², producing a ground state doublet Γ9 (|±3/2⟩) and two excited doublets Γ7 (|±1/2⟩) and Γ8 (|±5/2⟩) at an energy of 7.4 meV [2]. There is no dipole matrix element coupling the |±3/2⟩ states, so conventional magnons would not be measurable with neutrons [2]. Moreover, the excitation energies and linewidths are only weakly Q-dependent, indicating that interionic interaction interactions are small. Nevertheless, the development of this inelastic peak approximately coincides in temperature with peaks in both the specific heat and bulk susceptibility, so we have performed both µSR and neutron diffraction measurements to look for evidence of magnetic ordering below 2 K.

Zero-field µSR spectra were measured on samples of Ce0.8La1-xAl3 with x = 0.8, 0.5, 0.3 and 0.1. All spectra could be fitted to the function G(t), comprising the sum of a Lorentzian and a Kubo-Toyabe (KT) depolarization function, G(t) = A₁ exp(-λt) + A₂GKT(t), where GKT accounts for muon precession due to the dipole fields of the 27Al nuclear magnetic moments [3,14]. Lorentzian damping usually arises from dynamical processes, but here (as in CeAl3 [3,14]), it is likely to arise from inhomogeneous broadening due to a static, or quasi-static, distribution of fields arising from magnetic ordering of the 4f electrons. There was no evidence, within our limited time resolution, for a second Kubo-Toyabe function or an oscillatory component, as seen in the µSR data for CeAl3 [3,14].
interest to us, i.e., when $\alpha < 1/3$, $J_{\parallel} \gg J_{\perp}$.

It is possible to obtain independent estimates of the value of $\alpha$ from the specific heat and magnetic susceptibility results, using scaling relations predicted by the numerical calculations. Firstly, the theory predicts that there is a peak in $C(T)/T$, at a temperature $T^* = \alpha/\gamma$, where $C(T)$ is the specific heat and $\gamma$ is the value of $C(T)/T$ for $T \ll T^*$. The specific heat has only been measured down to 1 K in Ce$_{0.8}$La$_{0.2}$Al$_3$, but we estimate that $\gamma$ should be in the range 0.4-0.5 Jmol$^{-1}$K$^{-2}$ from the extrapolated values measured for $0.9 < x < 1.0$. Since $T^* \approx 2$ K, we obtain $\alpha = 0.10 \pm 0.02$. As a check on this result, we note that $\alpha$ is also given by the inverse Wilson ratio $\gamma/\chi$. From $\chi(T = 1.88 K) = 0.03$ emu/mol, we obtain the identical value $\alpha \approx 0.10$. This represents the upper limit of $\alpha$, because we are likely to have overestimated $\gamma$.

This value of $\alpha$ falls in the regime where the AKM predicts an inelastic response. There have been several numerical calculations of $S(\epsilon)$, which show that it peaks at a renormalized energy $\Delta$, which scales as $\Delta_0(\Delta_0/\omega_c)^{\alpha/(\alpha - 1)}$ where $\omega_c$ is the conduction electron bandwidth. Combining the AKM prediction for the bulk susceptibility, $\chi(T = 0) = \mu_B^2 N_A/2\Delta$ with the measured value of 0.03 emu/mol, gives $\Delta \sim 0.54$ meV. Furthermore, the AKM predicts that $\gamma/\alpha = \pi^2 k_B^2/3\Delta$, from which we estimate that $\Delta$ is between 0.47 and 0.59 meV. The predicted values for $\Delta$ are consistent with the energy of the inelastic peak measured by neutron scattering.

If we apply the same arguments to pure CeAl$_3$, we find that $\gamma \approx 0.31$ (using $\gamma = C(T = 50$ mK$)/T = 1.35$ Jmol$^{-1}$K$^{-2}$ and $\chi(T = 40$ mK$) = 0.0295$ emu/mol), which is very close to the critical value of $\alpha = 1/3$ where the response function $S(\epsilon)$ become quasi-elastic. Inelastic neutron studies of CeAl$_3$ have shown that the magnetic dynamics remain quasi-elastic down to 60 mK. According to the AKM, the decrease in $\alpha$ with increasing $x$ would be due to the “negative” chemical pressure produced by lanthanum dilution. To test this, we have measured the effect of “positive” external hydrostatic pressure on the magnetic response of Ce$_{0.8}$La$_{0.2}$Al$_3$ to see if it drives the system closer to pure CeAl$_3$. Figure 3 shows that the effect of external pressure is remarkably strong, with a reduction in the peak energy evident at only $P = 0.5$ kbar. At 2 kbar, the magnetic response is once again quasi-elastic. $\Delta$ has an almost linear dependence on pressure with $d\Delta/dP = -0.24$ meV/kbar at 1.7 K. We only observe such a strong pressure dependence in the vicinity of $T^*$; at 5 K, the magnetic response is practically pressure-independent. The effects of pressure indicate that the differences in the dynamical behavior of CeAl$_3$ and Ce$_{0.8}$La$_{0.2}$Al$_3$ are not the result of chemical disorder. In the framework of the AKM, it means that the dissipation strength $\alpha$ is extremely pressure-dependent. The observation that the specific heat of pure CeAl$_3$ is only pressure-dependent close to the maximum

![FIG. 2. Temperature dependence of (1) the $\mu$SR relaxation rate $\lambda$ for Ce$_{0.8}$La$_{0.2}$Al$_3$: $x = 0.8$ (solid squares; absolute values divided by 3); $x = 0.5$ (open circles); $x = 0.3$ (open triangles), and $x = 0.1$ (open squares), (2) the neutron scattering function at zero energy transfer, $S(\epsilon \rightarrow 0)$ (solid triangles), and (3) the specific heat $C(T)/T$ (solid circles; from Ref. [8]). The lines are guides to the eye.](image-url)
in $C(T)/T$ is consistent with this explanation. We have argued that, as in CeAl$_3$, the µSR results are evidence of magnetic ordering, of either short or long range, with moments less than $\sim 0.05 \mu_B$, similar to what has been observed in several uranium heavy fermion compounds. The spectral weight associated with such weak magnetic correlations would not be measurable by neutron scattering from polycrystalline samples. Costi and Kieffer showed that the inelastic peak in $S(\epsilon)$ persists in the presence of a bias field, as long as this is small compared to $\Delta$. Equating the bias field with the internal exchange field, we infer that antiferromagnetism or spin glass order with weak moments would be unlikely to change the form of the single-ion dynamics. Such ordering would be insufficient to drive the thermodynamics and so is more likely to be a by-product of the reduced dissipation of the ground state. In this scenario, the transition at $T^*$ is produced by the single-ion AKM, but allows the development of a more coherent $f$-electron band at low temperature. The small-moment magnetism would be a manifestation of the itinerant nature of the $f$-electrons in this regime.

Other non-cubic heavy fermion systems, such as CeRu$_2$Si$_2$, have highly anisotropic susceptibilities, so they should be considered in the context of the anisotropic Kondo effect; indeed, the heuristic model used to describe the spin dynamics in CeRu$_2$Si$_2-x$Ge$_x$ alloys had many of the features of the dynamics of the AKM. The unusual properties of URu$_2$Si$_2$, which is also highly anisotropic, might be associated with the AKM. Most of the spectral weight is in the dynamical response, which is also characterized by longitudinal fluctuations. The ordered moment is very small (0.04 $\mu_B$), yet there is a large heat capacity anomaly at $T_N$. An extension of the AKM to account for intersite interactions would be necessary for a description of the strong dispersion of the magnetic excitations.

In conclusion, we have shown that anomalies in the specific heat and magnetic susceptibility of Ce$_{0.8}$La$_{0.2}$Al$_3$ are not driven by the development of static magnetic correlations, but are associated with the development of the single-ion inelastic response of the cerium $4f$-electrons, arising from their coupling to the conduction electrons. The scaling of the bulk and dynamic properties is consistent with the predictions of the anisotropic Kondo model, from which we conclude that the anisotropy of coupling in CeAl$_3$ is close to the critical value at which a weakly-dissipative response is observable. These results provide a new insight into the mechanisms by which low-temperature coherence is established in heavy fermions.

We thank I. Sashin, T. Hansen, C. A. Scott, and K. H. Andersen for their help with the experiments, and D. Grempel for useful discussions. EAG thanks the Rutherford Appleton Laboratory for hospitality and financial assistance. This work was supported by the US Department of Energy Contract No. W-31-109-ENG-38.

FIG. 3. Inelastic neutron scattering data from Ce$_{0.8}$La$_{0.2}$Al$_3$ vs. pressure at $T = 1.7$K. The symbols and lines are the same as in Fig. 1.