

Absence of Kondo lattice coherence effects in $\text{Ce}_{0.6}\text{La}_{0.4}\text{Pb}_3$: A magnetic-field study

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The specific heat of polycrystalline $\text{Ce}_{0.6}\text{La}_{0.4}\text{Pb}_3$ has been measured in magnetic fields ranging from 0 to 14 T. After subtraction of a lattice contribution, the specific heat between 1 K and 10 K is well described by the $S=\frac{1}{2}$ single-impurity Kondo model with just one adjustable parameter: the zero-field Kondo temperature. In particular, the variation in the temperature and the height of the peak in C vs T is captured with good accuracy. This fit suggests that lattice coherence effects play no significant role in the magnetic-field response of this concentrated Kondo system. © 2005 American Institute of Physics. [DOI: 10.1063/1.1855456]

I. INTRODUCTION

Lately, there has been renewed interest in the subject of the Kondo lattice and its relation to the single-impurity Kondo model. Nakatsuji *et al.*¹ have proposed a two-fluid model that describes thermodynamic and transport properties of $\text{Ce}_x\text{La}_{1-x}\text{CoIn}_5$ by the superposition of a single-impurity part and a coherent heavy-fermion liquid part. It has been shown for the concentrated Ce alloys ($x > 0.5$) that only 10% of the low-temperature specific heat corresponds to the single-impurity part, and that this part can be described by the same Kondo temperature T_K for all concentrations x .

This remarkable result, which suggests that the specific heat is essentially a coherent lattice property, requires reexamination of previous investigations concluding that single-impurity physics accounts rather well for the thermodynamic properties of a number of heavy fermions. Of particular note is the alloy series $\text{Ce}_x\text{La}_{1-x}\text{Pb}_3$, for which the zero-field specific heat scales with Ce concentration for $0 < x \leq 0.6$, and the specific heat per Ce is accounted for quantitatively by the $S=\frac{1}{2}$ single-impurity Kondo model.² The absence of coherence effects in $\text{Ce}_x\text{La}_{1-x}\text{Pb}_3$ is surprising given that the pure compound CePb_3 orders antiferromagnetically at $T_N=1.1$ K, pointing to the presence of significant inter-ion correlations.

In order to provide a more rigorous test of the single-impurity picture in $\text{Ce}_x\text{La}_{1-x}\text{Pb}_3$, we have measured the specific heat of polycrystalline $\text{Ce}_{0.6}\text{La}_{0.4}\text{Pb}_3$ in magnetic fields ranging from 0 to 14 T. The $x=0.6$ concentration was chosen to satisfy two criteria: the system should be sufficiently concentrated and it should not order magnetically at any temperature. After subtraction of a lattice contribution, we find that the specific heat between 1 K and 10 K is well described by the $S=\frac{1}{2}$ single-impurity Kondo model with just one adjustable parameter: the zero-field Kondo temperature, T_K

$=2.6 \pm 0.2$ K. In particular, the variation in the temperature and the height of the peak in C vs T is captured with good accuracy. This fit is nontrivial, given that the impurity has a field-dependent g factor arising from field-induced mixing of Ce crystalline electric field levels.

II. EXPERIMENT

Two polycrystalline samples of $\text{Ce}_{0.6}\text{La}_{0.4}\text{Pb}_3$ were synthesized independently in an arc melter using highest available grade elements (Ce and La from Ames National Laboratory, Pb 6N from AESAR Johnson Matthey). Because of the high vapor pressure of Pb, the starting material had additional Pb to compensate for vapor losses. The starting composition for sample 1 had 3% more Pb than indicated by stoichiometry, while that for sample 2 had an additional 2%. Each sample was repeatedly remelted to improve the homogeneity. After each remelting, the sample mass was compared to that expected for the stoichiometric material under the assumption that there were no vapor losses of Ce and La at the low arc current used. The process was repeated until the final stoichiometry (assuming no loss of Ce and La) was $\text{Ce}_{0.6}\text{La}_{0.4}\text{Pb}_{3.00 \pm 0.01}$. Each sample was then annealed for two weeks at 600 °C in the presence of additional free lead to minimize further Pb losses from the sample. (No such losses were detected.)

The specific heat C of sample 1 was measured by the thermal relaxation method over temperatures T between 0.4 K and 10 K in magnetic fields $H=0, 5, 8, 10,$ and 14 T. Sample 2 was measured between 0.7 K and 4.2 K at zero field and 10 T to provide a basis for estimating the likely degree of sample dependence in the data.

Figures 1 and 2 plot the excess specific heat $\Delta C = C - C_{\text{lat}}$ normalized per mole of Ce. Here, C_{lat} is the lattice (phonon) contribution, estimated from the data of Lin *et al.*² Figure 1 also shows error bars for a few representative points. The uncertainties reflect possible errors both in the measured total specific heat and in the lattice correction. By 10 K, C_{lat} makes up roughly 90% of C , so the uncertainty in ΔC is particularly large at the upper end of the measured temperature range.

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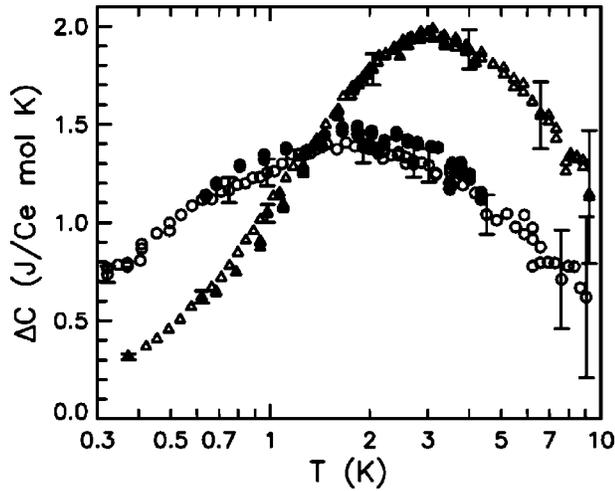


FIG. 1. Excess specific heat ΔC vs temperature T for sample 1 (open symbols) and sample 2 (filled symbols) at magnetic fields $H=0$ (circles) and $H=10$ T (triangles). Error bars are shown for a few representative points.

III. THEORY

We modeled the experimental data using the spin- $\frac{1}{2}$ Kondo impurity model described by the Hamiltonian

$$\hat{H}_K = \sum_{\mathbf{k}, \sigma} (\epsilon_{\mathbf{k}} + \sigma g_c \mu_B H) c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma} + g_i \mu_B H S_z + JS \cdot \sum_{\mathbf{k}, \sigma} \sum_{\mathbf{k}', \sigma'} c_{\mathbf{k}\sigma}^\dagger \sigma_{\sigma\sigma'}^j c_{\mathbf{k}'\sigma'} \quad (1)$$

Here $\epsilon_{\mathbf{k}}$ describes the conduction-band dispersion, $\sigma = \pm \frac{1}{2}$ labels the conduction-electron spin projection along the direction of the magnetic field, μ_B is the Bohr magneton, g_c and g_i are the conduction-band and impurity g factors, respectively, J is the Kondo exchange, \mathbf{S} is the impurity spin operator, and $\sigma_{\sigma\sigma'}^j$ ($j=1, 2, 3$) are the Pauli matrices.

At zero field, the model has a single low-energy scale³ $k_B T_K \approx \epsilon_F \exp[-1/\rho(\epsilon_F)J]$, where k_B is Boltzmann's constant and $\rho(\epsilon_F)$ is the density of states at the Fermi energy ϵ_F .

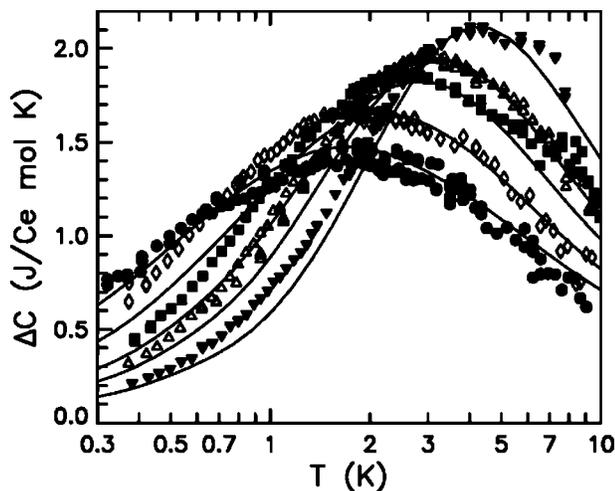


FIG. 2. Excess specific heat ΔC vs temperature T in fields of 0 T (●), 5 T (◇), 8 T (■), 10 T (△), and 14 T (▼). Data for samples 1 and 2 are combined. The lines show C_{imp} for the single-impurity Kondo model [Eq. (1)] calculated for the experimental H values, with g_i set to the corresponding $\langle g_i \rangle$ listed in Table I.

Since the quantities entering the Kondo temperature T_K were not measured experimentally, we calculated C_{imp} , the impurity contribution to the heat capacity, using the numerical renormalization group (NRG) method^{3,4} for an arbitrary choice $\rho(\epsilon_F)J=0.2$ and then fitted the temperature scale of the data for $\text{Ce}_{0.6}\text{La}_{0.4}\text{Pb}_3$, focusing particularly on the region around the peak in C (which occurs in both samples at $T \approx 1.8 \pm 0.2$ K). This process yielded a value $T_K = 2.6 \pm 0.2$ K, some 20% lower than that obtained by Lin *et al.*² We have no explanation for this discrepancy.

In magnetic fields, it is also necessary to know g_c and g_i . The calculated C_{imp} is insensitive to the value of the conduction-band g factor, which we took to be $g_c=2$. The impurity g factor g_i is deduced by mapping the lowest pair of energy levels of Ce^{3+} onto an effective spin- $\frac{1}{2}$ degree of freedom, as described in the remainder of this section.

In the cubic crystalline electric field (CEF) environment of $\text{Ce}_x\text{La}_{1-x}\text{Pb}_3$, the six $J=5/2$ atomic levels of atomic Ce^{3+} , $\{\lvert m_J \rangle\}$, split into a Γ_7 doublet and a Γ_8 quartet:⁵

$$\begin{aligned} |\Gamma_7, \pm\rangle &= \sqrt{\frac{1}{6}} \left| \pm \frac{5}{2} \right\rangle - \sqrt{\frac{5}{6}} \left| \mp \frac{3}{2} \right\rangle, \\ |\Gamma_8, 1, \pm\rangle &= \sqrt{\frac{5}{6}} \left| \pm \frac{5}{2} \right\rangle + \sqrt{\frac{1}{6}} \left| \mp \frac{3}{2} \right\rangle, \\ |\Gamma_8, 2, \pm\rangle &= \left| \pm \frac{1}{2} \right\rangle. \end{aligned} \quad (2)$$

The preponderance of experimental evidence⁶⁻⁸ indicates that CePb_3 has a Γ_7 ground state.⁹ The Γ_7 - Γ_8 splitting temperature T_{CEF} has been variously estimated from the magnetic susceptibility⁶ to be 67 K, from the elastic constants⁷ to be 76 K, and from inelastic neutron scattering to be 67 K (Ref. 8) and 72 K (Ref. 10). It is probable that the CEF scheme is affected only weakly by substitution of La for some Ce atoms since the immediate environment of each remaining Ce is unaffected, so we set $T_{\text{CEF}}=72$ K in our calculations.

In a magnetic field \mathbf{H} , the CEF states are mixed by the Zeeman interaction. Therefore, the effective Hamiltonian governing the atomic $J=5/2$ Ce^{3+} multiplet is

$$\hat{H}_{\text{Ce}} = k_B T_{\text{CEF}} \sum_{j=1}^2 \sum_{\sigma=\pm} |\Gamma_{8,j}, \sigma\rangle \langle \Gamma_{8,j}, \sigma| + g \mu_B \mathbf{J} \cdot \mathbf{H}, \quad (3)$$

where μ_B is the Bohr magneton and the Landé g factor for Ce^{3+} ($J=5/2$, $L=3$, $S=\frac{1}{2}$) is $g=6/7$.

For a given \mathbf{H} , we diagonalized \hat{H}_{Ce} and found the splitting ΔE between the lowest two energy eigenvalues. We then used the relation $\Delta E = g_i \mu_B H$ to deduce an effective value of $g_i(\mathbf{H})$ to insert into the $S=\frac{1}{2}$ Kondo impurity model. This effective g factor is dependent on both the magnitude of \mathbf{H} and its orientation relative to the crystal axes. The strongest variation of g_i with H is found for fields oriented along the $\langle 100 \rangle$ directions, while the weakest variation occurs for fields along $\langle 111 \rangle$.

To model the random orientation of our polycrystals relative to the field, we averaged $g_i(\mathbf{H})$ over all directions of \mathbf{H} at fixed $H=|\mathbf{H}|$. Table I shows the largest value, the smallest value, the mean, and the standard deviation of g_i for each

TABLE I. Properties of Ce^{3+} in CePb_3 cubic crystalline electric fields with an applied magnetic field of magnitude H . $k_B\tilde{T}_{\text{CEF}}$ is the minimum over all field orientations of the energy gap between the second and third levels. $\max g_i$ and $\min g_i$ are the maximum and minimum values over all field orientations of the effective impurity g factor deduced from the splitting between the first and second energy levels. $\langle g_i \rangle$ and $\sigma(g_i)$ are the mean and standard deviation of g_i , respectively.

$H(\text{T})$	$\tilde{T}_{\text{CEF}}(\text{K})$	$\max g_i$	$\min g_i$	$\langle g_i \rangle$	$\sigma(g_i)$
0	72	1.429	1.429	1.429	0.000
5	65	1.431	1.412	1.423	0.005
8	61	1.434	1.387	1.415	0.012
10	59	1.436	1.364	1.407	0.019
14	55	1.443	1.302	1.387	0.037

field H at which the specific heat was measured, as well as the minimum splitting \tilde{T}_{CEF} between the second and third lowest eigenenergies.

The table shows that over the range of fields covered in our experiments, the lowest-lying pair of states remains well separated in energy from the remaining four states. This separation ($\tilde{T}_{\text{CEF}} \geq 55$ K) justifies the neglect of the higher levels at temperatures $T \leq 10$ K.

Second, it turns out that although the field causes quite strong mixing of CEF levels, the distribution of g_i values remains fairly narrow, and the mean value shows a rather weak dependence on H . We used just the mean value $\langle g_i \rangle$ in a NRG calculation of C_{imp} at each magnetic field. (The NRG calculations are computer intensive, and it was therefore impractical to average over the entire distribution of g_i values.) We discuss the likely effect of this approximation in the next section.

IV. DISCUSSION

Figure 1 shows the excess specific heat for samples 1 and 2, at fields $H=0$ and $H=10$ T. At each field, the data for the two samples lie close to one another. In particular, the location (in temperature) and height of the peak in ΔC are very consistent between the samples. Given that samples 1 and 2 were synthesized independently, the agreement between their specific heats (both in zero and nonzero fields) suggests that the data represent the intrinsic properties of $\text{Ce}_{0.6}\text{La}_{0.4}\text{Pb}_3$, and are not merely sample-specific artifacts.

Figure 2 compares all the excess specific heat data for both samples with the impurity specific-heat contribution C_{imp} computed as described in the preceding section. Given

that there is no adjustable parameter beyond the Kondo temperature T_K deduced from the zero-field data, there is good agreement between experiment and theory. The Kondo model reproduces the field variation of the height of the specific heat peak with very high accuracy. The temperature of the peak in ΔC is also well described, the only significant deviation occurring for $H=5$ T.

In all cases $H>0$, the peak in the experimental data is somewhat broader than predicted by the Kondo model, particularly on the low-temperature side of the maximum in ΔC . This broadening may be partially attributable to the spread in g_i values arising from the distribution of angles between the magnetic field and the cubic crystal axes. As indicated by the maximum and minimum values listed in Table I, the g_i distribution has longer tails on the low- g_i side of $\langle g_i \rangle$, which should tend to produce an asymmetry of the type seen in ΔC vs T .

The rather minor differences between the field dependence observed in the specific heat of $\text{Ce}_{0.6}\text{La}_{0.4}\text{Pb}_3$ and that predicted by the single-impurity Kondo model suggest that there is no significant coherence effect in the specific heat of this system, at least at temperatures of order the single-ion Kondo scale and higher. This provides a clear counter example to the two-fluid model advanced¹ for $\text{Ce}_x\text{La}_{1-x}\text{CoIn}_5$ and $\text{Ce}_x\text{La}_{1-x}\text{IrIn}_5$, where coherence effects set in at temperatures more than an order of magnitude above T_K .

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¹S. Nakatsuji, D. Pines, and Z. Fisk, Phys. Rev. Lett. **92**, 016401 (2004).

²C. L. Lin, A. Wallash, J. E. Crow, T. Mihalisin, and P. Schlottmann, Phys. Rev. Lett. **58**, 1232 (1987).

³K. G. Wilson, Rev. Mod. Phys. **47**, 773 (1975).

⁴W. C. Oliveira and L. N. Oliveira, Phys. Rev. B **49**, 11986 (1994).

⁵K. R. Lea, M. J. M. Leake, and W. P. Wolf, J. Phys. Chem. Solids **23**, 1381 (1962).

⁶P. Lethuillier and J. Chaussy, J. Phys. (Paris) **37**, 123 (1976).

⁷D. Nickl, I. Kouroudis, W. Assmus, B. Lüthi, G. Bruls, and U. Welp, Phys. Rev. B **35**, 6864 (1987).

⁸B. Renker, E. Gering, F. Gompf, H. Schmidt, and H. Rietschel, J. Magn. Magn. Mater. **63–64**, 31 (1987).

⁹A Γ_8 CEF ground state split into two doublets by quadrupolar interactions was proposed by D. Dürkop, E. Braun, B. Politt, H. Schmidt, and D. Wohlleben, Z. Phys. B: Condens. Matter **63**, 55 (1986).

¹⁰C. Vettier, P. Morin, and J. Flouquet, Phys. Rev. Lett. **56**, 1980 (1986).