Low-temperature elastic constants of $CeAl_3$

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Measurements of longitudinal and shear elastic constants in polycrystalline $CeAl_3$ are presented. The measurements were extended to very low temperatures (50 mK). A strong softening for the longitudinal elastic constant was observed in the temperature region of the crystal-field splitting of Ce^{3+} (10–100 K), and an even stronger softening occurred below 4 K down to 0.6 K. Below 0.6 K down to 50 mK both elastic constants exhibit shallow minima. This behavior is correlated with other physical properties in the same temperature region. It is argued that one observes a transition from a region dominated by a single-ion Kondo effect of the crystal-field-split Ce^{3+} ion ($T > 0.6$ K) to a new coherent state below 0.5 K.

The intermetallic compound $CeAl_3$ exhibits anomalous physical properties both at high and low temperatures.[1] The electrical resistivity is unusually high, has a maximum at 35 K, and decreases again at higher temperatures. Down to 4 K the susceptibility shows a Curie-Weiss behavior expected for the crystal-field-split Ce^{3+} ($J = 5/2$) doublet states,[2] but no magnetic order is found even at 10 mK. Below 1 K an extremely high electronic specific heat[3] ($\gamma = 1620$ mJ/mole K^2) is observed, together with anomalous behavior in the susceptibility, thermal expansion,[3,4] and magnetoresistance.[5] All these phenomena are thought to arise from the close proximity of the $4f$ electron of Ce^{3+} to the Fermi energy, which makes the compound nearly unstable against a valence change to the Ce^{4+} $5d^2$ $4f^0$ configurations. It causes also the $4f$ - $5d$ exchange interaction to be unusually large.

Thus the resistivity behavior has been qualitatively described by resonant scattering on the broadened and crystal-field-split three doublet states, invoking either the Kondo effect[6] or virtual-bound-state scattering.[3] The width of the many-body $4f$ - $5d$ resonance[7] as estimated from the low-temperature properties, is about $T_K = 4$ K. The very-low-temperature behavior, which shows the characteristics of a heavy Fermi liquid, appears to arise from the onset of phase coherence among the single-ion "Kondo-condensed" states. It is still not well understood.

In order to characterize this compound further, we have performed eleastic constant measurements in a wide temperature and magnetic field range. The temperature dependence was measured from room temperature down to 50 mK using a dilution refrigerator system[8] for temperatures below 2 K, giving direct outside access to the mixing chamber. The magnetic field dependence was measured in a superconductivity magnet for temperatures at $T = 1$ and 4 K. The sample used for this investigation comes from the same batch as the one used for previous investigations.[3]

Figure 1 shows an overall view of the temperature dependence of the longitudinal and shear elastic constants (c_L and c_T). The density used for converting sound velocities to elastic con-

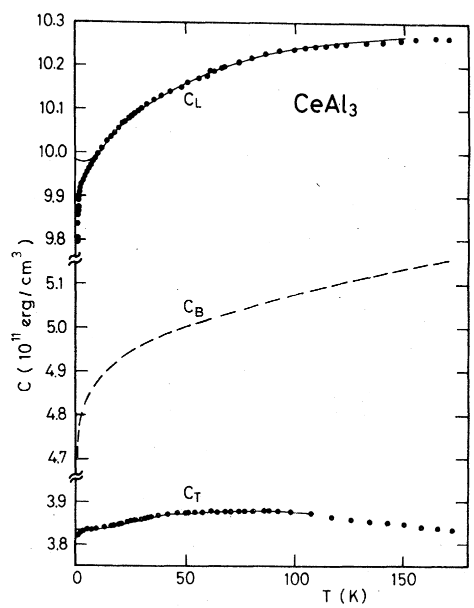

FIG. 1. Temperature dependence of elastic constants for polycrystalline $CeAl_3$. c_L is for the longitudinal mode, c_T for the transverse mode, and c_B is the calculated bulk modulus. Full lines are the theoretical fit of strain susceptibility.

stants is $\rho = 4.29$ g/cm^3. Since $CeAl_3$ cannot yet be prepared in single-crystal form, only c_L and c_T can be measured. The bulk modulus c_B is calculated using the averaging formula $c_B = c_L - \frac{4}{3} c_T$ for polycrystalline material and is also shown as the dashed line in Fig. 1. A brief account of the elastic constants for $T > 1$ K was given in a conference report.[9]

Figure 1 shows a strong softening of c_L and a somewhat less pronounced one for c_T for $T > 5$ K. The elastic constant behavior in this temperature region can be explained by magnetoelastic effects due to the crystal-field-split Ce^{3+} ions in the usual way.[10] However, in this polycrystalline material the strain susceptibility χ_S is determined by magnetoelastic coupling constants of different symmetries. In the notation of Ref. 10 the elastic constant is given by $c = c_0 - g_2^2 N \chi_S P(O_2^0) - g_3^2 N \chi_S(O_{xy}) - g_4^2 N \chi_S(O_{xz})$ with different g_i for longitudinal and shear waves and O_Γ denoting the quadrupolar operators for the different symmetries. One gets a good fit to the experiment for $T > 10$ K with a crystal-field splitting of $E_{3/2} - E_{5/2}$ (18 K) $-E_{1/2}$ (90 K) as shown by the full lines in Fig. 1. It should be stressed that, while the temperature dependence of c_L and c_T in this temperature region is definitely due to this magnetoelastic interaction, the large number of adjustable parameters (c_0, g_2, g_3, g_4) make a quantitative fit not so convincing a case as in the case of single-crystal materials.[10]

The interesting aspects of our measurements are the data for $T < 10$ K. In Fig. 2 we show the data for c_L and c_T for $T < 4$ K. One notices a further strong softening, especially for c_L, for the temperature region $0.6 < T < 3$ K and a rather abrupt flattening for $T < 0.6$ K with a shallow minimum at $T \sim 0.5$ K for both modes. We interpret the strong softening (of about 2% for c_L in this small temperature region) to a renormalization of the elastic constant due to the many-body resonant interaction, with the strain derivative $\partial T_K / \partial \epsilon$ being the coupling constant. A qualitative fit to the thermal expansion data[3,4] for $T > 1$ K using the resonance model[7] gives $\partial T_K / \partial \epsilon \sim -31$ K/ion a value found for Ce^{3+} Kondo ions in other compounds.[11] A quantitative theory for the elastic constants in this Kondo regime has not been given yet.

For $T < 1$ K one observes for both c_L and c_T modes shallow minima at $T \sim 0.5$ K, indicating that for $CeAl_3$ in this temperature region a new phenomena develops, possibly a coherent low-temperature state. This experimental observation is substantiated by the following other facts: (1) The magnetic susceptibility χ_m exhibits a small maximum at $T \sim 0.6$ K. This maximum cannot be explained by the single-ion resonance model.

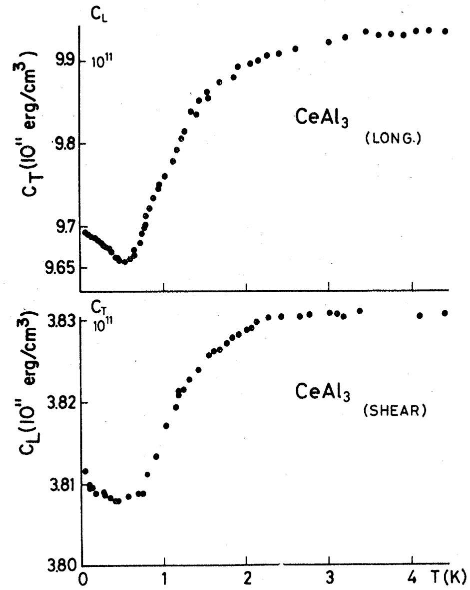

FIG. 2. Elastic constants c_L and c_T in the temperature region from 50 mK to 4 K.

(2) The thermal expansion β changes sign at $T \lesssim 1$ K and exhibits a minimum[4] at $T \lesssim 0.5$ K. Again this cannot be explained by the single-ion resonance model which gives for $T_K \sim 4$ K a maximum at $T \sim 2$ K as observed experimentally, and decreases monotonically to 0 K. (3) The magnetoresistance exhibits an extremum at $T \sim 0.5$ K and changes sign at ~ 0.8 K. (4) The electrical resistivity exhibits a T^2 law up to $T \lesssim 0.3$ K and changes to linear dependence for higher temperatures.

All these experimental facts taken together, indicate that for $CeAl_3$ the physical properties for $T > 1$ K are probably well described by Kondo-type effects in the presence of a crystal-field-split stable Ce^{3+} state, but that for $T < 1$ K a com-

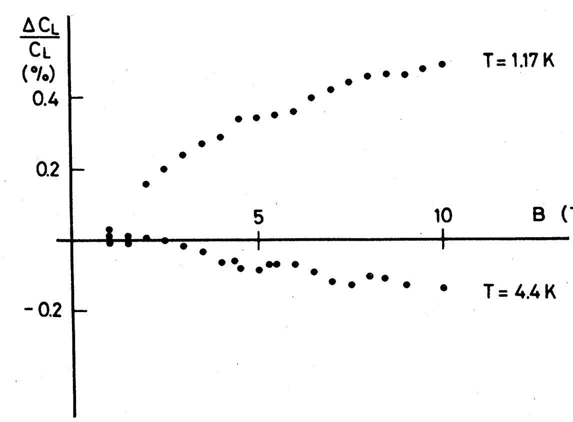

FIG. 3. Magnetic field dependence of the longitudinal elastic constant c_L for polycrystalline $CeAl_3$.

pletely new state develops which seems to be describable as a heavy Fermi liquid.[12] Our new elastic constant results fit well into the picture which has emerged already from the other physical properties listed above. Comparing our low-temperature elastic constants results with the magnetic susceptibility[3] in the same temperature region, one can give a rough empirical formula $c = c_0 - a\chi$ for $T < 4$ K.

Finally we show in Fig. 3 the magnetic field dependence of c_L for $T = 4.4$ and 1.15 K up to 10 tesla, indicating a rather strong magnetic field dependence for low temperatures. The magnetic field was applied along the propagation direction of the sound waves. The magnetic field effects are not so strong as the ones observed in $CeAl_2$ in the same temperature region.[13]

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[1]K. H. J. Buschow, H. J. van Daal, F. E. Maranzana, and P. B. van Aken, Phys. Rev. B 3, 1662 (1971).
[2]K. H. Mader and W. M. Swift, J. Phys. Chem. Solids 29, 1759 (1968).
[3]K. Andres, J. E. Graebner, and H. R. Ott, Phys. Rev. Lett. 35, 1779 (1975).
[4]M. Ribault, A. Benoit, J. Flouquet, and J. Palleau, J. Phys. 40, L-413 (1979).
[5]A. S. Edelstein, C. J. Tranchita, O. D. McMasters, and K. A. Gschneidner, Solid State Commun. 15, 81 (1974).
[6]B. Cornut and B. Qogblin, Phys. Rev. B 5, 4541 (1972).
[7]K. D. Schotte and U. Schotte, Phys. Lett. 55A, 38 (1975); G. Gruner and A. Zawadowski, Rep. Prog. Phys. 37, 1497 (1974).
[8]G. Binnig and H. E. Hoenig, J. Phys. (Paris) Colloq. 39, C6-1148 (1978).
[9]R. Takke, W. Assmus, B. Lüthi, T. Goto, and K. Andres, in *Crystalline Electric Field and Structural Effects in f-Electron Systems*, edited by J. E. Crow, R. P. Guertin, and T. W. Mihalisin (Plenum, New York, 1980), p. 321.
[10]B. Lüthi, in *Magnetism and Magnetic Materials—1976*, edited by J. J. Becker and G. H. Lander (AIP, New York, 1976).
[11]See, e.g., J. Schilling, Adv. Phys. 28, 657 (1979).
[12]M. T. Béal Monod (unpublished).
[13]T. Goto and B. Lüthi (unpublished).