Classification
Physics Abstracts
71.20 - 74.40 - 74.60E

LOW TEMPERATURE MAGNETORESISTIVITY OF UBe_{13}

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(Reçu le 26 août 1985, accepté sous forme définitive le 29 novembre 1985)

Résumé.- Des expériences de magnétorésistance montrent une relation simple quadratique T_c de la résistivité de la phase normale juste au-dessus de la température supraconductrice T_c. La résistivité résiduelle dépend, elle, fortement du champ magnétique. Pour 12 T ⩾ H ⩾ 4 T, la magnétorésistivité peut être décomposée en :

$$\rho(T,H) = \rho_0(H) + A(H)T^2$$

ρ_0 et A suivant des variations respectives en $\sqrt{H}$ et H. L'accent est mis sur la dualité particules légères et lourdes.

Abstract.- Very low temperature magnetoresistivity experiments exhibit a simple quadratic T_c dependence of the resistivity just above the superconducting transition temperature T_c. Extrapolated residual resistivity depends strongly on the magnetic field. For 12 T ⩾ H ⩾ 4 T, the magnetoresistivity can be fitted by :

$$\rho(T,H) = \rho_0(H) + A(H)T^2$$

ρ_0 and A following respectively $\sqrt{H}$ and H variations. Emphasis has been given on the duality between light and heavy particles.

The heavy fermion compounds (HFC) have the common property of showing huge values of the ration C/T (the specific heat divided by the temperature). However, large differences occur among them, notably between the well known heavy fermion superconductors (HFS) UBe_{13} [1], $CeCu_2Si_2$ [2] and UPt_3 [3]. The last two compounds have axial symmetry whereas UBe_{13} has a cubic symmetry ; the U-U interdistance in UBe_{13} larger (d ~ 5.13 A) is larger than the corresponding distance (d ~ 4.1 A) between the f atoms in the other two [4]. In zero applied field (H), in UBe_{13}, just above the superconducting transition $T_c \sim 0.9$ K, the resistivity $\rho(T_c^+) \sim 180$ μΩcm is enormous and close to the maximum value of $\rho_M \sim 220$ μΩcm, which is reached for $T_M \sim 2.4$ K. For $CeCu_2Si_2$, $\rho(T_c^+) \sim$ μΩcm, $\rho_M = 150$ μΩcm and $T_M \sim 24$ K while for UPt_3, $\rho(T_c^+) \sim 1.5$ μΩcm and no resistivity maximum can be detected whithout phonon correction [4].

The strong scattering observed for UBe_{13} at T_c^+ precludes characterizing the low temperature properties of UBe_{13} by an unique Fermi temperature $T_F \sim 8$ K as chosen in the analysis of its specific heat and of its magnetization. The latter is linear up to H = 10 T and quasi independent of temperature below T = 4.2 K [5]. A large negative magnetoresistivity has been already observed for UBe_{13} [6]. New experiments performed down to 56 mK at fields up to 12 T, and down to 0.42 K at fields up to 24 T will be reported. The aim is to determine when simple laws, such as T^2 dependence, can be recovered in the resistivity of the normal phase. Data on the upper critical fields H_{c_2} (T) will be briefly given for comparison with published results [6]. Previous experiments [6] were limited to fields up to 10 T and the data analysis were almost restricted to the superconducting transition. Finally magnetization measurements up to 24 T down to 1.3 K will be also reported.

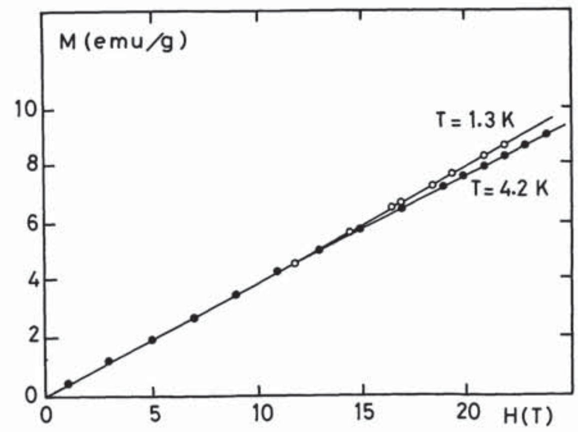

Fig. 1.- Magnetization of UBe_{13} up to 24 T, down to 1.5 K.

Experimental conditions.

The sample was cut from the same batch as that sample recently used for performing thermal conductivity and thermoelectric power (TEP) measurements [7]. Four leads were soldered using indium and the resistivity was measured by an ac cryogenics bridge with a low power dissipation. The linear voltage output allows a continuous field sweep of the resistor at a constant temperature. The error bar on the resistivity data is less than 0.1 μΩcm. For H < 12.5 T the magnetic field was produced by a superconducting coil and the sample immersed in the mixing chamber of a dilution refrigerator (which reaches 56 MK). Between 12.5 T and 24.6 T the field was produced by a polyhelix type resistive magnet and temperatures were achieved in a ^3He cryostat down to 0.42 K. In zero field, the temperature was measured with a Ge thermometer, which was calibrated on ^3He vapour pressure, and was controlled with a capacitance under magnetic fields. DC magnetization (M) measurements show the strong linearity of M with H up to the highest applied field of 24 T at the lowest measured temperature of 1.3 K and the quasi independence of M with the temperature below 4.2 K (Fig. 1).

Magnetoresistivity.

Figure 2 represents the magnetoresistivity for different temperatures and figure 3, at constant field, the temperature variation of the resistivity, measured up to 12.5 T, while figure 4 shows results obtained in resistive magnets. As was observed previously, the magnetoresistivity is large and negative [6,7]. The weak maximum in the temperature variation of ρ at T_M ~ 2.4 K for H = 0 clearly disappears for H > 2 T (for H = 2 T, ρ is quasi constant for 2.4 < T < 3 K). The drop of ρ at H = 21 T has been observed with field increasing or decreasing ; unfortunately, due to the failure of the magnet, it was not possible to repeat these high field experiments.

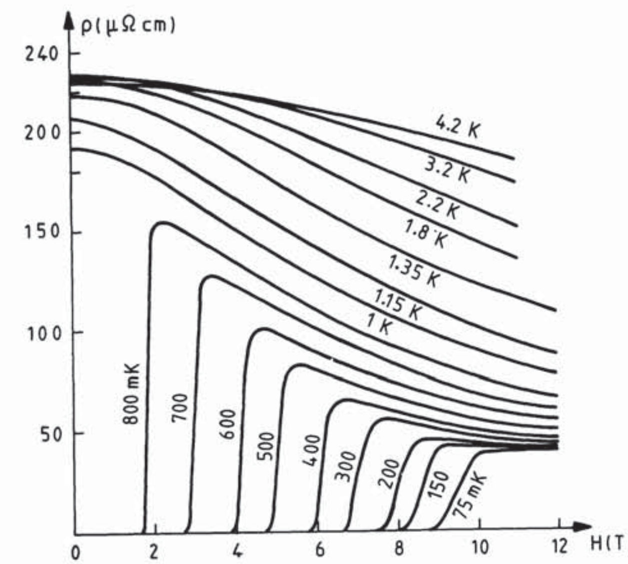

Fig. 2.- Magnetoresistivity of UBe_{13} at different temperatures.

Defining T_c as the temperature corresponding to one half of the resistivity drop, the upper critical field phase diagram can be drawn (Fig. 5). The value found of T_c (H=0) = 880 mK is slightly higher that the temperature T_c (H=0) = 854 mK measured for a sample extracted from the same batch [7] ; this difference may result from difficulties due to thermal gradients in the mixing chamber, when large power must be applied to stabilize the temperature. The low field regime I (H <20 kOe) and high field regime II (H ⩾ 20 kOe) of the phase diagram correspond respectively to an enormous initial slope $\left(\frac{\partial H_{c_2}}{\partial T}\right)_{T_c}$ > -500 kOe /K and to a quasi linear temperature dependence down to 0 K with a slope $\left(\frac{\partial H_{c_2}}{\partial T}\right)_{T \to 0}$ ~

-102 kOe/K, in excellent agreement with published data [1,6,7]. Contrary to other recent reported work [8], no departure from a field linearity of H_{c_2} (T) is observed at T ⟶ 0 K ; an extrapolation to T ⟶ 0 K gives $H_{c_2}(0)$ = 10.2 T.

The main interesting feature is, that over all the field range $H < H_{c_2}(0)$, $\rho(T_c^+)$ follows a T_c^2 dependence i.e. for $H < H_{c_2}(0)$, $\rho(T_c^+)$ obeys the relation :

$$\rho\ (T_c^+) = \rho_0 + \alpha T_c^2$$

with α = 180 µΩcmK^{-2} and ρ_0 = 38 µΩcm as shown in figure 6.

This relation between $\rho(T_c^+)$ and T_c^+ is especially relevant taking into account the apparent enormous disorder scattering of the normal phase.

Figure 3 clearly shows that extrapolations to 0 K lead to residual resistivities which are strongly field dependent. For H > 4 T, ρ(H) can be represented at very low temperature by the relation :

$$\rho(H) = \rho_0(H) + A(H)T^2$$

The striking results are : a) below $H_{c_2}(0)$, dependences of ρ_0 and A in $\sqrt{H}$ and H respectively and b) above H_{c_2}, the linear extrapolation to zero of A for H ∼ 20 T, i.e. near the field, where a drop in the resistivity has been observed (Fig.7). The $\rho_0(H)$ and A(H) curves intersect at low field for H ∼ 2 T and in high field for $H \sim H_{c_2}(0)$. It is worhwhile mentioning that A and T_c vary linearly with H in the same field high regime II. If a connection exists with the regimes I and II of the phase diagram, the crossing of $\rho_0(H)$ and A(H) must imply a characteristic temperature $T^* \sim 1$ K by homogeneity arguments (ρ_0 in µΩcm), A in µΩcmK^{-2}). A similar temperature $T^* \sim 1$ K corresponds also to the mapping of the temperature dependence of the normalized magnetoresistivity by $\frac{H}{T+T^*}$ above 1 K in agreement with previous results (see ref. [4]).

Discussion.

Before discussing in more detail the experiments, let us underline, that the valence i.e. the nature of the magnetism carried by the U centres is not so well known [9], as for trivalent cerium ions in HFC. For example, in a cubic symmetry, a trivalent cerium ion (Kramer's ion) can only be in a non magnetic ground state via a Kondo like coupling while an U^{4+} ion can be in a singlet ground state by the sole mechanism of the crystal field effect like the Pr^{+3} ions. This leads to basic difficulties for decomposing (in a single impurity scheme) crystal field and Kondo effects and also for understanding the nature of the ground state of the lattice notably the occurrence of a non magnetic ground state. In the discussion, the terminology "Kondo temperature", applied to the magnetization, is related to the quenching of the angular momentum even if one of its mechanism is the formation of a singlet crystal field ground state. The presence of a Kondo like mechanism is obvious at high temperature as ρ increase on cooling down to 2.3 K.

At 4.2 K, the magnetoresistivity Δρ is negative but not yet quadratic in H i.e. in M over all the field range as it is the

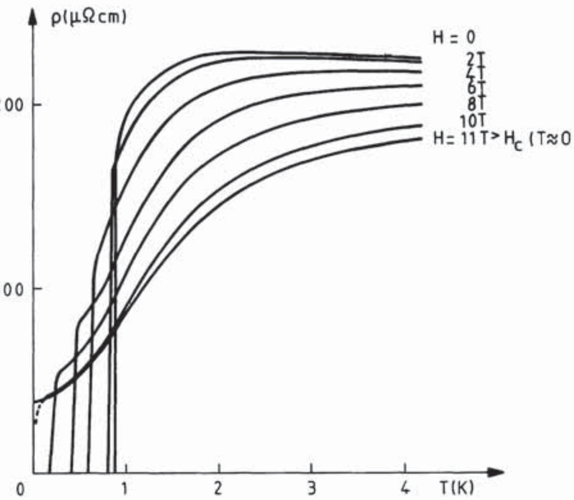

Fig.3.- Temperature dependence of the resistivity at constant field.

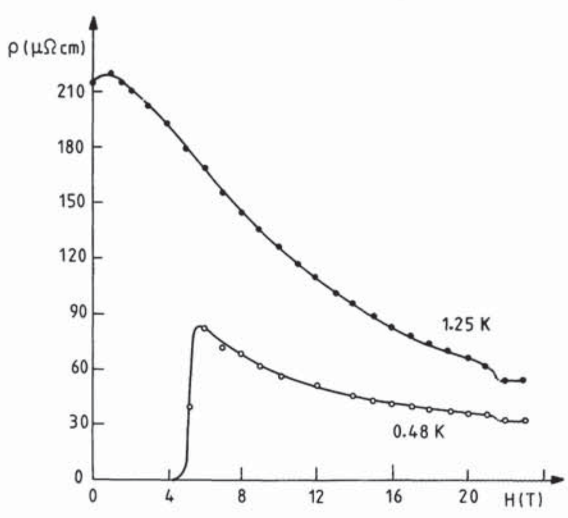

Fig.4.- High field resistivity curve.

case for paramagnetic impurities (Fig. 8). The strong temperature dependence of the magnetoresistivity contrasts with the inpendence of the magnetization with the temperature. That implies different characteristic energies in tranport ($T^* \lesssim 1$ K) and in magnetization measurements ($T_k \gtrsim 8$ K). On cooling, departure from the single ion behaviour increases since lattice effects become important below $T_M \lesssim 2.4$ K. However, the magnetoresistivity remains negative contrary to other HFC like UPt_3 [10,11], $CeAl_3$ [12], $CeRu_2Si_2$ [13] and $CeCu_2Si_2$ [14], where a positive contribution appears on cooling when the coherent lattice regime is approached.

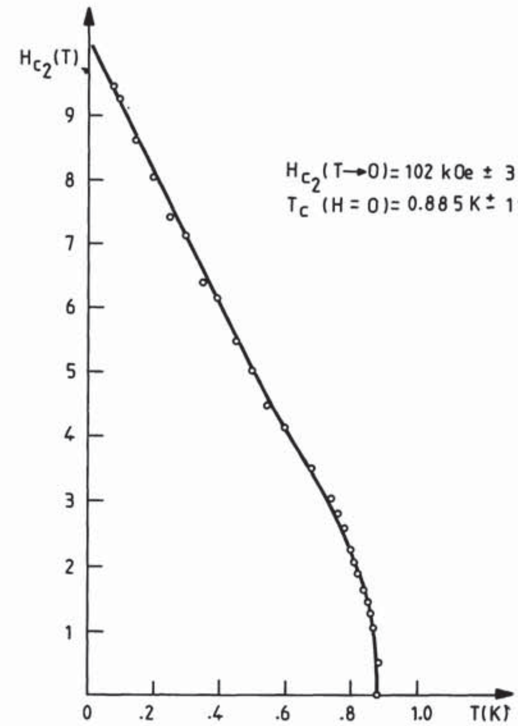

Fig.5.- Upper critical field phase diagram.

In UBe_{13}, a strong scattering of the itinerant electron by the U centres is observed almost down T_c. This behaviour must be correlated with the weak value of $T_M \sim 2.4$ K as compared to the Kondo temperature $T_K \sim 8$ K necessary to explain the weak temperature dependence of its magnetization (Fig. 1). Further evidence is the strong scattering disorder found in TEP experiments, since a negative minimum of Q is not achived for UBe_{13} at T_c (H = 0) [7]. Clearly, the band-width suitable to describe an ordering of the f electrons in the k space must be lower than 8 K. In constrast, as previously emphasized, in $CeCu_2Si_2$ $T_M \sim 24$ K > $T_K \sim 5$ K and in UPt_3 $T_M \gtrsim 300$ K > T_K or T_{sf} the spin fluctuation temperature [15-17] ; a negative minimum of the TEP occurs at 20 K for $CeCu_2Si_2$ [18] and a positive maximum at $T \sim 8$ K for UPt_3 [15]. The almost single ion behaviour of the U atoms in UBe_{13} may be due to the fact that the U ions are strongly isolated from each other by the surrounding 13 Be atoms. Furthermore, the quasi independence of the U atom may be reinforced also from the large number ($n_s \sim 28$) of s electrons per unit cell [19]. Howver, the relation between the number of f centres and the number of itinerant conduction, electrons in the lattice properties is still an open question.

Basically, there are two types of particles involved in the problem : light electrons, given mainly by the Be atoms, and heavy f electrons of the U atoms. These last particles correspond to a weak delocalization of the f electrons by their Kondo like coupling with the itinerant electrons

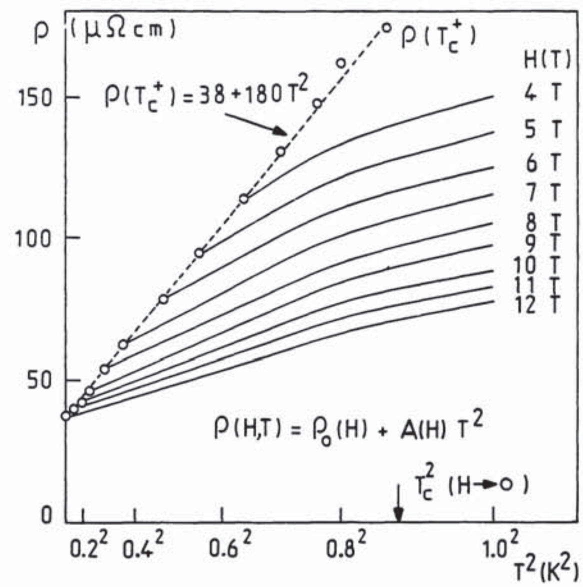

Fig. 6.- Quadratic dependence of $\rho(T_c^+)$ with the superconducting transition and representation of ρ(H) in T^2.

[20,21]. A two component picture of the normal phase has been already considered above T_c in thermal conductivity [21] and Hall effects experiments [22]. Here, it seems supported by a better linearity of the magnetoconductivity ($\Delta\sigma = \sigma(H) - \sigma(H=0)$) than that of the magnetoresistivity in a plot log $\Delta\sigma$(or $\Delta\rho$) versus log H. Such a decomposition must fail when the phase of the two components must be mutually adjusted below T_c or when, in the normal phase, any auxiliary energy reservoir has collapsed. Clearly at $H \to 0$ from resistivity and TEP data [7] the temperature range of the coherent phase in the normal phase is very small, i.e. $\ll T_c$. It has been observed that the A coefficient of the T^2 term of the resistivity can be scaled with the square of

the coefficent γ of the specific heat [31]. The linear decrease of A with the magnetic field suggests a $\sqrt{H}$ field decrease of γ.

The quadratic T_c dependence of $\rho(T_c^+)$ shows that in the vicinity of the superconducting transition one has the temperature dependence usually observed for electron-electron interactions [23,24], the magnitude $\alpha = 180\ \mu\Omega cmK^{-2}$ of the T_c^{+2} term is still larger than that found in the normal phases of HFC (for $CeAl_3$, $A = 35\ \mu\Omega cmK^{-2}$ [25] and for $CeCu_6$ $A = 120\ \mu\Omega cmK^{-2}$ [26,27]. The superconductivity may follow some precursor coherence among the f electrons in the normal phase. Thus, at T_c^+, the T_c^2 law of the resistivity appears to correspond to coherence effects driven by the inset of the superconductivity. There is a mutual push-pull between superconductivity and interactions among particles. Correlatively, the disorder scattering enhances the upper critical fields. In regime I the large disorder observed in resistivity and TEP is associated with an enhancement of $H_{c_2}(T)$ in agreement with the idea that the disorder enhances the paramagnon strength and weaken the pair breaking parameter [28]. Qualitatively, the important of localization effects agree also with the $\sqrt{H}$ dependence of the residual resistivity ρ_0 found for $4\ T < H < H_{c_2}(0)$ as a similar negative magnetoresistivity is observed for weak 3d localization [29]. Quantitatively, the variation of the residual magnetoresistivity is not a weak perturbation and it is necessary to convert resistivity in conductivity. The negative magnetoresistivity is lower by a factor 3 from that predicted by the theory of a weak localization. Interaction effects in disordered systems must also be considered.

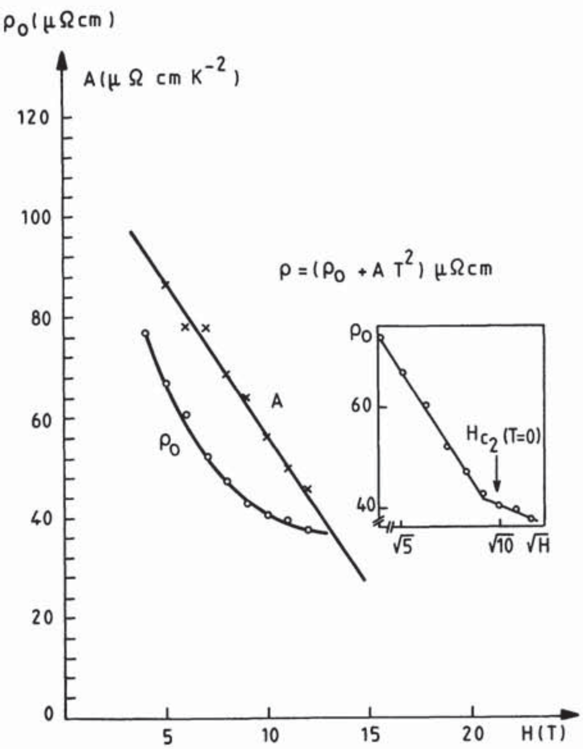

Fig. 7.- Field variation of the coefficient $\rho_\bullet(H)$ and $A(H)$, obtained by the fitting of the resistivity $\rho(T,H)$ by $\rho_\bullet(H) + A(H)T^2$.

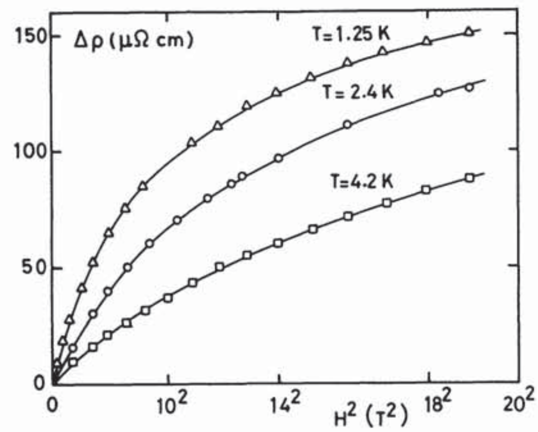

Fig. 8.- Dependence $\Delta\rho = \rho(0) - \rho(H)$ at different temperature as a function of H^2 The linearity of $\Delta\rho$ with H^2 is only observed for H < 6.3, 4 and 2.5 T respectively at T = 4.2 K, 2.2 K and 1.25 K.

The regime change in $\rho(H)$ for $H \sim H_{c_2}(0)$ corresponds to a domain, where a T^2 law is obeyed up to $0.8\ K \approx T_c(H=0)$ and and where a change in the sign of TEP seems to occur from negative to positive [7]. For H = 0, the electronic mean free path (l_e = = 13 A for $\rho \sim 130\ \mu\Omega cm$) is smaller than the superconducting coherence length $\psi_\bullet \sim$ 65 A). The interesting feature is that the large negative magnetoresistivity leads to an increase of the mean free path such that it becomes comparable to $\psi_\bullet$ for $H \sim H_{c_2}(0)$.

The applicability of the dirty limit approximation is doubtful for UBe_{13} since a decrease of $\rho(T_c^+)$ by 30 % [6] gives results similar to those reported here. Experiments on samples of different purities performed in the same apparatus to minimize experimental errors must be performed to clarify this point and also the interplay between localization, interaction and disorder. Finally up to H < 24 T and T > 0.46 K no reentrant superconductivy has been found as recently proposed [30].

Conclusion.

The interplay between coherence effects of a lattice, localization and interaction effects is obvious for the specific case of UBe_{13}. The main interesting feature is the connection between superconductivity and the properties of the normal phase. Reducing the physics of HFC to the sole f particles neglects the interesting problem of coupling between two interacting systems. Finally, the diversity of the hierarchy between basic parameters (T_K, T_M, anisotropy and crystal field effects) leads to drastic differences between HFS which needs to be understood via their normal phases.

Acknowledgments. - Work at Los Alamos was performed under the auspices of the US Department of Energy. A major part of these results was presented orally at the Meeting of Baltimore (1984) of the American Physical Society [32].

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