Scaling behavior of the giant anisotropic magnetoresistance in thin films of the heavy-fermion compound $CeAl_3$

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(Received 12 August 1991; revised manuscript received 6 January 1992)

High-quality thin films of $CeAl_3$ with preferentially oriented crystal structure have been prepared, allowing unambiguous measurement of the orientation dependence of the magnetoresistance $\Delta\rho$: a very large anisotropy is observed. When a magnetic field is applied along the crystal c axis, a positive transverse $\Delta\rho$ is exhibited over a much larger region of H and T than is the case with bulk polycrystalline samples. Kondo scattering is diminished in this orientation when T is below 6 K. Two qualitatively different regimes are observed in which $\Delta\rho$ is described by universal functions of H/T.

Even before the discovery[1] of the enormous electronic coefficients of specific heat which characterize the heavy fermion compounds, $CeAl_3$ was under investigation due to its peculiar electronic properties. Polycrystalline bulk samples of $CeAl_3$ displayed highly anomalous and temperature-dependent resistivity;[2] a Curie-Weiss behavior of the magnetic susceptibility without magnetic ordering;[3-5] and a large variability of the resistivity with small doping levels.[6] More recent observations include a large positive peak in the Hall coefficient[7] at a temperature of 5–6 K; a peak in thermal conductivity[8] below 1 K; an enormous negative Grüneisen parameter;[9] and a change in the spin-lattice relaxation rate measured by NMR.[10] A positive magnetoresistance was observed at temperatures below 1 K.[5,11] The availability and quality of single crystals of $CeAl_3$ are restricted by the tendency of other phases to be present in the melt. The only published measurements on single crystals are the data by Jaccard *et al.*[12] on two submillimeter-sized specimens. To our knowledge, no measurements of quantum oscillations have been reported; thus the Fermi surface has not been mapped.

Here, we report our findings regarding the regime of positive magnetoresistance in $CeAl_3$ in very high magnetic fields, which were obtained using high-quality thin film samples. Some preliminary results have been reported.[13] The hexagonal crystal structure is found to have a preferential orientation in these films. Our consequent ability to resolve the effect of crystal orientation on transport phenomena led to the discovery that the regime of positive transverse magnetoresistance extends as high in temperature as 5.4 K. Fields up to 20 T are required to cause a decreasing resistivity at low temperatures. We examine the H/T dependence of the magnetoresistance and find two regions of qualitatively different behavior. We identify 6 K as the lower limit of temperature where the scattering behavior is similar to that predicted for noninteracting Kondo ions.

Thin films of 1100–2500 Å as-deposited thickness were prepared by electron beam coevaporation of Ce and Al onto Al_2O_3 substrates at ambient temperature. The intermetallic compound was formed by subsequent *in situ* annealing at 800–900 °C for several minutes. The properties of these films are identical to bulk $CeAl_3$ when appropriate comparison is made. The films displayed the resistivity-temperature behavior[2] of polycrystalline bulk $CeAl_3$, in particular the peak at 37 K and the knee[5] at 5.5 K. Only the $CeAl_3$ phase was observed in powder x-ray diffraction patterns. Resistivity was measured with standard four-terminal techniques. The magnetic susceptibility showed the Curie-Weiss behavior of bulk[5] $CeAl_3$, with no indication of a ferromagnetic transition at 6 K, indicating that no significant quantity of Ce_3Al_{11} was present.

X-ray diffraction measurements taken as a function of substrate orientation (i.e., x-ray pole figures) showed that the (001) plane was oriented almost exclusively parallel to the substrate plane, as occurs in many hexagonal systems when prepared in thin-film form. The resistivity was found[13] to be highly dependent on the orientation of the magnetic field with respect to the substrate or basal plane, while maintaining the transverse geometry. The heavy fermion compound $CeCu_6$ has similar anisotropy.[14] Another heavy fermion compound, $CePb_3$, develops a positive magnetoresistance at low temperatures,[15] which has been interpreted in terms of an antiferromagnetic state. In our measurements on $CeAl_3$ films, the anisotropy becomes pronounced below 6 K, and the positive magnetoresistance below 5.4 K with the magnetic field along the c axis. The specific-heat peak associated[15] with the onset of positive magnetoresistance in $CePb_3$ does not occur[9] in $CeAl_3$. $CeAl_3$ does not order magnetically at 6 K,[16,17] and displays only weak static magnetic correlations[18,19] below about 1.2 K. The latter may be associated with the small peak in magnetoresistance observed[11] in polycrystalline materials. This small peak is also observed in some of our thin films, but only with the magnetic field parallel to the basal plane.

The observed positive magnetoresistance in the thin-film samples cannot be ascribed to size effects arising from the thin-film geometry, as films of widely differing thicknesses prepared with preferential crystal orientation had quantitatively the same resistivity behavior. A film of similar quality (single phase in XRD) and thickness

but with multiple c-axis orientations, formed by altering the annealing conditions, showed positive magnetoresistance only below 2 K, similar to bulk polycrystalline samples, and had greatly reduced anisotropy. Nor does the giant positive magnetoresistance extending to 5.4 K appear to be related to a theory[20] based upon distributions of Kondo temperatures due to defects, as the data are the same for various film thicknesses, and are strongly temperature dependent.

Figure 1(a) shows the transverse resistivity $\rho_\perp(H,T)$ of $CeAl_3$ with the current **j** in the basal plane, and with **H** along the c axis. Negative initial magnetoresistance disappears below 5.4 K. Figure 1(b) shows the transverse resistivity $\rho_\parallel(H,T)$ with **H** perpendicular to **j**, and both in the basal plane. Initial magnetoresistance remains negative down to about 2 K. When **H** is along the c axis, two regions are found in which the dimensionless magnetoresistance $\Delta\rho_\perp=[\rho_\perp(H,T)-\rho(0,T)]/\rho(0,T)$ follows qualitatively different functions of H/T. These regions are enclosed by dot-dashed lines and labeled A and B in Fig. 1(a). The maxima in resistivity with the field along the c axis describe a parabolic curve in the H-T plane.[13]

The scaling behavior of $\Delta\rho_\perp$ is shown in Fig. 2. In region A, $\Delta\rho$ is almost isotropic, and scales with H/T at all fields, and for temperatures down to 6.0 K [Fig. 2(a)]. At 5.6 K, departure from the high temperature H/T behavior is marked. Kohler's rule,[21] that is, scaling with $H/\rho(0,T)$, is not obeyed in region A [Fig. 2(b)]. This is not surprising if resistivity in region A is a result of Kondo scattering. The scattering phase shift, and hence the scattering rate, is expected[22] to be a function of field in Kondo scattering, whereas Kohler's rule will be obeyed if the scattering rate is field independent. The resistivity in this region is qualitatively similar to a prediction[22] for scattering from noninteracting spin-$\frac{1}{2}$ Kondo ions. The sudden failure of scaling below 6 K indicates that the energy scale which controls the scattering is changing. Apparently, the noninteracting Kondo ion description fails below 6 K. H/T scaling has been observed in dilute Kondo systems.[23]

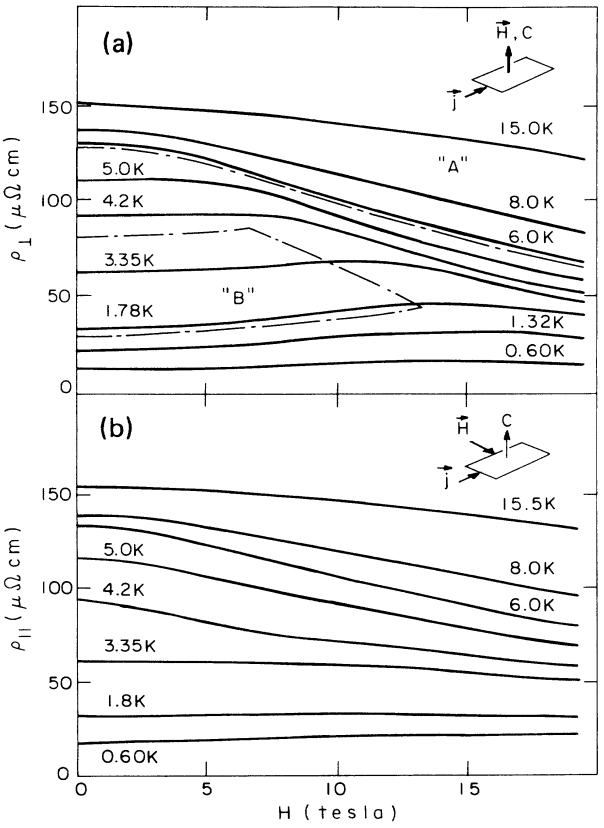

FIG. 1. Resistivity of $CeAl_3$ in a magnetic field with current in the basal plane. (a) Magnetic field perpendicular to basal plane; (b) magnetic field in basal plane perpendicular to current. The regions in which scaling is observed are delineated by —·—·—· lines.

In region B, which extends only up to 3.8 K, $\Delta\rho_\perp$ is positive and displays H/T scaling [Fig. 2(c)], and also $H/\rho(0,T)$ scaling [Fig. 2(d)]. The data for **H** in the basal plane, $\Delta\rho_\parallel$, is negative and does not scale. Thus the magnetoresistivity is both anisotropic and of opposite sign. Although the scaling trajectory in Fig. 2(d) is slightly curved, the fact that magnetoresistance scales with $H/\rho(0,T)$ is consistent with a field-independent mean-free path. Ordinary metals deviate from pure H^2 magnetoresistance as saturation occurs at high fields, while still obeying $H/\rho(0,T)$ scaling. Furthermore, in region B, the longitudinal magnetoresistance (not shown) is equal to the transverse in-plane magnetoresistance $\Delta\rho_\parallel$ to within experimental error.

Some aspects of the data suggest that an electronic phase transition is occurring at 6.0 K. The spatial symmetry of the scattering in the presence of a magnetic field is broken at that temperature. In region B, the scattering behavior has the axial symmetry of the hexagonal crystal lattice of $CeAl_3$. The occurrence of H/T scaling in regions A and B, with opposite signs of magnetoresistance, show that qualitatively different behaviors, each with its own characteristic energy scale, occur in these regions. The absence of a sharp peak[9] in the specific heat at 6 K would imply that the transition is of higher than second order. A second transition may occur at the low temperature end of region B, that is, at 1.7 K. It is at this temperature that $\Delta\rho_\parallel$ changes sign, and below which weak static magnetic correlations have been reported.[18,19]

The magnitude of magnetoresistance in region B is quite large for a metal of such high resistivity. Since the longitudinal magnetoresistance was measured in region B and found to be identical to the in-plane transverse magnetoresistance $\Delta\rho_\parallel$, the negative sign of $\Delta\rho_\parallel$ is apparently due to a spin effect, as it is isotropic, whereas the positive sign of $\Delta\rho_\perp$ is apparently an orbital effect.

The appearance of positive magnetoresistance below 6 K may be related to the onset of some type of coherence among the Kondo ions, an idea which has been broadly invoked to explain heavy fermion behavior. The breaking of spatial symmetry at 6.0 K followed by a second transition at 1.7 K is consistent with the Ce-Ce spacing in the hexagonal $CeAl_3$ crystal lattice. Within the basal plane, the Ce-Ce spacing is smaller than the out-of-plane spacing. This would lead to a stronger Ce-Ce coupling, and hence a higher transition temperature to a state with axial symmetry. The out-of-plane Ce-Ce interaction

would occur at a lower temperature, and produce a less axial (although not necessarily isotropic) state, as is observed. In addition to the magnetoresistance data, it is noted that many of the anomalous transport and magnetic properties of $CeAl_3$ undergo qualitative changes at a temperature of about 6 K, including the following.

(1) The inverse magnetic susceptibility, whose slope changes there, but whose overall form remains Curie-Weiss-like.[5] The increase in slope of $1/\chi$ as temperature falls indicates a lowering of the Ce moments. This may be related to the onset of anisotropy and positive magnetoresistance.

(2) The zero-field resistivity, whose slope changes there.[5] Furthermore, a knee at 5–6 K is not unique to

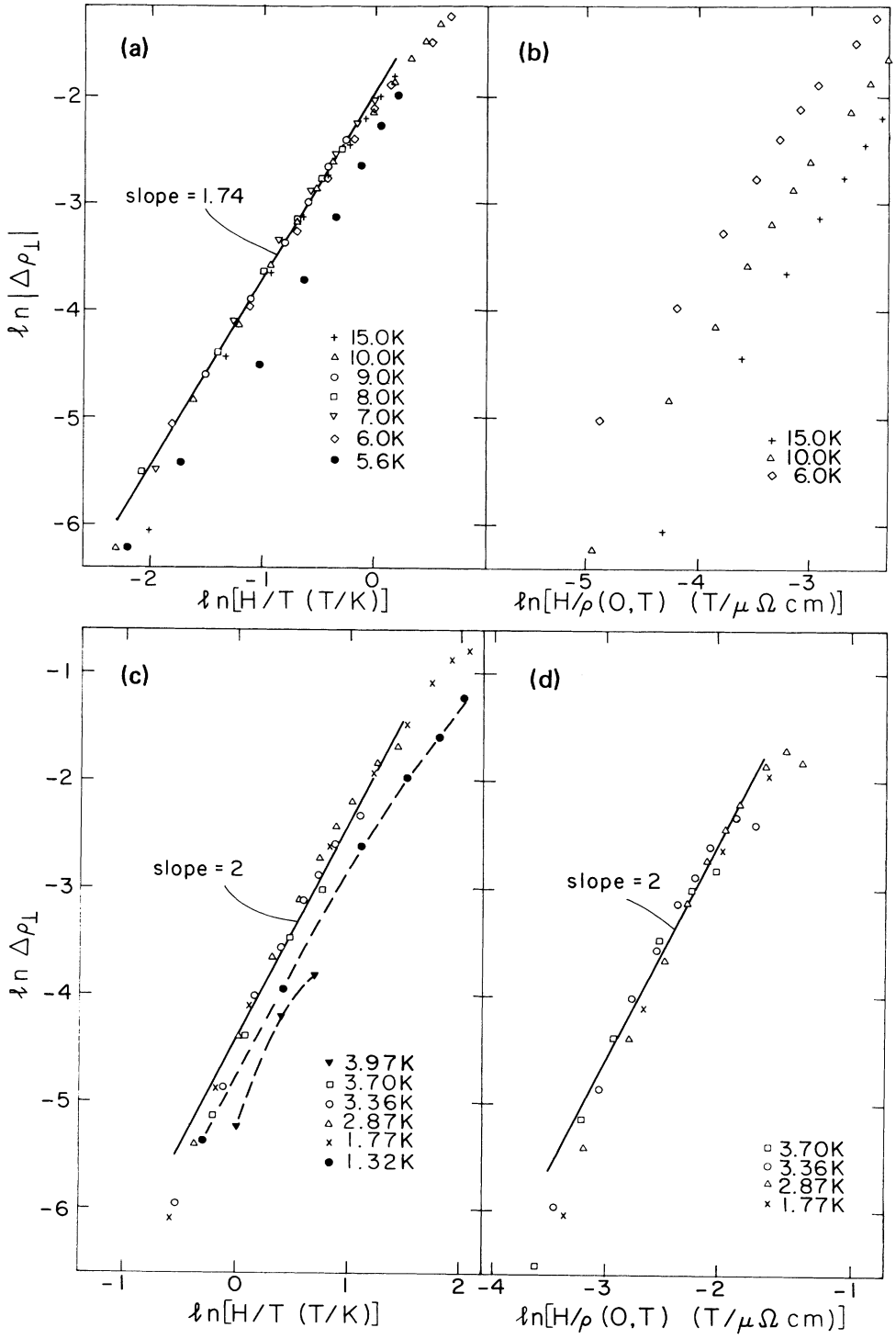

FIG. 2. Scaling behavior of magnetoresistance of $CeAl_3$ with **j** parallel to, and **H** perpendicular to, basal plane: (a) H/T scaling in region A; (b) failure of Kohler's rule in region A; (c) H/T scaling in region B; (d) Kohler's rule scaling in region B.

polycrystalline $CeAl_3$, but appears also[2,6] in pseudobinary compounds with La, Th, or Y substituted for Ce, in the neighboring phases $CeAl_2$ (Ref. 24) and Ce_3Al_{11},[25] and in our own thin films where stoichiometry and annealing conditions varied from the optimum. The alloying studies and our nonoptimum films both show clearly that, whereas features in the resistivity vary widely above 6 K, the knee at 6 K is ubiquitous. (One study of a small $CeAl_3$ single crystal[12] found the knee at 6 K to be more rounded, and saw a sharper knee at 1.5 K. Our thin-film samples are polycrystalline rather than single crystals.)

(3) The Hall coefficient, which deviates[7] from the predictions of skew-scattering theory[26] there. Coleman, Anderson, and Ramakrishnan[27] have asserted that the formation of a heavy quasiparticle band is expected in Kondo-lattice compounds, and that it will significantly influence the measurement of the Hall coefficient.

(4) The resistivity at low temperatures also deviates[5] from the theory of Cornut and Coqblin,[28] which is based on Kondo scattering in compounds with Ce taking account of the crystalline electric field. In a lattice, it is clear that such a theory must break down at a sufficiently low temperature, since the periodicity of the potential and the absence of inelastic scattering must result in zero resistivity at zero temperature. The discrepancy in fact appears to develop at about 6 K. The large resistivity peak of $CeAl_3$ at 37 K is well explained by the Cornut-Coqblin theory, and is observed experimentally to be sensitive to the crystalline environment, as mentioned earlier.

It is clear from our data that the scattering behavior alters radically when the temperature of $CeAl_3$ is reduced below 6 K. The scattering Hamiltonian of the carriers must be quite different from that for higher temperatures. The large anisotropy in magnetoresistance could be due either to mass anisotropy or to an anisotropy in Kondo scattering cross section; in the latter case, a near-zero cross section for carrier momenta parallel to the basal plane would be required to explain the observed behavior. The formation of carriers which are insensitive to Kondo scattering would be consistent with the observed departure from the theory of Cornut and Coqblin. The ubiquity of the resistivity knee could mean that the conditions for the occurrence of the transition at 6 K are relatively insensitive to the crystalline environment.

Since the discovery of the heavy-fermion phenomenon, attempts have been made to model a band of heavy carriers using standard band-structure calculations and renormalized band calculations.[29] It is interesting to consider whether the observed behavior reflects in some way the existence of heavy-mass carriers, which appear by specific heat measurements[1,9] to be forming in the same temperature range. Although we cannot, from our measurements alone, exclude the possibility that the magnetoresistance is unrelated to the specific heat enhancement, the mundane explanation would be that the carriers which furnish the dominant contribution to the magnetoresistance below 6 K are also responsible for the enhancement of the specific heat coefficient.

As the magnetic field is increased above the limit of H/T scaling in region B, the resistivity develops a maximum and then decreases. It is possible that the reappearance of negative magnetoresistance means that the electron gas reverts to the Kondo state in high field; however, high-field specific-heat measurements[30] imply that heavy-fermion behavior persists in fields of 23 T. It may be instead that a second band of carriers, whose scattering by the Kondo mechanism is suppressed by the field, accommodates a larger fraction of the current density as the field intensity is increased.

It is interesting to note that the change of resistivity with temperature at zero field is about three times larger than the change of resistivity with temperature at a constant high field, e.g., 20 T. This may be important in constructing a microscopic model of these phenomena.

In summary, careful measurements of the magnetoresistance of thin films of $CeAl_3$ have revealed a remarkable behavior of the carriers at temperatures below 6 K. Anisotropy and scaling behavior have been determined. A model in which carriers with different transport properties form below 6 K may account for the data, and it has been discussed in light of several other unusual properties of this material. It remains to be determined what aspects of the magnetoresistance behavior of $CeAl_3$ are common to the entire family of materials known as heavy-fermion compounds.

We would like to acknowledge helpful discussions with B. L. Alt'shuler, A. S. Edelstein, P. A. Lee, R. Meservey, J. S. Moodera, and P. Schlottmann. The Francis Bitter National Magnet Laboratory is operated for the National Science Foundation by the Massachusetts Institute of Technology. This work was performed under NSF Grant No. DMR-8618072. One of us (G.M.R.) acknowledges the support of the Office of Naval Research at the beginning stages of this work.

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