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Resistivity and magnetoresistance of CeAl_3 single crystals

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In honor of Martin Peter's 60th birthday.

Abstract. We discuss the anisotropic resistivity of single crystalline CeAl_3 . At low temperature, both the resistivity and the magnetoresistance provide evidence that some kind of magnetic order starts to develop at 1.6 K. At very low temperature, the residual resistivity rapidly decreases as temperature increases with approximately the same characteristic temperature which governs the pure Kondo lattice resistivity.

Introduction

Among the heavy fermion compounds (HFC), CeAl_3 is one of the most studied. Its ground state was characterized as normal conducting and paramagnetic [1] and it was considered as archetype of non magnetic HFC. However, the peritectoidic formation of this compound at 1135 K prevents the growth of the large single crystals necessary for studying carefully the nature of the ground state. To overcome this difficulty we developed an experimental set up for transport properties measurements on the very small single crystals one can extract from an annealed polycrystalline ingot. At the same time we started to anneal different Ce–Al alloys under very high vacuum in order to obtain the largest single crystalline grains by recrystallization. The temperature dependence of the resistivity was measured on two samples with current parallel or perpendicular to the hexagonal c -axis. Longitudinal and transverse magnetoresistances were also measured. Details about the samples and the measurement technique can be found in Ref. 2.

Resistivity

Figure 1 shows the temperature dependence of the resistivity of a CeAl_3 single crystal with the current I parallel or perpendicular to the hexagonal c -axis. Along both directions the resistivity exhibits a maximum around 35 K. From these

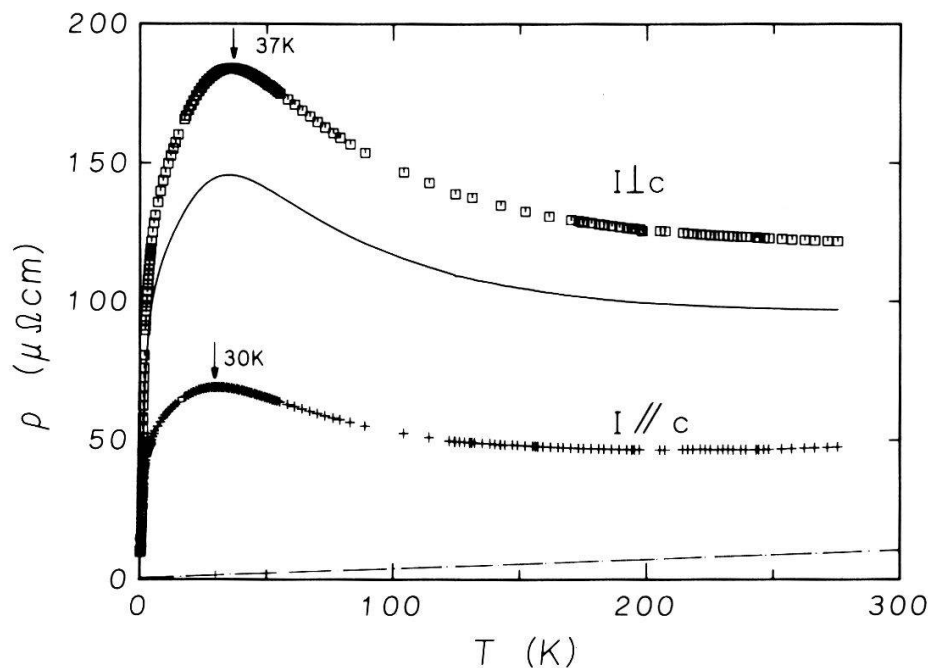


Figure 1

Temperature dependence of the resistivity of CeAl_3 single crystals with the current parallel or perpendicular to the c -axis. Solid line: calculated resistivity $\rho = \frac{1}{3} (\rho_{\parallel} + 2\rho_{\perp})$ of a polycrystal. Dotted-dashed line: normal phonon contribution estimated from LuAl_3 measurements [5].

results one can calculate the polycrystalline resistivity curve $\rho = \frac{1}{3} (\rho_{\parallel} + 2\rho_{\perp})$ shown as a solid line on the figure, in very good agreement with the previous experimental results of Ref. 3.

The $\ln T$ variation of the resistivity, often considered as a signature of single ion Kondo coupling, is followed here in a rather narrow temperature range ($50 \leq T \leq 90$ K). In fact, as pointed out in Ref. 4, a better fit of our data can be obtained up to room temperature according to an empirical law $\rho = aT + bT^{-1/2}$, but the phonon term aT is larger than that extracted from the resistivity measurements of polycrystalline isostructural LuAl_3 [5].

Figure 1 also shows that ρ_{\perp} is larger than ρ_{\parallel} , indicating a stronger scattering in the basal plane than along the c -axis. This agrees with single ion Kondo theory for a highly anisotropic ground state of the 4f electron [6, 7]. In such theory the resistivity along the c -axis is small if the crystal electric field (CEF) ground state is a doublet constructed from the $|J_z = \pm \frac{5}{2}\rangle$ and/or $|J_z = \pm \frac{3}{2}\rangle$ states and the CEF splitting Δ is large. For infinite Δ the parallel resistivity is even zero. However, neutron scattering experiments [8, 9] indicate an overall CEF splitting $\Delta \cong 90$ K and consequently the anisotropy of ρ should strongly decrease for $T > \Delta$ as observed for instance in CeCu_2Si_2 [10].

So, the most intriguing feature in Fig. 1 is the nearly temperature independent ratio $\rho_{\perp}/\rho_{\parallel} \cong 3$ for $T > 10$ K. This probably originates in the anisotropy of the coupling constant J of 4f electrons with the Fermi sea, not taken into account by the theory.

It can be seen in Fig. 2 that for $T < 4$ K the anisotropy of ρ_{\perp} and ρ_{\parallel} rapidly decreases and that, below 0.6 K, ρ_{\parallel} even becomes larger than ρ_{\perp} . The break of

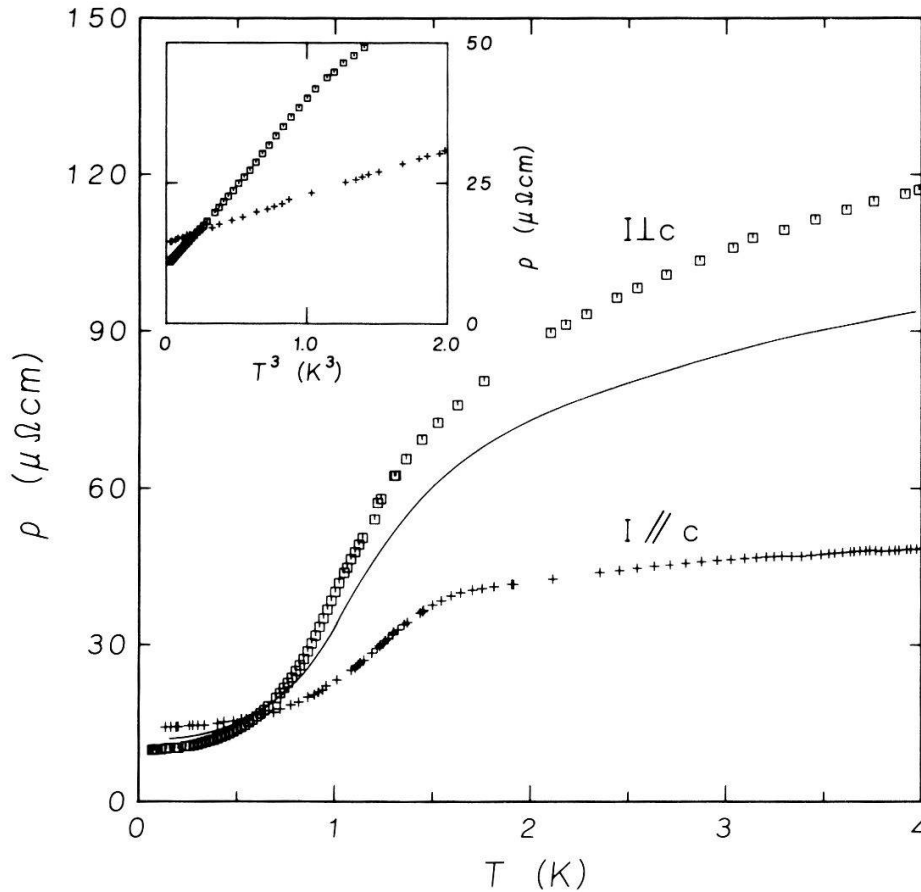


Figure 2

Low temperature dependence of the resistivity of CeAl_3 single crystals with the current parallel or perpendicular to the c -axis. The insert shows the T^3 variation. Solid line: see Fig. 1.

ρ_{\parallel} at 1.6 K with no corresponding anomaly in ρ_{\perp} is rather unexpected. Recent muon spin resonance experiments [11] seem to indicate at nearly the same temperature ($T = 1.5$ K) the development of short range magnetic correlations between the 4f electron moments. Also surprising for a non magnetic HFC are the T^3 variations of ρ_{\perp} and ρ_{\parallel} for $0.4 < T < 1$ K (insert of Fig. 2).

Usually, in this temperature range, the resistivity of HFC, such as CeCu_2Si_2 , CeCu_6 or polycrystalline CeAl_3 , shows approximate linear (or even less rapid) T dependence. T^n power laws with $n > 2$ are reminiscent of electron magnon scattering and have been observed in antiferromagnetic ordered Kondo compounds such as CeAl_2 ($n = 3$) [12, 13] and CeB_6 ($n = 3.5$) [14].

Below 0.35 K, the resistivity ρ_{\perp} and ρ_{\parallel} both recover the standard law $\rho = \rho_0 + AT^2$ expected for a strongly interacting Fermi liquid but with much higher residual resistivity values ρ_0 and lower A coefficients than in polycrystalline samples (see Table 1). In spite of these discrepancies, the temperature range where the resistivity follows a T^2 law is the same for both single and polycrystal. In fact, the disagreement between the calculated resistivity $\rho_{\text{poly}}^{\text{cal}} = \frac{1}{3}(\rho_{\parallel} + 2\rho_{\perp})$ of a polycrystal and the experimental values $\rho_{\text{poly}}^{\text{exp}}$ is large only for $T < 1$ K. Therefore, it appears that the difference $\Delta\rho_{\text{imp}} = \rho_{\text{poly}}^{\text{cal}} - \rho_{\text{poly}}^{\text{exp}}$, identified as an impurity contribution, decreases as T^2 up to 0.35 K. From Table 1 this

Table 1

Residual resistivity ρ_0 , quadratic A and cubic B temperature coefficients of the resistivity for $CeAl_3$ single crystals and various polycrystals.

	ρ_0 ($\mu\Omega$ cm)	A ($\mu\Omega$ cm/K ²)	B (Ω cm/K ³)
$I \perp c$	9.9	13.3	30
$I \parallel c$	14.5	4.6	8
polyc (15)	2.0	32.0	—
polyc (15)	5.0	30	—
polyc (1)	0.7	35	—
polyc (14)	3.8	29	—

impurity contribution can be written for $T < 0.35$ K as

$$\Delta\rho_{\text{imp}} \cong (10 - 20T^2)\mu\Omega \text{ cm}$$

or put in another form

$$\Delta\rho \cong 10\{1 - [T/(0.7 \text{ K})]^2\}\mu\Omega \text{ cm} \quad (1)$$

to underline the impurity scale temperature T_i of 0.7 K.

A similar behavior has been observed in $(Ce, La)Pd_3$ but with a higher scale temperature of about 30 K and has been interpreted by scattering of heavy quasiparticles from Kondo hole when the system is in its coherent regime [16]. However, apart from the resistivity, the other physical properties (magnetic susceptibility, specific heat) do not show clear evidence of the Kondo hole mechanism.

Another interpretation has been given by Fetisov and Khomskii [17] assuming that impurities induce scattering from the light d band to the heavy f band in complete analogy with the two band models for d transition metals. In this scheme the impurity resistivity takes the form

$$\rho_{\text{imp}} = \rho_0[1 - (\pi^2/3)(T/T_f)^2] \quad (2)$$

where T_f is the scale temperature of the pure system. To evaluate T_f from the T^2 law of pure $CeAl_3$, we note that the resistivity should reach its saturation value ρ_s as $\rho(T) \cong \rho_s(T/T_f)^2$. Taking $\rho(T_{\text{max}}) = 150 \mu\Omega \text{ cm}$ as an estimate of ρ_s , we obtain $T_f \cong 2.3$ K. From equation (1) and (2), the relation between the scale temperatures T_i and T_f of the impurity and pure systems is quite natural.

Furthermore, according to Fetisov and Khomskii, the second term in (2) renormalizes the coefficient A_p of the T^2 law of the pure lattice. So the experimental term is

$$A_{\text{exp}} = A_p - (\pi^2/3)(\rho_0/T_f^2)$$

This nicely agrees with the values listed in Table 1. Figure 3 shows that A_{exp} , as a function of ρ_0 , fits a straight line which slope gives $T_f \cong 1.2$ K. Evidently, $A_p \cong 37 \mu\Omega \text{ cm/K}^2$ can also be extrapolated as $\rho_0 \rightarrow 0$.

Finally, we want to note that the impurity resistivity should be associated with a positive contribution of the thermoelectric power located at a temperature

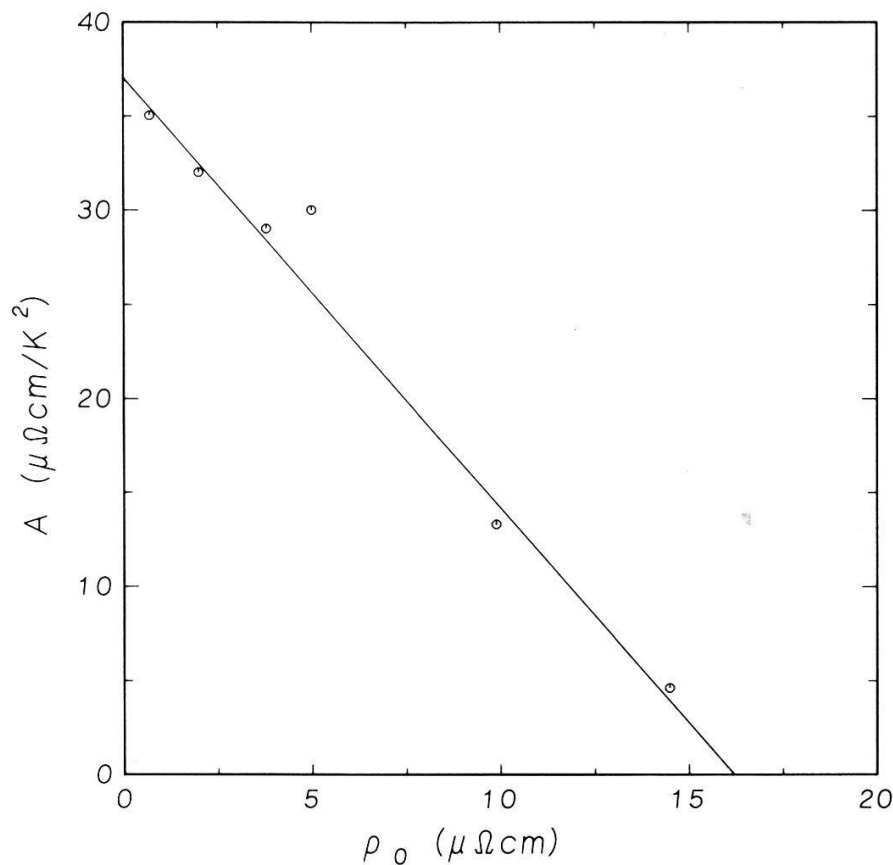


Figure 3

Values listed in Table 1 of the coefficient A in the law $\rho = \rho_0 + AT^2$ as a function of the residual resistivity ρ_0 .

somewhat below T_f as observed in CeAl_3 [15] and CePd_3 [18]. This mechanism also explains why the largest thermoelectric power has been measured for samples with high residual resistivity values in apparent contradiction to the Nordheim–Gorter rule [15].

Magnetoresistance

Let us now discuss the magnetic field dependence of the low temperature resistivity. Results for $I \parallel c$ and $H \parallel c$ are reported in Fig. 4.

Above 2 K, the magnetoresistance $\Delta\rho/\rho$ is negative for H parallel to c whatever the direction of the current (\parallel or $\perp c$), whereas $\Delta\rho/\rho$ is positive but very weak for $H \perp c$. Usually, the negative magnetoresistance is characteristic for the presence of local magnetic moments [19]. The anisotropic magnetic field response of the resistivity agrees with predictions of Ref. 6 and confirms the doublet $|J_z = \pm 3/2\rangle$ ground state in CeAl_3 .

At very low temperature, the resistivity is dominated by its residual value ρ_0 . It can be seen in Fig. 4 that the magnetoresistance for $I \parallel c$ and $H \parallel c$ is positive up to 8 T and exhibits a large maximum at a field $H_{\text{max}} \cong 2.2$ T.

In polycrystalline samples similar results have been observed with the same field H_{max} but with $\Delta\rho/\rho$ values lower by a factor of about 4 [15, 20, 21]. This

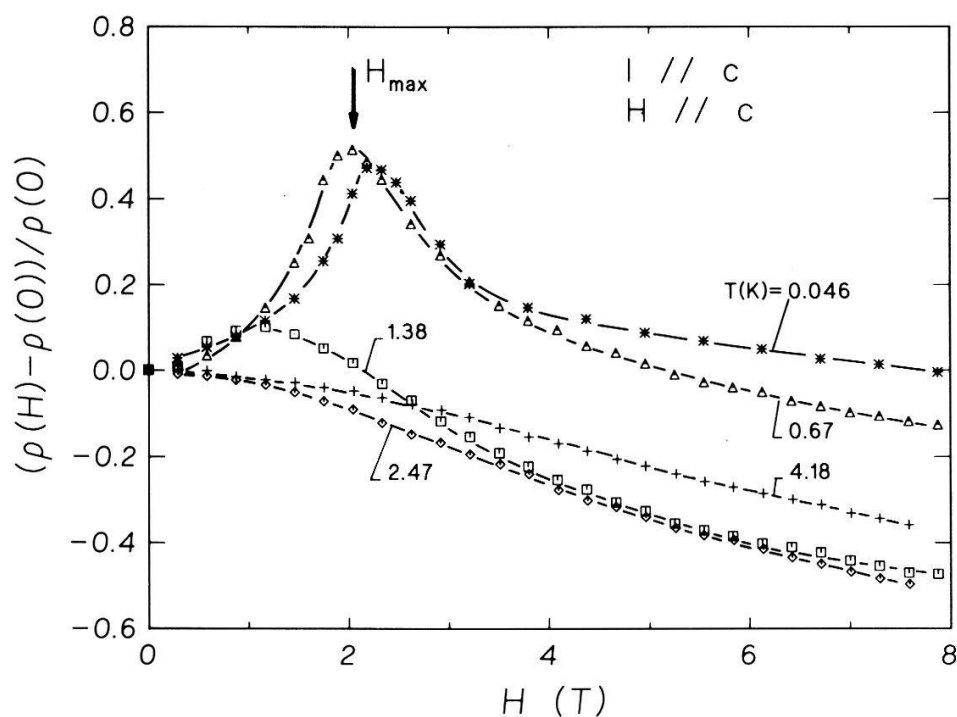


Figure 4

Longitudinal magnetoresistance along the c -axis of CeAl_3 at different low temperatures.

factor roughly corresponds to the reduction expected from random projections of an hexagonal magnetoresistance tensor (preliminary measurements indicate that $\Delta\rho/\rho$ is large only for $I \parallel c$ and $H \parallel c$).

As the residual resistivity ρ_0 is much higher in single crystals than in polycrystals, one concludes that the very low temperature magnetoresistance is independent of ρ_0 , i.e. dominated by its impurity part. Such a behavior agrees

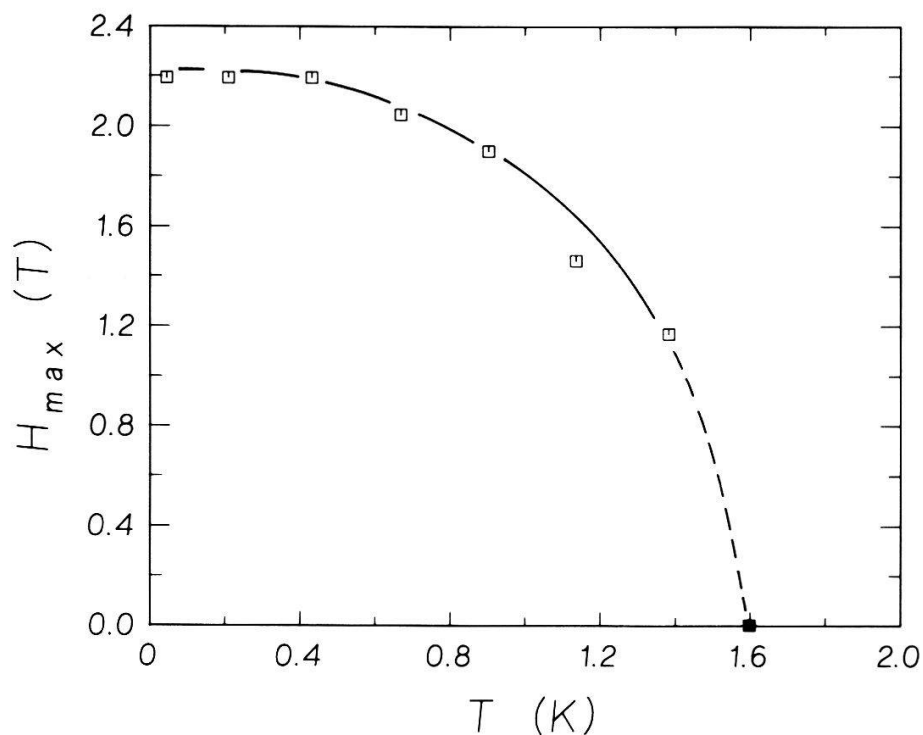


Figure 5

Magnetic phase diagram deduced from the magnetoresistance measurements. H_{\max} is defined in Fig. 3.

with the above-mentioned two band model [17] and the maximum of $\Delta\rho/\rho$ can be interpreted assuming the Fermi level is located in a pseudogap of the density of heavy states.

In fact, the positive magnetoresistance for I and $H \parallel c$ can be observed up to 1.6 K, the temperature at which a break occurs in $\rho_{\parallel}(T)$ (Fig. 2). Figure 5 shows the temperature variation of the crossover field H_{\max} . Such a plot is strongly analogous to (H, T) phase diagrams of magnetic systems and then suggests some 'magnetic' phase in CeAl_3 . An appealing possibility is a magnetic order with very weak moments. Hence, the above-mentioned pseudogap in the density of heavy states which has been often presented as a characteristic of the Kondo resonance in the coherent state of HFC, might have a more conventional magnetic origin in CeAl_3 .

Conclusion

To conclude we summarize our main results:

- the large anisotropy of the resistivity at temperatures higher than the CEF splitting underlines the anisotropy of the mixing interaction in the CeAl_3
- the break of ρ_{\parallel} at 1.6 K, the T^3 dependence of ρ_{\parallel} and ρ_{\perp} at lower temperature and the (H, T) phase diagram deduced from the magnetoresistance measurements give evidence of some kind of magnetic transition
- at very low temperature impurity scattering plays an essential role in transport properties.

The origin of the large residual resistivity of the single crystals is still not established. Chemical impurities can be ruled out since both single crystalline and polycrystalline samples were prepared from the same starting materials. Measurements of samples with small variations of composition have shown the insensibility of the residual resistance to stoichiometric deviations. Since the single crystalline grains result from recrystallization and are composed of nearly oriented small crystallites, stresses might be important in our single crystals. This possibility has to be studied further.

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