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TRANSPORT PROPERTIES OF HEAVY ELECTRON METALS

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Abstract. – Resistivity (ρ) and thermoelectric power (Q) experiments are described for two different ground states at zero pressure (P): a normal phase (CeAl_3) and a superconducting phase (UBe_{13}). In CeAl_3 , antiferromagnetism occurs clearly at $P = 0$ and may disappear rapidly under pressure. Magnetic field and pressure experiments in UBe_{13} show that antiferromagnetism and superconductivity may be antagonist.

1. Introduction.

Recent measurements in heavy fermion metals underline the importance of the magnetism. For example, the previous classification of the ground state as a normal Pauli paramagnetic phase in CeAl_3 or a superconducting and non magnetic phase in CeCu_2Si_2 , is now revised; both of these compounds seem to undergo an antiferromagnetic order with a tiny ordered moment suggesting itinerant magnetism [1]. Indeed most heavy fermion superconductors (URu_2Si_2 , CeCu_2Si_2 , UPt_3 , $\text{U}_{1-x}\text{Th}_x\text{Be}_{13}$ with $x < 0.015$) [2-4] are now considered as antiferromagnetic. Inelastic neutron measurements on (yet assumed heavy fermion paramagnets), CeCu_6 and CeRu_2Si_2 show the coexistence of antiferromagnetic correlations and strong local spin fluctuations. Their relative weights may be monitored by applied magnetic field (H) or pressure (P) [5, 6].

Low ordered magnetic moments have been mainly detected by muon spin relaxation (μSR) [4], nuclear magnetic resonance (NMR) [2] or neutron scattering experiments when large single crystals can be realized as for UPt_3 [3]. Here, focus will be given on transport measurements realized on a non superconducting (CeAl_3) and a superconducting (UBe_{13}) compound. The reported magnetoresistivity data are obtained on very small single crystals of CeAl_3 . Details about samples and experimental techniques can be found in reference [7-9]. Measurements on UBe_{13} were performed down to 50 mK in magnetic field up to 10 T at very high pressure ($P \leq 80$ kbar) generated by a pair of opposed Bridgman anvils made of non magnetic tungsten carbide. Due to the large value of the upper critical field H_{c2} ($T = 0$) ~ 13.9 T of UBe_{13} the high magnetic fields of the Service National des Champs Intenses and the Max-Planck Institut of Grenoble were used to study the resistivity (ρ) of the normal phase [10].

CeAl_3 : an antiferromagnet with tiny ordered magnetic moments?

μSR experiments down to 35 mK lead to the observation of a spontaneous muon spin precession frequency in zero external field below 0.7 K and of a non oscillatory component which collapses only above 1.5 K. Clearly, a quasistatic internal magnetic field of finite average value occurs below 0.7 K and below 1.5 K magnetic correlations seem to set up [11].

In the same time [7], resistivity measurements on single crystals of CeAl_3 show unusual behaviours not yet detected on polycrystals. From 300 K to 10 K, ρ_{\parallel} and ρ_{\perp} i.e. parallel and perpendicular to the c -axis of the hexagonal structure, exhibit a constant large anisotropy with a ratio $\rho_{\perp}/\rho_{\parallel} \sim 3$. The average ($\langle \rho \rangle = 1/3 (\rho_{\parallel} + 2\rho_{\perp})$) is in excellent agreement with experimental data obtained for polycrystalline samples made from the same starting materials. The weak temperature dependence of $\rho_{\perp}/\rho_{\parallel}$ well above the CF splitting ($\Delta \sim 90$ K) indicates that the anisotropy of the mixing interaction must be taken into account in theoretical models.

At low temperature, the resistivity of the single crystals differs markedly from that of polycrystals. Evidences of magnetic order have been found i) at 1.6 K where ρ_{\parallel} shows a break while no anomaly occurs in ρ_{\perp} and ii) for $0.4 < T < 1$ K, where both ρ_{\parallel} and ρ_{\perp} have T^3 dependences. T^n power laws with $n > 2$ are reminiscent of electron-magnon scattering; for example, they are observed in the antiferromagnetic ordered Kondo compound CeAl_2 [12]. In this temperature range, the polycrystalline samples have a quasi-linear T resistivity [13].

The obtention of single crystals gives the possibility of significant magnetoresistivity experiments as realized on CeRu_2Si_2 [14]. The amplitude of the magnetoresistivity is coupled to that of the magnetization

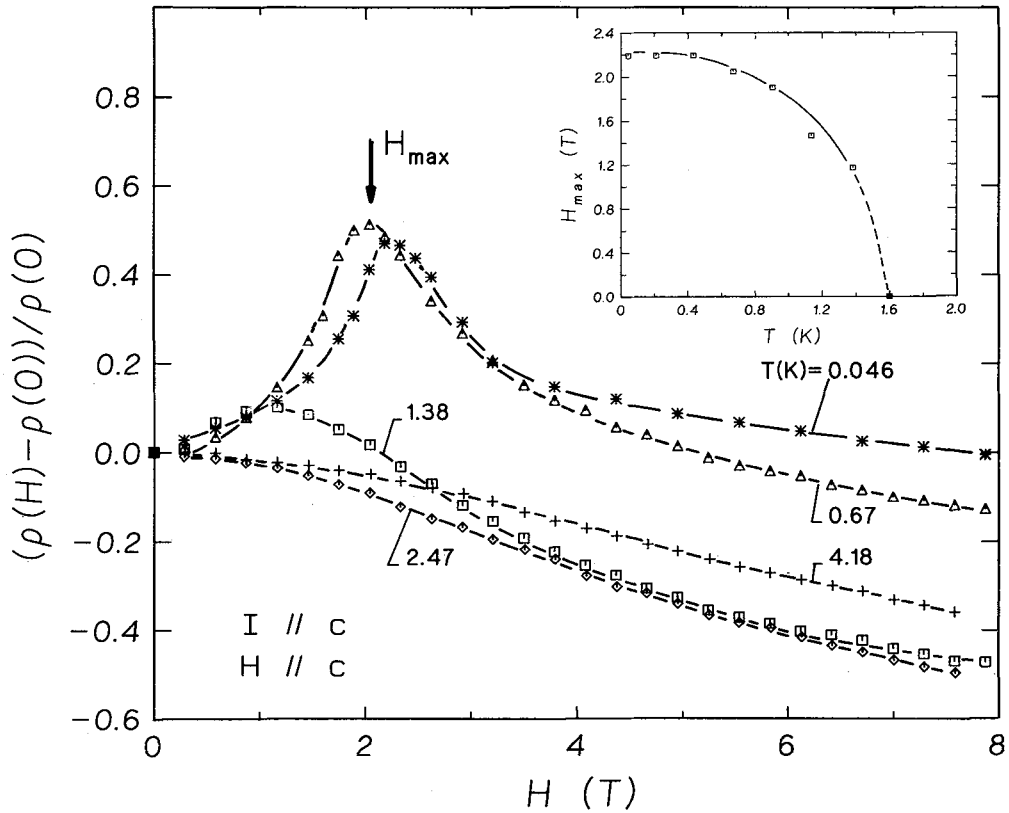


Fig. 1. - Longitudinal magnetoresistance along the *c* axis of CeAl₃ at different low temperatures. Insert is the magnetic phase diagram deduced from the temperature dependence of H_{max} , the maximum of the isothermal magnetoresistivity.

which is induced by the magnetic field. In agreement with the observation that the *c*-axis is the easy magnetization axis, the magnetoresistivity is large only when the magnetic field is parallel to the *c*-axis. As shown in figure 1, below 1.6 K, an initial positive magnetoresistivity appears up to a magnetic field H_{max} . The temperature dependence of H_{max} shown in the insert is strongly analogous to H, T phase diagrams of magnetic systems.

Below 0.35 K, $\rho_{||}$ and ρ_{\perp} follow the standard law $\rho(H) = \rho_0(H) + A(H)T^2$, with $\rho_{0||} = 14.5 \mu\Omega\text{cm}$, $\rho_{0\perp} = 9.9 \mu\Omega\text{cm}$, and $A_{||} = 4.6 \mu\Omega\text{cm}/\text{K}^2$, $A_{\perp} = 12.8 \mu\Omega\text{cmK}^{-2}$. Figure 2 shows that the magnetic field dependence of $A_{||}(H)$ has a narrow peak at $H \sim 2$ T i.e. nearly at the crossover field H_{max} . Such a coincidence is found at the metamagnetic like transition of CeRu₂Si₂ [14]. It is worthwhile to mention that non linearity of the magnetization with H was already reported in the pionnering work of reference [15]. In polycrystals, field maxima of $\Delta\rho/\rho_0(H)$ and $A(H)$ have been reported with the same field H_{max} but with lower magnitude corresponding roughly to the random projection of an hexagonal magnetoresistance tensor. The occurrence of maxima in the magnetoresistivity

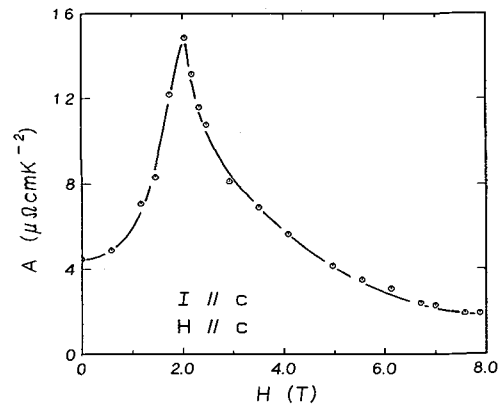


Fig. 2. - Magnetic field dependence of the $A_{||}(H)$ coefficient in the law $\rho_{||} = \rho_{0||} + A_{||}T^2$ of CeAl₃.

are predicted for anisotropic weakly antiferromagnets [16]. Evidence of magnetic order is also given by the NMR observation that the NMR linewidth increases markedly below 1.2 K and the nuclear spin lattice relaxation rate $1/T_1$ exhibits a sharp maximum also at

1.2 K [2, 17]. The initial increase of $A(H=0)$ with P in contrast to the rapid decrease with P above 3 kbar is interpreted as a transition from an antiferromagnetic lattice to a Pauli paramagnet [18].

The values of ρ_0 obtained in these crystals are far higher than those reached in polycrystals ($\rho_0 \leq 2 \mu\Omega\text{cm}$ [15-18]). For $\rho_0 \leq 2 \mu\Omega\text{cm}$, quasiconstant determinations of $A = 33 \mu\Omega\text{cmK}^{-2}$ are reported in polycrystals [15-18]. The large dependence of A with ρ_0 (comparison between polycrystals and single crystals) suggests that A cannot be entirely attributed to intrinsic interactions between quasiparticles built in a perfect lattice. An hypothesis to explain this unusual interference between ρ_0 and A is that impurities behave like "Kondo holes" highly sensitive to any change of the Kondo lattice [8]. As it is yet difficult to control the perfection of the lattice, one way to explore the interplay between lattice and impurities is to probe their relative sensitivity to the pressure (P) variable. Extensive P resistivity experiments [18] show that large variations of $A(P)$ are also attended by a strong P variation of ρ_0 . Magnetic field or pressure dependence of the residual resistivity reflects the microscopic change in the interactions between the quasiparticles of the CeAl_3 lattice.

If this notion of "Kondo holes" is widely used in heavy Fermion systems particularly to discuss their superconducting properties at low temperature [19-21], few theoretical works [22, 23] concern the normal state. The strong correlations between ρ_0 and A lead to ask the crucial question of the influence of disorder on the nature of the ground state. The observation of tiny ordered moments supports a picture where f electrons are itinerant so that their physical properties (for example connected with nesting) may depend strongly on doping. As the occurrence of tiny ordered moments seems just now established, there is yet no systematic work which may give a clear answer. For CeAl_3 , as similar field dependences of $A(H)$ and $\rho_0(H)$ are observed for samples with substantial differences in ρ_0 , no drastic change should occur here on the nature of the ground state.

UBe_{13} : interplay between superconductivity and magnetism

UBe_{13} is a fascinating heavy Fermion compound as superconductivity appears for a temperature T_c where no Fermi liquid behaviour has still been achieved i.e. the characteristic temperature T^* of the normal ground state is comparable to T_c . The remarkable experimental facts at $P = 0$ are i) in the normal phase an unusual large resistivity just above T_c ($\sim 150 \mu\Omega\text{cm}$) [24] and a thermoelectric power Q which has even not reached its negative maximum ($\sim -20 \mu\text{V/K}$) at $T^{Q_{\min}}$ when superconductivity sets up at $T_c \sim 950 \text{ mK}$

[25] and ii) in the superconducting state, the possible occurrence of large critical fluctuations [10, 21] and evidences for an axial pairing with an energy gap having point nodes on the Fermi surface [27].

The high pressure thermoelectric power $Q(P)$ data of figure 3 show the emergence of a new feature under pressure [28]: a low T positive contribution with a maximum clearly resolved for $P = 67$ and 80 kbar. The negative peak of Q centered at $T^{Q_{\min}}$ was not observable at $P = 0$ for $H = 0$ since $T^{Q_{\min}} < T_c$. It emerges rapidly above T_c under P . Resistivity measurements performed simultaneously show that the pressure dependence of the temperature $T^{\rho_{\max}}$ of the maxima of the resistivity has the same P variation that $T^{Q_{\min}}$. Thus this correlation asserts the same physical origin of $T^{\rho_{\max}}$ and $T^{Q_{\min}}$: the crossover between high and low temperature paramagnetic regimes. The appearance of a positive peak below $T^{Q_{\min}}$ suggests the occurrence of an antiferromagnetic order since large positive peaks of Q have been observed for well known antiferromagnets like UCu_5 [29], TmSe or TmS [30]. In these cases, the change of sign of Q corresponds to the ordering temperature T_N . The high magnetic field dependence of the positive contribution of Q [28] is an other evidence of its magnetic origin. As P increases, larger fields are needed to destroy this positive contribution.

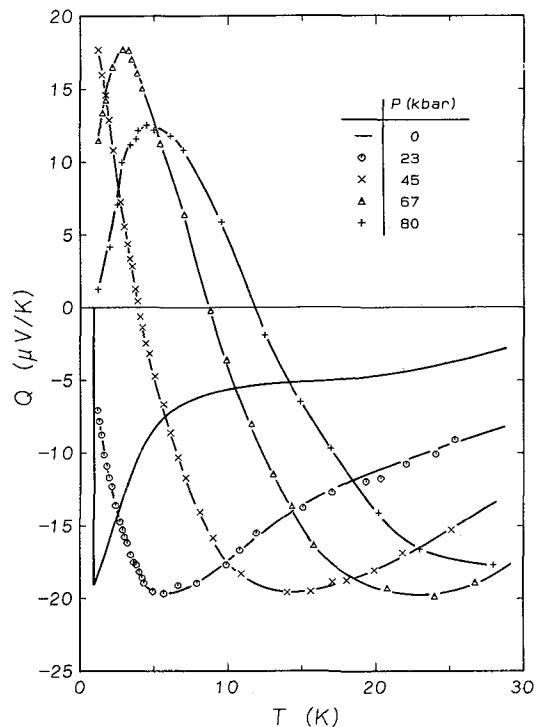


Fig. 3. — Absolute thermoelectric power of UBe_{13} different pressures.

From our previous study at $P = 0$, a positive peak of Q can be inferred below 150 mK. And new specific heat experiments [1, 10] under magnetic field lead to the idea that the normal phase (observed through the contribution of the vortices) could be magnetically ordered below 100 mK.

The occurrence of magnetic ordering is often detected by a change of regime in the magnetoresistivity at T_N , for example, in the cubic magnetic Kondo lattices CeAl_2 ($T_N = 3.8$ K) or CePb_3 ($T_N \sim 1.2$ K) [31, 32]. Below T_N , positive contributions to the magnetoresistivity are found while above T_N , the magnetoresistivity is negative. In UBe_{13} at $P = 0$, the magnetoresistivity has always been found negative (above $T_c(H)$!), down to 35 mK and for $H < 21$ T [10]. So UBe_{13} is paramagnetic in zero pressure for $H > H_{c2}$. At $P = 45$ kbar, measurements were performed down to 1.2 K and only negative magnetoresistivity was observed [28]: T_N should be lower than 1.2 K for $P \leq 45$ kbar.

But at $P = 67$ kbar a positive magnetoresistivity appears at low temperature ($T < 4$ K) : T_N should then be comparable or higher than 4 K. At this pressure, experiments performed down to 50 mK and up to $H = 10$ T, show no trace of superconductivity and a large positive residual magnetoresistivity: $\Delta\rho/\rho = 32\%$ at $H = 10$ T. The T^2 dependence of ρ was observed below 1 K, with a coefficient $A(67 \text{ kbar}) \sim 2 \mu\Omega\text{cmK}^{-2}$ at $H = 0$.

Up to now, microscopic experiments [3, 4] give no sign of antiferromagnetic order for the superconducting phase of UBe_{13} . It may appear that here superconductivity and antiferromagnetism cannot coexist in contrast to the situation observed where uranium atoms are substituted by thorium atoms ($\text{U}_{1-x}\text{Th}_x\text{Be}_{13}$) with $x > 0.015$ [33]. The P disappearance of superconductivity ($T_c(67 \text{ kbar}) < 50$ mK), may be due to the P increase of the ordering temperature T_N of the normal phase. Pressure studies are actually performed to clarify the situation.

At $P = 0$, extensive magnetoresistivity experiments were realized down to 35 mK up to 20 T. Below $T = 900$ mK $\sim T_c(H = 0)$, and above $H = 4$ T, the resistivity can be decomposed in $\rho_0(H)$ plus $A(H)T^2$ terms [34]. Figure 4 shows the large simultaneous decrease of $A(H)$ and $\rho_0(H)$ with H ; the dashed line shows extrapolated values at low field obtained in order to respect the initial H^2 dependence of the magnetoresistivity. The interesting phenomena is that the usual scaling (see [1]) between $A(H)$, the differential susceptibility $\chi(H)$ and the linear T term $\gamma(H)$ of the specific heat ($\sqrt{A(H)} \sim \chi(H) \sim \gamma(H)$) breaks down: $\gamma(H)$ and $\chi(H)$ are field independent almost up to 8 T [35, 10] and to 20 T [34] respectively. This suggests that the field dependence of $A(H)$ is entirely

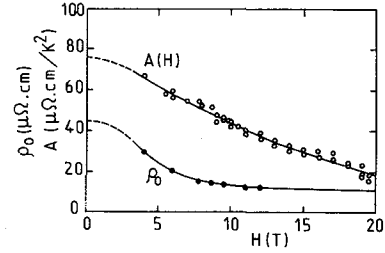


Fig. 4. - Field dependence of (0) $\rho_0(H)$ and (0) $A(H)$ of UBe_{13} at $P = 0$. At $P = 67$ kbar and $H = 0$, $\rho_0(0) = 32 \mu\Omega\text{cm}$ and $A(0) \sim 2 \mu\Omega\text{cmK}^{-2}$.

due to that of the interactions between quasiparticles, not to the field dependence of "band properties". As a funny check of this hypothesis, one can relate $A(H)$ with superconductivity in analogy with pure aluminium. For Al, the A coefficient is strongly enhanced due to the virtual phonon mediated interaction which is also governing the superconductivity. So in UBe_{13} the hypothesis made for the origin of the H variation of the T^2 term of ρ would imply a (known!) field dependence of the attractive potential and thus a field dependent superconducting condensation energy [10].

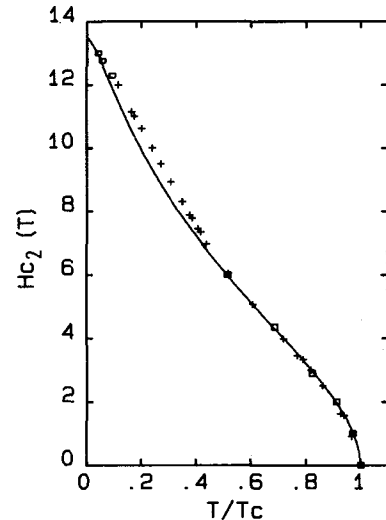


Fig. 5. - For UBe_{13} , comparison of the experimental determination of the upper critical field $H_{c2}(T)$ with an estimation of $H_{c2}(T)$ (full line) assuming an orbital limitation and a BCS condensation energy depending on the magnetic field as described in the text.

Figure 5 shows how the measured unusual shape of the upper critical field $H_{c2}(T)$ can be reproduced assuming an orbital limitation and a BCS field depen-

dent interaction $V(H) \sim \sqrt{A(H)}$ taken from figure 4 [10]. Such an agreement is another strong support to a non phonon mechanism for the superconductivity of UBe_{13} ! [10]. The fit yields an initial slope of H_{c2} at T_c of about 180 T/K, consistent with the experimental estimate of [36] and providing strong support on the possibility of significative critical fluctuation effects near T_c in UBe_{13} [10].

Conclusion

Transport experiments are unique by the facility to operate under extreme conditions: high magnetic field, high pressure and small samples. They could be the appropriate measurement to determine the dependence of ground states (notably magnetism) on lattice's imperfections.

At $P = 0$, CeAl_3 appears as an antiferromagnet with a metamagnetic transition. Above $P \sim 2$ kbar, it is a Pauli paramagnet, and no superconducting phase has been observed down to 40 mK and $P < 8$ kbar. At high pressure, $P > 23$ kbar, UBe_{13} may be in a normal antiferromagnetic ground state. The low pressure superconducting ground state seems incompatible with antiferromagnetism however the magnetic field may restore this long range order in the vortices.

It seems now admitted (1) that the driving mechanism of heavy fermion superconductors is given by antiferromagnetic couplings. However, the existence of only few cases of superconductors may indicate that, over and above these couplings, specific pairing conditions have to be fulfilled (for example on the wavevector and amplitude of antiferromagnetic correlations).

More systematic measurements must be performed to know the importance of disorder on lattice properties: influence on the nature of the ground state, on the magnitude of the effects (size of the ordered moment), on the low temperature excitations... Therefore, special effort must be made to improve the samples quality.

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