Normal and superconducting properties of UBe13

J.P. Brison, O. Laborde, D. Jaccard, J. Flouquet, P. Morin, Z. Fisk, J.L. Smith

► To cite this version:

J.P. Brison, O. Laborde, D. Jaccard, J. Flouquet, P. Morin, et al.. Normal and superconducting properties of UBe13. Journal de Physique, 1989, 50 (18), pp.2795-2810. 10.1051/jphys:0198900500180279500 . jpa-00211103

HAL Id: jpa-00211103

<https://hal.science/jpa-00211103>

Submitted on 1 Jan 1989

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Classification

Physics Abstracts

72.15Gd — 72.15Qm — 74.70Tx — 65.40f

Normal and superconducting properties of UBe_{13}

J. P. Brison (¹, *), O. Laborde (¹), D. Jaccard (¹, **), J. Flouquet (¹), P. Morin (²), Z. Fisk (²) and J. L. Smith (³)

(¹) Centre de Recherches sur les Très Basses Températures, C.N.R.S., BP 166 X, 38042 Grenoble Cedex, France

(²) Laboratoire Louis-Néel, C.N.R.S., BP 166 X, 38042 Grenoble Cedex, France

(³) Los Alamos National Laboratory, Los Alamos NM 87545, U.S.A.

(Reçu le 25 avril 1989, accepté sous forme définitive le 30 mai 1989)

Résumé. — Nous décrivons des expériences de magnétorésistivité dans une large gamme de température ($30 \text{ mK} < T < 80 \text{ K}$) et de champ magnétique ($H < 20 \text{ T}$). A haute température ($T > 4 \text{ K}$), la magnétorésistance suit une loi d'échelle en H/H^* à un seul paramètre $H^*(T)$. Cependant, il n'existe pas de lien simple entre magnétorésistance et aimantation. A basse température, les termes $A(H) T^2$ de la résistivité varient fortement sous champ tandis que la chaleur spécifique de la phase normale est indépendante du champ. Cette forte variation de $A(H)$ est due à la forte dépendance en champ de l'interaction directe entre quasi particules. Ces propriétés de la phase normale sont reliées à celles de la phase supraconductrice (notamment, la chaleur spécifique et la conduction thermique).

Abstract. — Extensive magnetoresistivity experiments in large temperature ($30 \text{ mK} < T < 80 \text{ K}$) and field ($H < 20 \text{ T}$) ranges are reported. At high temperatures ($T > 4 \text{ K}$), the magnetoresistivity obeys a scaling law in H/H^* with one temperature adjustable parameter $H^*(T)$. However no simple relation can be found between magnetoresistivity and magnetization. At low temperatures the $A(H) T^2$ terms of the resistivity are strongly field dependent although the specific heat of the normal phase is field independent. It is proposed that the field variation of $A(H)$ reflects the strong field dependence of the direct interactions between quasi particles. The low temperature magnetoresistivity data are connected with the unusual superconducting properties of UBe_{13} (specific heat and thermal conductivity).

Introduction.

The heavy fermion UBe_{13} is thought to be one of the good examples of a « Kondo lattice » system. This means that the occurrence of the heavy quasi-particles at low temperatures would be due to the coupling between localized f electrons and conduction electrons. One is led to distinguish between a « high temperature regime » where the uranium ions are

(*) *Present address* : Department of Physics, University of Florida, Gainesville, U.S.A.

(**) Département de la Matière Condensée, 1211 Genève, Switzerland.

considered like magnetic impurities scattering the conduction electrons, and a « low temperature coherent state », with heavy quasiparticles in a « perfect » lattice, described within the framework of strongly interacting Fermi liquids and in a non-magnetic ground state.

In this article we report mostly magnetoresistivity and magnetization (M) experiments performed in a large range of magnetic fields (H) and temperatures (T). These measurements are appropriate to check the preceding ideas and explore the properties of UBe_{13} in both regimes. Their analysis suggests links between normal and superconducting properties like the upper critical field (H_{c2}), the low temperature specific heat (C) and the thermal conductivity κ .

1. Experimental set up.

The magnetoresistivity measurements were performed on the same polycrystalline sample using an ac bridge. The contacts were realized with indium ; the current was parallel to H .

The different temperature ranges were obtained in a He^4 bulb (for $T \geq 4.2$ K) with the sample immersed in pumped He^4 ($1.5 \text{ K} < T < 4.2 \text{ K}$) or in dilution refrigerators ($30 \text{ mK} < T < 4.2 \text{ K}$). The magnetic field was produced either by superconducting magnets (for fields up to 12 T) or in a resistive Bitter magnet, at the S.N.C.I. (Grenoble-France) for H up to 20 teslas. Particular attention was given to the thermometry under magnetic field : the isothermal $\rho(H)$ curves were obtained by a regulation of the temperature with the helium bulb, by the vapor pressure of the bath or by the use of a reference capacitor which is only weakly field dependent. The $\rho(T)$ curves at fixed field were made after calibration of the carbon or platinum thermometers under H .

Isothermal magnetization measurements were collected on a single crystal coming from the same batch as the polycrystalline sample. $H(< 8 \text{ T})$ was applied along $a(0, 0, 1)$ axis ; the temperature varied from 1.5 to 300 K.

2. High temperature magnetoresistivity.

2.1 RESISTIVITY FOR $H = 0$. — Figure 1 shows the resistivity of UBe_{13} in zero magnetic field. We see an increase on cooling from $T = 300 \text{ K}$ down to 2.5 K where ρ passes through a

Fig. 1. — Present determination of the resistivity of UBe_{13} in zero field showing an increase on cooling down to 2.5 K, followed by a decrease attributed to the occurrence of coherence before the superconducting transition (at 950 mK).

maximum. This temperature of 2.5 K is often called « coherence temperature » (T_{coh}), because it is assumed to separate the high temperature regime from the low temperature coherent one. The upturn of ρ below 10 K is reminiscent of that of the single impurity Kondo behavior, and the decrease of ρ below 2.5 K is ascribed to the formation of the heavy quasiparticles (due to the Kondo coupling between localized f electrons and conduction electrons) which restores the translational symmetry of the lattice. In this scheme, most of the resistivity above T_{coh} originates from the diffusion of the conduction electrons on uranium ions considered as magnetic impurities. Thus, magnetoresistivity should provide useful information on this high temperature incoherent regime.

2.2 MAGNETORESISTIVITY FOR $T > T_{\text{coh}}$. — Figure 2 shows the corresponding magnetoresistivity $\rho(H)$ at a fixed temperature ($T = 1.6$ K to 80 K) and from 0 to 20 teslas. It is negative for all temperatures and the magnitude of $\Delta\rho$ increases on cooling. The interest of the high field measurements appears clearly. For $T = 4.2$ K, a value of $\Delta\rho/\rho = \frac{\rho(0) - \rho(H)}{\rho(0)}$ of the order of 0.45 is achieved for $H = 20$ T : this will allow us to check the models.

Fig. 2. — Magnetoresistivity curves of UBe_{13} ($0 \leq H \leq 20$ T), for temperatures $1.6 \text{ K} < T < 80 \text{ K}$.

When the magnetoresistivity is due to the polarization of magnetic impurities and is governed by one energy scale, the isothermal $\rho(H)$ curves follow scaling laws on H with a temperature dependent effective field $H^*(T)$. In the reduced variable H/H^* , the magnetoresistivity is given by $\rho(H, T) = \rho(0, T) \phi(H/H^*(T))$ where ϕ is a universal function. For Kondo impurities [1], the condition is satisfied at least for $T \gg T_K$ (where T_K is the Kondo temperature). Renormalizing $\rho(H, T)$ to $\rho(0, T)$ takes into account the temperature dependence of the effective interaction between the impurities and the conduction electrons. $H^*(T)$ should vary like $(T + T_K)$ [1] to be consistent with the behaviour of the susceptibility (χ) in this temperature range (the magnetization scales like $M = M_{\text{sat}} g(H/H^*(T))$ and thus $1/\chi \propto H^*(T)$). To check the scaling laws in UBe_{13} in the temperature range $T > T_{\text{coh}}$, the $\rho(H)$ curve at 4.2 K ($\rho(H, 4.2)$) was used as a reference.

The best superposition of the $\rho(H)/\rho(0)$ curves (taken from Fig. 2) on $\rho(H, 4.2)/\rho(0, 4.2)$ through a scaling : $H \rightarrow H/\alpha(T)$ is shown in figure 3. It is perfect within experimental accuracy, in the whole range of fields and temperatures ($4.2 \text{ K} < T < 35 \text{ K}$) explored. The temperature dependence of $H^*(T)$ is shown in figure 4 ; the value at 4.2 K was fixed so that $\rho(H)/\rho(0) = 0.5$ at $H = 0.64 H^*$, as for spin 1/2 Kondo impurities [2]. The results of [1] are recovered : above 15 K, $H^*(T)$ follows a linear T dependence : $H^*(T) \sim (T + T_K)$ with $T_K \sim 13 \text{ K}$ while below 15 K strong deviations from linearity appear.

Fig. 3. — Superposition of the curves of figure 2 from $T = 4.2 \text{ K}$ to 40 K , through a scaling on the magnetic field as explained in the text.

Fig. 4. — Behaviour of the effective characteristic field $H^*(T)$ derived from the scaling relations.

2.3 MAGNETORESISTIVITY FOR $T < T_{\text{coh}}$. — For $T < T_{\text{coh}}$, the description of f electrons in one impurity schemes is expected to break down. Instead, a Fermi liquid picture of strongly interacting quasiparticles might be relevant, as for other heavy fermion systems in their low temperature regime. Thus in UBe_{13} , a change in the behaviour of the $\rho(H)$ curves at 1.6 and 2.1 K was expected.

Figure 5 shows that, indeed, the scaling laws on H do not work anymore on the last two curves : taking the curve $\rho(H, 1.6)/\rho(0, 1.6)$ as a reference spoils completely the superposition of the curves described previously. This alone does not mean that one impurity schemes are obsolete in the coherent regime : even for the one impurity Kondo theory it is not expected that scaling laws work in the whole temperature range $T \gg T_K$ down to $T \ll T_K$. But, it prevents a meaningful discussion of the behaviour of $H^*(T)$ below 4.2 K.

Fig. 5. — Best superposition of all the curves of figure 2, taking the curve at 1.6 K as a reference for the scaling described previously, showing a spoiling of the result when curves in the coherent regime are taken into account.

2.4 SUSCEPTIBILITY AND MAGNETIZATION. — Even at low temperatures the magnetization is strictly linear in H . For $H = 8$ T, M reaches a value of $0.2 \mu_B$ per uranium atom. Using Arrott's method (Fig. 6), the linearity of M with H is obvious. However one notices a non reversible behavior of M in decreasing field which exists in the whole temperature range investigated and is not yet explained. It has already been reported that M is linear in H up to 24 T down to 1.5 K [5].

Fig. 6. — Representation of the magnetization (M) data by Arrot's plot : M^2 versus H/M .

Arrott's method allows an accurate determination of the reciprocal initial susceptibility χ . χ follows a Curie-Weiss law above 150 K with $\theta = -85$ K and $\mu_{\text{eff}} = 3.36 \mu_B$: thus value is slightly different from the value $\mu_{\text{eff}} = 3.05 \mu_B$ of [3] but is consistent with the data of [4] which $\mu_{\text{eff}} = 3.4 \mu_B$ between 300 K and 1 000 K. This is not too far from the free ion value of

$J = 4$ ($\mu_{\text{eff}} = 3.58 \mu_B$) and $J = 9/2$ ($\mu_{\text{eff}} = 3.62 \mu_B$). Between 150 K and 20 K, $1/\chi$ seems to follow a linear T dependence with $\theta = -130$ K (Fig. 7). The apparent increase of θ may be due to crystal field effects. Below 20 K, our data agrees with those of [3]. The strong downwards curvature of $1/\chi$ at low temperatures is a good indication of a magnetic ground state for the crystal field levels of the U ions.

Fig. 7. — Temperature dependence of the susceptibility χ in full line the data of reference [3]. Insert is the low temperature regime.

2.5 DISCUSSION. — The observation of scaling laws on $\rho(H)$ and the temperature dependence of the susceptibility point towards a picture of a localized magnetism for the U ions. This is consistent with spectroscopic measurements [6] which favor a $5f^3$ configuration. A crystal field scheme Γ_6 - Γ_8 - Γ_8 with a Kramers doublet Γ_6 was proposed for explaining specific heat data [7]. However the splitting $\Gamma_6 \rightarrow \Gamma_8$ of 180 K appears rather large in relation to the strong isotropy of the U sites and in comparison to the REBe_{13} isomorphous compounds. Indeed in NdBe_{13} and in PrBe_{13} [8], the overall splitting reaches only 70 and 125 K respectively although they are the largest across the REBe_{13} series. Thus the comparison of UBe_{13} with REBe_{13} is difficult; the nature of low lying levels in UBe_{13} remains an open question.

The energy scale $T_K \sim 13$ K deduced from the temperature dependence of H^* (i.e. from the scaling law on $\rho(H, T)$) is consistent with the large increase of ρ on cooling below 10 K and roughly with the estimation of the Kondo temperature made from specific heat data [7] $T_K = 7.7$ K. Below 15 K, the departure of $H^*(T)$ from a linear T dependence is quite natural since in the one impurity Kondo theory a crossover occurs at $T \sim T_K$ and there are no reasons for the same scaling law being valid for $T < T_K$. Nevertheless, it appears to be verified in UBe_{13} down to $T_K/3$. Below 15 K, the curvature of $H^*(T)$ is consistent with that of $1/\chi$.

But a further comparison of the magnetoresistivity and magnetization is disappointing. For example, at $T = 4.2$ K, M is linear in H almost up to 24 T [5] whereas at the same temperature, $\Delta\rho$ deviates significantly from an H^2 (i.e. M^2) behaviour for fields above 5 T. This discrepancy also clearly appears in the comparison with exact results derived for spin 1/2

Kondo impurities at $T = 0$ [2]. This comparison is somewhat questionable as $\rho(H/H^*)$ is certainly different at 4.2 K and 0 K (scaling laws do not work below 4.2 K) but should give good « orders of magnitude ». (As regards to magnetization, it does not vary much on cooling below 4.2 K). Figure 8 shows in full lines the exact theoretical solutions for $\rho(H/H^*)/\rho(0)$ and $M(H/H^*)/M_{\text{sat}}$, with the experimental scaling curve $\rho(H/H^*(T))/\rho(0)$ and the magnetization at 4.2 K as a function of H/H^* (4.2 K).

Fig. 8. — Full lines : theoretical predictions for the magnetoresistivity and the magnetization of spin 1/2 Kondo impurities at $T = 0$. Dots : experimental points for the magnetoresistivity of UBe_{13} in the temperature region where scaling laws work. Dotted line : magnetization of UBe_{13} at 4.2 K (from [5]) as a function of H/H^* (4.2 K) and normalized so that its slope in $H = 0$ coincides with the theoretical one.

This strongly suggests that the magnetoresistivity and the magnetization of UBe_{13} are not controlled by the same processes. This idea is also supported by the different energy scales relevant for the magnetoresistivity ($T_K \sim 13$ K) and the magnetization ($\theta \sim 85$ K to 130 K) ; the latter being consistent with the linearity of M up to high fields even at 4.2 K. Recent thermal expansion and magnetostriction experiments [9] have also ruled out a description of the high temperature properties of UBe_{13} with a unique parameter.

Furthermore, this study of UBe_{13} at high temperatures shows no direct evidence of a Kondo effect in this compound. The classification of UBe_{13} as a « Kondo lattice » relies essentially on the appearance of a coherent regime without long range magnetic ordering, whereas it is described in terms of a localized magnetism at high temperatures (up to now, neutron diffraction measurements have failed to detect any static sublattice magnetization down to 50 mK [12]). But the shape of $\rho(H/H^*(T))$ cannot be precisely compared to theoretical predictions for Kondo impurities, available only at $T = 0$, and the curvature of $H^*(T)$ below 15 K is opposite to that expected for an antiferromagnetic coupling between conduction and localized electrons. Moreover, the existence of two energy scales points out that the effects of Kondo couplings (if any) in UBe_{13} are at least strongly altered by competition with some type of R.K.K.Y. interactions (and not only by the very high concentration of « magnetic impurities »). Indeed, the occurrence of magnetic correlations in UBe_{13} was perhaps already directly observed in preliminary neutron measurements [10] and indirectly in Hall effect experiments [11]. Still, to keep a simple picture, one could guess that magnetoresistivity is mostly sensitive to interactions between f electrons and conduction electrons (energy scale

$T_K \sim 13$ K) while magnetization is mostly governed by the f electron-f electron coupling (energy scale $\theta \sim 85$ to 130 K). In fact, the need for two different energy scales is also recovered in the low temperature heavy fermion regime.

3. Low temperature magnetoresistivity.

3.1 MEASUREMENTS. — Below 1 K, two different types of measurements have been performed

— for $H \leq 12$ T, measurements at fixed fields to determine the temperature dependence of the resistivity. It was found to be quadratic for $T \leq 900$ mK, and T higher than the critical temperature of the superconducting transition in the given field, restricting the analysis to $H \geq 4$ T (cf. Fig. 9) :

$$\rho = A(H) T^2 + \rho_0(H). \quad (1)$$

Fig. 9. — T^2 analysis of the magnetoresistivity of UBe_{13} , for H between 12 and 4 T, and valid below 900 mK.

— Up to 20 T, measurements at fixed temperatures were performed (Fig. 10), from which the values of $A(H)$ and $\rho_0(H)$ could be determined up to 20 teslas. Below 150 mK, the resistivity does not show any temperature dependence, yielding directly $\rho_0(H)$ between 15 and 20 teslas. Knowing $\rho_0(H)$ from 4 to 20 T, $A(H)$ was then easily extracted from the curves at 620, 480 and 320 mK of figure 10.

The superconducting transition occurring at 950 mK prevents a direct measurement of $A(H)$ and $\rho_0(H)$ in low fields. Nevertheless, as shown in figure 2 with the curves for 2.1 K and 1.6 K, the field dependence of $\rho(H, T)$ for T slightly above T_c is nearly quadratic in low field, and for $H > 4$ T, the analysis (1) could be even extended to the curve at 1.6 K, just

Fig. 10. — Low temperature measurements in high fields up to 20 T of the magnetoresistivity of UBe_{13} .

assuming a weaker temperature dependence. This allows to extrapolate $A(H)$ and $\rho_0(H)$ down to $H = 0$ with an almost quadratic H law, consistent with the $\rho(H)$ curves at 1.6 K. The field dependence of $A(H)$ and $\rho_0(H)$ determined that way is reported in figure 11.

Fig. 11. — Determination of $A(H)$ and $\rho_0(H)$ through the measurements at fixed field (some of them presented in Fig. 9) and at fixed temperature (from Fig. 10). Below 4 T, extrapolated values as explained in the text.

3.2 COMPARISON WITH PREVIOUS RESULTS. — A T^2 behaviour of the resistivity at low temperatures is common in heavy fermion systems. It is often associated with the Fermi liquid behaviour of the heavy quasiparticles. A simple idea is then to consider $A(H)$ as an intrinsic property of the system, linked to the interactions between quasiparticles, and $\rho_0(H)$ as a residual resistivity, determined by the impurities within the sample.

The analysis (1) had been performed previously on another sample of UBe_{13} [5] and it is thus possible to check the sample dependence of $A(H)$ and $\rho_0(H)$. The « old » and « new » results are reported in figure 12, showing a weaker sample dependence of $A(H)$ (factor 1.3)

Fig. 12. — Comparison of the previous results of [5] and the present one from the fixed field measurements : while $A(H)$ changed by a factor 1.4, $\rho_0(H)$ decreased by a factor 3 on the new sample.

and a stronger decrease of $\rho_0(H)$ for this new sample (factor 3) ; this is in good agreement with the origin proposed for both quantities.

It had also been reported in [5] that the limit of validity of the T^2 law (almost the superconducting transition T_c in zero field) was field independent. This important feature is also verified here at least up to $H = 12$ T.

3.3 DISCUSSION. — The high temperature impurity scheme is now left for a Fermi liquid picture, although other authors still apply the high temperature analysis to this low temperature regime [1, 13]. One important feature is that the magnetoresistivity of UBe_{13} is always negative in the whole range of fields and temperatures yet explored. This is more easily accounted for in the one impurity picture which nevertheless implies the occurrence of superconductivity at 950 mK in the presence of a large number of efficient paramagnetic centers.

In cubic heavy fermion compounds such as CeAl_2 , CeIn_3 , CePb_3 , above their Néel temperature T_N , a negative magnetoresistivity is observed while positive contributions of the magnetoresistivity appear below T_N [14]. In UBe_{13} at zero pressure (P), the fact that only negative magnetoresistivity is detected suggests that UBe_{13} is paramagnetic above the upper critical field H_{c2} ($T \rightarrow 0$). Recent specific heat measurements under magnetic field seem to indicate a magnetic ordering of the normal phase of UBe_{13} below 150 mK [12, 15]. Our magnetoresistivity data imply an associated critical field $H_M(T \rightarrow 0) \leq H_{c2}(T \rightarrow 0)$. It is worthwhile to notice that transport measurements under pressure show for $P = 67$ kbar the

emergence of a positive magnetoresistivity below $T < 4$ K [14]. At $P = 67$ kbar, UBe_{13} may be antiferromagnetic ; the appearance of this long range magnetic ordering also coincides with the P disappearance of superconductivity. In UBe_{13} , superconductivity and antiferromagnetism seem to be antagonistic, in contrast to the situation observed when U atoms are substituted by Th atoms ($\text{U}_{1-x}\text{Th}_x\text{Be}_{13}$ with $x > 0.015$) [16].

3.4 ANALYSIS OF THE T^2 TERM. — The discussion of the $A(H) T^2$ term has already been reported in connection with the field dependence of the upper critical field [25]. Here only complementary points will be given.

Usually, it seems possible to describe the low temperature coherent regime of heavy fermion systems as a Fermi liquid with one energy scale governing both the band properties (like the specific heat) and the interactions between quasiparticles (as seen in transport properties). In UBe_{13} , as already found for the high temperature regime ($T > T_{\text{coh}}$), there are no connections between the field variation of its specific heat (at least up to 8 teslas [15, 17, 18]) and the strong decrease of $A(H)$ in field. One of the simplest way to account for this is to rely upon the distinction between the different bare interactions made in the high temperature regime :

1) The conduction electron-f electron interactions which give rise to the strong resistivity of UBe_{13} above 2.5 K and are assumed to be responsible for the formation of the heavy quasiparticles and thus of the band properties in the low temperature regime. The observation of scaling laws shows that these interactions are weakly field dependent. (The basic assumption justifying the scaling is that the magnetoresistivity arises only from the polarization of the « impurities »).

2) f electron-f electron interactions which produce magnetic correlations. When the heavy quasiparticles are formed, the conduction electrons and f states are hybridized. Thus, these interactions could give an effective quasiparticle-quasiparticle scattering in the coherent regime. We propose that the latter interactions are field dependent and dominate the magnetoresistivity at low temperatures, the effect of previous interactions being mostly suppressed by the formation of the quasiparticles (coherence).

Two different mechanisms may lead yet to a T^2 term in the resistivity [19]. The more common is through the diffusion of the quasiparticles by thermally excited magnetic fluctuations. If the high value of the Curie-Weiss temperature θ derived at high temperatures (85 K) is taken as a characteristic of magnetic fluctuations, few thermal magnetic fluctuations should remain at low temperatures, resulting only in a small contribution of this mechanism to the large $A(H) T^2$ term. Besides, current theories on weak or nearly antiferromagnets predict no T^2 term above T_N [20, 21].

The other mechanism is a direct quasiparticle-quasiparticle interaction mediated by virtual spin fluctuations. It has been argued [25] that in UBe_{13} , it could produce a large T^2 term. Notably, the strength of this interaction might be emphasized by the proximity of T_c and T_{coh} (the T^2 term is not even observed at T_c in zero field) : the attractive potential leading to superconductivity could be due to such a coupling. In this scheme, the T^2 dependence of ρ arises from the usual phase space arguments of Fermi liquid theory [19], the limit of validity of the T^2 law could then be related to band properties (thus its field independence and also its pressure dependence). On this last point, it is worthwhile to notice that the pressure dependence of the A coefficient seems to follow that of the linear temperature term γ of the specific heat according to the scaling law $A \sim \gamma^2$. Our extrapolation of A for $H = 0$ is $70 \mu\Omega\text{cmK}^{-2}$ while A is found equal to $20 \mu\Omega\text{cmK}^{-2}$ and $2 \mu\Omega\text{cmK}^{-2}$ respectively for $P = 15$ kbar [22] and 67 kbar [14, 23]. Assuming a compress-

ibility $K = 10^{-6} \text{ bar}^{-1}$, that leads to a Grüneisen parameter at $P \rightarrow 0$ of $\Omega = \frac{\partial \text{Log } \sqrt{A}}{\partial \text{Log } V}$ equal to 43, in relative good agreement with the values 60 and ~ 50 found respectively by specific heat [24] and thermal expansion [9] experiments.

A previous paper [25] provided support to the idea that the field variation of A reproduces the field dependence of the interaction potential, notably the attractive « B.C.S. » potential $V(H)$. The unusual curvature of the upper critical field of UBe_{13} near T_c could be explained assuming that $A(H) \propto V^2(H)$ and then using a field dependent « bare » transition temperature : $T_c(H) \propto \exp(-1/\rho_d V(H)) = \exp(-\alpha/\sqrt{A(H)})$, where ρ_d is the density of states at the Fermi level. In fact, not only can we assume $A \propto V^2$, but also $A \propto \rho_d^2 V^2$: using the simple kinetic formula : $\rho = \frac{m^*}{ne^2} (1/\tau)$ one can see that the scattering rate contributes a factor $\rho_d |V|^2$ (cf. the Fermi Golden Rule) and the « effective mass » the other factor ρ_d . The interest is that the pressure measurements confirm these ideas. Under pressure, both ρ_d (cf. C_p measurements [24]) and, very likely, V will vary ; but still the link between T_c and A should remain (as long as the same « virtual » interactions control A). With the value $\alpha = 3.3 \times 10^{-2}$ used to fit H_{c2} in [25] one can then predict a relation between T_c and A in zero field under pressure :

$$\frac{1}{\sqrt{A(P)}} = \frac{1}{\sqrt{A(0)}} + 3.3 \times 10^{-2} \ln \frac{T_c(P=0)}{T_c(P)}.$$

Careful measurements will be needed to check this relation, but at least the order of magnitude estimates deduced from the data of [22] are not in disagreement with this relation. Let us finally stress that the pressure increase of the quasiparticle Fermi energy (or the pressure decrease of γ) is associated with a P decrease of the superconducting temperature [24]. Similar phenomena are found for UPt_3 and liquid ^3He . Such effects are opposite to the general tendency recently reported that T_c increases when γ decreases [26].

3.5 ANALYSIS OF $\rho_0(H)$. — In heavy fermion compounds, the residual resistivity is a non-trivial quantity since it is connected with the lattice properties. For example in CeAl_3 , the pressure and field dependences of A also correspond to large variations of ρ_0 [27]. In compounds where tiny magnetic moments are ordered, one can ask if this would not be driven by impurities. In UBe_{13} , the occurrence of superconductivity at 950 mK prevents us from observing the low field behavior of $\rho_0(H)$ directly but its large decrease above 4 T can be also interpreted as an interplay between extrinsic and lattice properties. In those systems considered as « Kondo lattices », it has been argued [28] that in the low temperature coherent regime, a single impurity is a major defect in the regular array of Ce or U ions ; it could be viewed by the quasiparticles as a Kondo hole with a phase shift near $\pi/2$ at the Fermi level. The idea would be that, at $H = 0$, the impurities could have a relative phase shift of $\pi/2$ which moves away from this value as H is increased (giving rise to a negative magnetoresistivity). The saturation of $\rho_0(H) \sim 10 \mu\Omega\text{cm}$ above 10 teslas would coincide with the recovery of the true (bare) residual resistivity. Again, the properties of the superconducting state provide a check for this hypothesis.

3.6 LOW TEMPERATURE SPECIFIC HEAT AND THERMAL CONDUCTIVITY IN THE SUPERCONDUCTING STATE. — In anisotropic superconductors, even simple impurities have strong « pair breaking » effects. As a result, a kind of gap-less behaviour is observed in impure samples for $T \ll T_c$, with manifestations such as residual linear T terms in the specific heat and thermal

conductivity (instead of higher order power law behaviours) [28-31], with a high sensitivity to the phase shift of the impurities [29, 31]. Specific heat measurements (on a sample from the same batch as the one used for the present magnetoresistivity measurements) show a minimum of C_p/T at 90 mK before it reaches a residual finite term $\gamma_0 \sim 70 \text{ mJ.mole}^{-1} \text{ K}^{-2}$. This « resonance » structure at low values of T/T_c is in agreement with theoretical predictions on the effect of a small amount of impurities on C_p/T in the unitary limit [29, 31] (Fig. 13).

Fig. 13. — At $H = 0$, temperature variation of C/T . The extrapolation of C/T to 0 K above 200 mK gives a vanishing residual term [12]. The insert shows the unusual sharp anomaly of C_p/T at T_c due to the occurrence of large critical fluctuations (see [25]).

Furthermore, this can be compared with measurements on the same polycrystalline sample of the thermal conductivity (κ). The problem with the latter quantity is that in UBe_{13} , κ is not simply proportional to T at T_c . Between 1 K and 4.2 K, κ can be analyzed as : $\kappa = \alpha T + \beta T^2$; $\alpha = 0.29 \text{ mW/K}^2 \cdot \text{cm}$; $\beta = 0.26 \text{ mW/K}^3 \cdot \text{cm}$ (Fig. 14). The T^2 term is likely to be attributed to phonons (diffused by electrons) whereas the αT term represents the electronic contribution. Thus, at T_c , around 50 % of the thermal conductivity is still mediated by the phonons (the resistivity of UBe_{13} at 1 K is several orders of magnitude higher than that of ordinary metals). This part should decrease at lower temperatures because the mean free path of the phonons should then be limited by the size of the crystallites (with $\beta T^2 = 1/3 C_p v_s \ell$, one can estimate that ℓ is already $1 \mu\text{m}$ at 1 K). Thus, even though the general shape of $\kappa(T)$ between 0 K and T_c might be difficult to interpret, it appears reasonable to assume that below 300 mK, κ is essentially controlled by the electronic contribution. Then, following [33], one can compare the effect of the impurities on C_p and κ at low temperatures in the superconducting state. For an axial state, we determined, from the specific heat measurements, a pair breaking parameter $\gamma = [\gamma_0 T_c / C_N(T_c)]^2 / \pi / 2 = 0.006$. Such a value of γ gives : $\frac{\alpha_0}{\alpha} = \frac{3 \pi}{4}$ $\gamma = 1.4 \times 10^{-2}$ where

Fig. 14. — Temperature dependence of the thermal conductivity κ below 4.2 K at $H = 0$. Insert is the Lorentz number after subtraction of the phonon contribution to the thermal conductivity.

α_0 is the coefficient of the residual linear T term of κ at low temperatures. Thus, α_0 should be very small, of the order of $4 \mu\text{W/K}^2\text{cm}$. Experimentally, no linear T term has been observed on this sample down to 30 mK (Fig. 15), allowing us to put an upper limit on α_0 of $6 \mu\text{W/K}^2\text{cm}$. This is another confirmation that the phase shift of the impurities is very near $\pi/2$, since the calculations predict a drastic increase of α_0 when this phase shift deviates from $\pi/2$ [29, 31].

Let us emphasize that these low values of the pair breaking parameter γ imply very high values of the mean free path ℓ of the quasiparticles, of the order of $10^5 \text{ \AA}$ [34]. On the one hand, it clearly rules out the values of ℓ of a few angströms deduced from free electron models. This is consistent with what had been previously discussed [25] on estimates of the mean free path from the resistivity above T_{coh} and use of recent band calculations [35], showing that UBe_{13} was in the clean rather than dirty limit. On the other hand, $10^5 \text{ \AA}$ would still be more than one order of magnitude higher than the more optimistic estimate of ℓ , even if the value of the residual resistivity were taken in these estimates (instead of the high temperature value).

In previous experiments on a less pure sample ($\rho_0(H \rightarrow \infty) \sim 20 \mu\Omega\text{cm}$) [32], residual linear temperature terms in C and K were observed. $\gamma_0 \sim 110 \text{ mJ.mole}^{-1} \text{ K}^{-2}$ and $\alpha_0 = 30 \mu\text{W/K}^2\text{cm}$. The emergence of also a non vanishing α_0 coefficient is directly connected with an increase of the pair breaking parameter by a factor 2.5 by comparison to the present work. Evidence for resonant impurity scattering was also found in the specific heat of UBe_{13} [36] with a low value of $\gamma_0 \sim 20 \text{ mJ.mole}^{-1} \text{ K}^{-2}$ up to a rather high temperature ($T = 180 \text{ mK}$). Progress must be made in the purity of the bare lattice for selecting the nature and the content of the defects. There are clear experimental proofs that the regime of a strong interaction between impurities and the superconducting phase appears only at very low

Fig ; 15. — At $H = 0$, the temperature variation of K/T shows the weakness of any linear temperature term by contrast to the effect reported on figure 13 for C/T .

temperatures. The specific heat in the temperature range (150 mK-500 mK) has a T^3 variation with no supplementary linear temperature contribution.

Conclusion.

In this paper an attempt was made for a coherent interpretation of the high and low temperature data of magnetoresistivity and magnetization in UBe_{13} . The properties of the normal and superconducting phases were correlated. At high as well as at low temperatures two different energy scales occur. Even if UBe_{13} is thought to be a Kondo-lattice, magnetic correlations should exist and play an important role in its behaviour. A « classical » Fermi liquid picture has been proposed for the $A(H)T^2$ term of ρ . The field variation of A could be related to the unusual shape of the upper critical field while the large field dependence of the residual resistivity underlines the strong interplay of defects with the lattice properties. Microscopically, there is a need for inelastic neutron measurements. Notably, subtle effects between antiferromagnetic and ferromagnetic couplings (neglected here) may play an important role in UBe_{13} .

Acknowledgements.

One of us (JPB) thanks Profs. P. Wölfle and P. Hirschfeld for stimulating discussions. JF is deeply indebted to Prof. J. Friedel for his continuous support in our « experimental approach ».

References

- [1] BATLOGG B., BISHOP D. J., BUCHER E., GOLDING B. Jr, RAMIREZ A. P., FISK Z., SMITH J. L. and OTT H. R., *J. Mag. Mag. Mat.* **63 & 64** (1987) 441.
- [2] SCHLOTTMANN P., *Z. Phys. B* **51** (1983) 223.

- [3] OTT H. R., RUDIGER H., FISK Z. and SMITH J. L., in *Moment Formation in Solids*, W. J. L. Buggs Ed. (Plenum, New York) p. 305.
- [4] TROC R., TRZEBIATOWSKI W. and PIPREK K., *Bull. Acad. Pol. Sci., Ser. Sci. Chim.* **19** (1971) 427.
- [5] REMENYI G., JACCARD D., FLOUQUET J., BRIGGS A., FISK Z., SMITH J. L. and OTT H. R., *J. Phys. France* **47** (1986) 367.
- [6] WUILLOUD E., BAER Y., OTT H. R., FISK Z. and SMITH J. L., *Phys. Rev. B* **29** (1984) 5228.
- [7] FELTEN R., STEGLICH F., WEBER G., RIETSCHEL H., GOMPF F., RENKER B. and BEVERS J., *Europhys. Lett.* **2** (1986) 323.
- [8] BESNUS M. J., PANISSOD P., KAPPLER J. P., HEINRICH G. and MEYER A., *J. Mag. Mag. Mat.* **31-34** (1983) 227 and
BUCHER E., MAITA J. P., HULL G. W., FULTON R. C., COOPER A. S., *Phys. Rev. B* **11** (1975) 440.
- [9] DE VISSER A., FRANSE J. J. M. and FLOUQUET J., Contribution to the international symposium on magnetoelasticity and electronic structure of transition metal alloys and films, Duisburg (1989), to be published.
- [10] NEUMANN K. U., CAPPELMANN H., FISK Z., SMITH J. L. and ZIEBECK K. R. A., *Solid State Commun.* **60** (1986) 641.
- [11] BRISON J. P., BRIGGS A., LAPIERRE F., FLOUQUET J., FISK Z. and SMITH J. L., *J. Mag. Mag. Mat.* **76-77** (1988) 243.
- [12] BRISON J. P., Thesis, Grenoble 1988, unpublished.
- [13] RAUCHSCHWALBE U., STEGLICH F. and RIETSCHEL H., *Physica* **148B** (1987) 33.
- [14] See JACCARD D., SIERRO J., BRISON J. P. and FLOUQUET J., published in Proceedings of ICM of Paris, *J. Phys. Colloq. France* **49** (1988) C8-741.
- [15] BRISON J. P., RAVEX A., FLOUQUET J., FISK Z. and SMITH J. L., *J. Mag. Mag. Mat.* **76-77** (1988) 525.
- [16] See FISK Z., OTT H. R. and AEPPLI G., *Jpn J. Appl. Phys.* **26** (1987) 1882.
- [17] MAYER H. M., RAUCHSCHWALBE U., BREDEL C. D., STEGLICH F., RIETSCHEL H., SCHMIDT H., WÜHL H. and BEVERS J., *Phys. Rev. B* **33** (1986) 3168.
- [18] OTT H. R., RUDIGIER H., FELDER E., FISK Z. and SMITH J. L., *Phys. Rev. B* **33** (1988) 126.
- [19] See KAVEH M. and WISER N., *Adv. Phys.* **33** (1984) 257.
- [20] UEDA K., *J. Phys. Soc. Jpn* **43** (1977) 1497.
- [21] USAMI K., *J. Phys. Soc. Jpn* **45** (1977) 466.
- [22] THOMPSON J. D., MC ELFRESH M. W., WILLIS J. O., FISK Z., SMITH J. L. and MAPLE M. B., *Phys. Rev.* **35** (1987) 48.
- [23] MAO S. Y., JACCARD D., SIERRO J., FISK Z. and SMITH J. L., *J. Mag. Mag. Mat.* **76-77** (1988) 241.
- [24] PHILLIPS N. E., FISHER R. A., FLOUQUET J., GIORGI A. L., OLSEN J. A. and STEWART G. R., *J. Mag. Mag. Mat.* **63-64** (1987) 332.
- [25] BRISON J. P., FLOUQUET J. and DEUTSCHER G., to be published in *J. Low Temp. Phys.* (1989).
- [26] TOURNIER R., SULPICE A., LEJAY P., LABORDE O. and BEILLE J., *J. Mag. Mag. Mat.* **76-77** (1988) 552.
- [27] FLOUQUET J., HAEN P., LAPIERRE F., FIERZ C., AMATO A. and JACCARD D., *J. Mag. Mag. Mat.* **76-77** (1988) 285.
- [28] PETHICK C. J. and PINES D., *Phys. Rev. Lett.* **57** (1986) 118.
- [29] HIRSCHFELD P., VOLLHARDT D. and WÖLFLE P., *Solid State Commun.* **59** (1986) 111.
- [30] SCHMITTRINK S., MIYAKE K. and VARMA C. M., *Phys. Rev. Lett.* **57** (1986) 2575.
- [31] HIRSCHFELD P., WÖLFLE P. and EINZEL D., *Phys. Rev. B* **37** (1988) 83.
- [32] BRISON J. P., LASJAUNIAS J. C., RAVEX A., FLOUQUET J., JACCARD D., FISK Z. and SMITH J. L., *Physica C* **153-155** (1988) 437.
- [33] WÖLFLE P., *J. Mag. Mag. Mat.* **76-77** (1988) 492.
- [34] HIRSCHFELD P. and WÖLFLE P., private communication.
- [35] NORMAN M. R., PICKETT W. E., KRAHAUER H. and WANG C. S., *Phys. Rev. B* **36** (1987) 4058.
- [36] OTT H. R., FELDER E., BRUDER C. and RICE T. M., *Europhys. Lett.* **3** (1987) 1123.