Evidence for Anisotropic Kondo Behavior in $Ce_{0.8}La_{0.2}Al_3$

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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 quasielastic 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.

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$CeAl_3$ was the first material to be classified as a heavy fermion system over 20 years ago [1]. At first, it was thought to have a nonmagnetic 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 quasistatic 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 $Ce_xLa_{1-x}Al_3$ through measurements of the dynamic magnetic correlations, using neutron scattering and muon spin relaxation (μSR). We find a dramatic evolution of the dynamic magnetic susceptibility, from a quasielastic 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 μSR 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 [6–8], which has been attracting considerable theoretical attention recently because of its relation to statistical models of two-level systems with Ohmic dissipation [9]. 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 μSR 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 line shape

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

where $F(Q)$ is the Ce^{3+} magnetic form factor, χ_0 is the static susceptibility, and Γ is the half width at half-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 $K^{-1/2}$

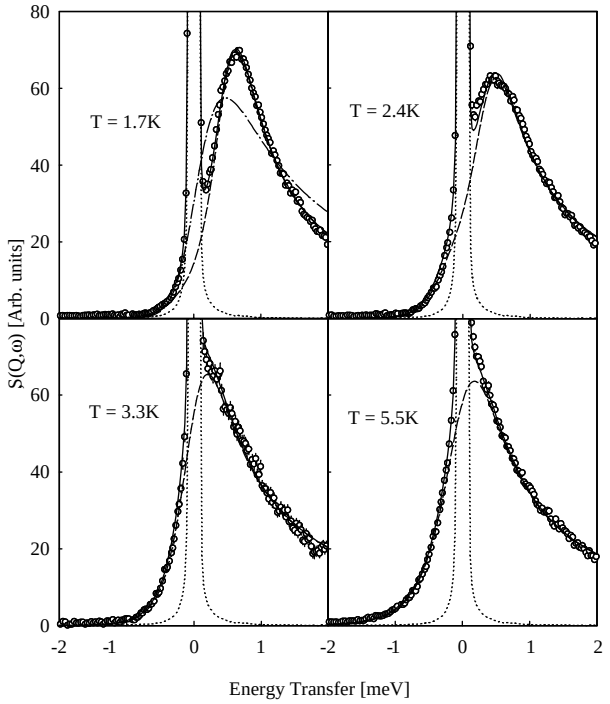

FIG. 1. Inelastic neutron scattering data (open circles) from $Ce_{0.8}La_{0.2}Al_3$ measured on the IN6 spectrometer with an incident energy of 3.15 meV. The spectra were summed over momentum transfers from 0.3 to 1.1 Å $^{-1}$. The dotted line is the elastic nuclear scattering, the dashed line is the profile fit of Eq. (1), and the solid line is the sum of both contributions. The dash-dotted line is the best fit at $T = 1.7$ K to a quasielastic line shape.

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 quasielastic to an inelastic form (i.e., $\Delta \neq 0$). Figure 1 shows that the fit to a quasielastic 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 magnonlike excitation within the ground-state doublet. The crystal field (CF) potential in $Ce_{0.8}La_{0.2}Al_3$ is a polynomial in $(J_z)^2$, producing a ground state doublet Γ_9 ($|\pm 3/2\rangle$) and two excited doublets Γ_7 ($|\pm 1/2\rangle$) and Γ_8 ($|\pm 5/2\rangle$) at an energy of 7.4 meV [12]. There is no dipole matrix element coupling the $|\pm 3/2\rangle$ states, so conventional magnons would not be measurable with neutrons [13]. Moreover, the excitation energies and linewidths are only weakly Q dependent, indicating that interionic exchange 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 $Ce_xLa_{1-x}Al_3$ 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, $G(t) = A_1 \exp(-\lambda t) + A_2 G_{\mathrm{KT}}(t)$, where G_{KT} accounts for muon precession due to the dipole fields of the ^{27}Al nuclear magnetic moments [3,14]. Lorentzian damping usually arises from dynamical processes, but here (as in $CeAl_3$ [14]), it is likely to arise from inhomogeneous broadening due to a static, or quasistatic, 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 $CeAl_3$ [3,14]. At high temperatures, the spectra are mainly determined by the relaxation of the ^{27}Al nuclear moments. Below a characteristic temperature, T^* ($\sim$3 K for $x = 0.8$ and 0.5, $\sim$1.5 K for $x = 0.3$, and $\sim$0.5 K 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. 2).

In support of our attribution of the Lorentzian damping in the μSR data to inhomogeneous broadening arising from quasistatic 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 $\lambda \approx \lim_{\epsilon \to 0} T\chi''(\epsilon)/\epsilon \approx S(\epsilon \to 0)$. It follows that we would expect λ to *decrease* below T^*, since $S(0)$, determined from the analysis of the inelastic line shape, falls dramatically at T^* (see Fig. 2). 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

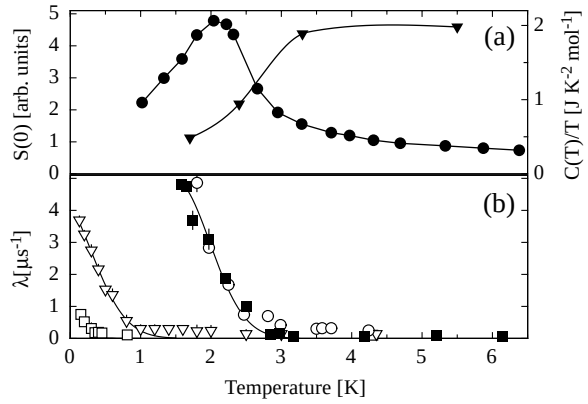

FIG. 2. Temperature dependence of (a) the neutron scattering function at zero energy transfer, $S(\epsilon \to 0)$ (solid triangles), and the specific heat $C(T)/T$ (solid circles; from Ref. [5]); (b) the μSR relaxation rate λ for $Ce_xLa_{1-x}Al_3$: $x = 0.8$ (solid squares; absolute values divided by 3); $x = 0.5$ (open circles); $x = 0.3$ (open triangles); and $x = 0.1$ (open squares). The lines are guides to the eye.

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 $Ce_{0.8}La_{0.2}Al_3$ to be less than $0.05\mu_B$. 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 $Ce_{0.8}La_{0.2}Al_3$ is $0.34\mu_B$ [5], 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 moment may be discounted. The evidence from the neutron diffraction experiments suggests that if magnetic ordering occurs below T^*, as suggested by the μSR data, it must be associated with an extremely small amplitude of the magnetic moment. We can then infer that the exchange field will be far too small to drive the change in the dynamical response below T^*. In order to find an explanation that can encompass the entire composition range from $x = 0.1$ to 1.0, we have explored an alternative account of the dynamical behavior.

In recent years, there has been substantial interest in the anisotropic Kondo model (AKM) [6–8] as a theoretical realization of a dissipative two-state system (DTSS) [9]. In the AKM, the interaction between a localized spin and the conduction electrons is anisotropic and there are two parameters representing the strength of this interaction, $J_\parallel$ and $J_\perp$, with $J_\parallel \neq J_\perp$. $CeAl_3$ is a natural candidate for such a model, because the anisotropy of the low-temperature magnetic susceptibility is very strong; the $|\pm 3/2\rangle$ crystal field ground state is Ising-like ($g_\perp = 0$ and $g_\parallel = 18/7$) and the bulk susceptibility is dominated by a divergent Curie term $\chi_\parallel(T)$ below 40 K [12]. The Van Vleck contribution from the excited crystal field states, $\chi_\perp(T)$, is much weaker.

The dynamics of the DTSS model are governed by the bare splitting of the two states at an energy Δ_0, and the strength of the dissipation produced by the bosonic continuum, which is characterized by the dimensionless parameter α. Below a critical value of α, estimated to be roughly $1/3$ by Costi and Kieffer [7], the dynamic response is predicted to be inelastic, but when $\alpha > 1/3$, the response is overdamped. The DTSS model may be mapped onto the AKM, with $J_\perp$ proportional to Δ and $J_\parallel$ associated with α. In the weakly dissipative regime of 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 α from the specific heat and magnetic susceptibility results, using scaling relations predicted by the numerical calculations. First, 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 γ is the value of $C(T)/T$ for $T \ll T^*$ [8]. The specific heat has been measured only down to 1 K in $Ce_{0.8}La_{0.2}Al_3$, but we estimate that γ should be in the range 0.4–0.5 $\mathrm{J\,mol^{-1}\,K^{-2}}$ from the extrapolated values measured for $0.9 < x < 1.0$ [5]. Since $T^* \approx 2$ K [5], we obtain $\alpha = 0.10 \pm 0.02$. As a check on this result, we note that α is also given by the inverse Wilson ratio γ/χ. From $\chi(T = 1.8\ \mathrm{K}) = 0.03$ emu/mol [5], we obtain the identical value $\alpha \approx 0.10$. This represents the upper limit of α, because we are likely to have overestimated γ.

This value of α 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 Δ, which scales as $\Delta_0(\Delta_0/\omega_c)^{\alpha/(1-\alpha)}$ where ω_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 $\gamma/\alpha = \pi^2 k_B^2/3\Delta$ [8], from which we estimate that Δ is between 0.47 and 0.59 meV. The predicted values for Δ 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 $\alpha \approx 0.31$ (using $\gamma = C(T = 50\ \mathrm{mK})/T = 1.35\ \mathrm{J\,mol^{-1}\,K^{-2}}$ [15] and $\chi(T = 40\ \mathrm{mK}) = 0.0295$ emu/mol [16]), which is very close to the critical value of $\alpha = 1/3$ where the response function $S(\epsilon)$ become quasielastic. Inelastic neutron studies of $CeAl_3$ have shown that the magnetic dynamics remain quasielastic down to 60 mK [10]. According to the AKM, the decrease in α 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 quasielastic. Δ has an almost linear dependence on pressure with $d\Delta/dP = -0.24$ meV/kbar at 1.7 K. We observe such a strong pressure dependence only 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 α is extremely pressure dependent. The observation that the specific heat of pure $CeAl_3$ is only pressure dependent close to the maximum in $C(T)/T$ [15] 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

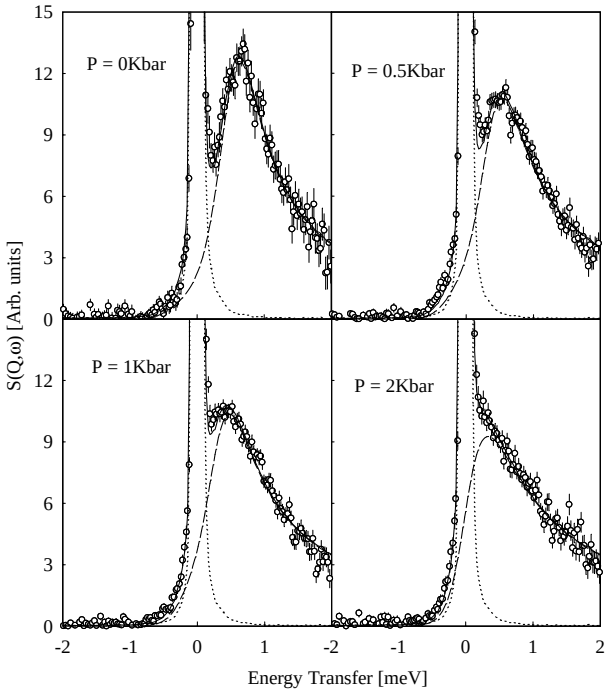

FIG. 3. Inelastic neutron scattering data from $Ce_{0.8}La_{0.2}Al_3$ vs pressure at $T = 1.7$ K. The symbols and lines are the same as in Fig. 1.

has been observed in several uranium heavy fermion compounds [14]. The spectral weight associated with such weak magnetic correlations would not be measurable by neutron scattering from polycrystalline samples. Costi and Kieffer [7] showed that the inelastic peak in $S(\epsilon)$ persists in the presence of a bias field, as long as this is small compared to Δ. 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 byproduct 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 noncubic heavy fermion systems, such as $CeRu_2Si_2$, 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 $CeRu_2Si_{2-x}Ge_x$ alloys [13] had many of the features of the dynamics of the AKM [7]. The unusual properties of URu_2Si_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 [17,18]. 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.

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