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Title
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Permalink
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Journal
EPL (Europhysics Letters), 46(17)

ISSN
0295-5075

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Publication Date
1985

DOI
10.1051/jphyslet:019850046017081100

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J. Physique Lett. **46** (1985) L-811 - L-817

Classification
Physics Abstracts
72.15E — 72.15J — 74.30E

Thermal conductivity and thermoelectric power of UBe_{13}

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(*Reçu le 8 avril 1985, accepté sous forme définitive le 8 juillet 1985*)

Résumé. — Des mesures de résistivité, pouvoir thermoélectrique et de conductivité thermique du composé à fermions lourds UBe_{13} ainsi que leurs relations avec le diagramme de phase température-champ critique sont présentées. A basse température ($T < 1$ K), le pouvoir thermoélectrique de la phase normale est gigantesque. La conductivité thermique suit une loi en T^2 dans la phase supraconductrice sans contribution linéaire en T contrairement à ce qui est observé dans UPt_3.

Abstract. — Measurements of the resistivity, thermoelectric power and thermal conductivity of the heavy fermion compound UBe_{13} and their relations to the temperature-upper critical field phase diagram are reported. At low temperature ($T < 1$ K), enormous thermoelectric powers are detected in the normal phase. The thermal conductivity obeys a T^2 law in the superconducting phase with no supplementary T contribution which is not observed in the other heavy fermion compound UPt_3.

1. Introduction.

The discovery of superconductivity in UBe_{13} at $T_c \sim 0.9$ K [1] has revived the debate on the origin of the superconductivity in heavy fermion compounds first found in $CeCu_2Si_2$ [2]. The heavy fermion character of UBe_{13} is proved by the large value of the ratio of the specific heat C to the temperature T near T_c : $C/T \sim 1$ J/mole/K^2 [3]. One of the peculiarities of UBe_{13} is that the electron mean free path is extremely short near T_c. At 1 K, the electrical resistivity ($\rho \sim 190$ μΩ cm) [1-4] is almost that estimated for the unitary limit; no sign of Fermi liquid behaviour ($\rho \sim T^2$) is detected. By contrast with liquid ^{3}He for which the superfluid transition ($T_c \sim 2$ mK) is far below the Fermi temperature T_F [5], the definition of T_F is not obvious.

Article published online by EDP Sciences and available at http://dx.doi.org/10.1051/jphyslet:019850046017081100

To clarify the specific case of UBe_{13}, a study of transport properties will be reported. Measurements of the thermoelectric power Q provide a sensitive method for mapping the density of electronic states and simultaneous measurements of the thermal conductivity are performed in both normal (N) and superconducting phase (S).

2. Experimental set up.

The single crystalline UBe_{13} sample has been prepared as described in reference [1]. The sample was a parallelepiped with cross sectional area 0.63 mm^2 and length 1.5 mm. The temperature gradient ∇T is measured by two 68 Ω Matshushita resistors soldered with indium at each extremity. A four lead measurement gives the heater power. The thermoelectric voltage is detected by a conventional nanovoltmeter manufactured by Tekelec with a sensitivity better than 2 nV.

The experiments are done with magnetic fields (H) up to 75 kOe in a dilution refrigerator cryostat. An auxiliary Matshushita resistor is located in the compensated region of the superconducting magnet for correcting the thermometers for magnetoresistivity. The lowest applied field H, used to quench the superconductivity of indium, is 1 kOe. The magnetic field, the electric current and the thermal gradient were applied along the length of the sample i.e. parallel to a primitive vector of the cubic Bravais lattice.

3. Results.

3.1 RESISTIVITY (ρ). — As previously reported [1], a maximum of paramagnetic scattering $\rho_M \sim 220$ μΩ cm is observed at $T_M^\rho \sim 2.5$ K. By resistivity measurements, the superconducting transitions T_c at 0 and 75 kOe are respectively 854 mK and 220 mK with widths (defined by 10 % and 90 % changes in ρ) equal to 12.5 mK and 58 mK. The upper critical field $H_{c_2}(T)$ derived from resistivity confirms the published phase diagrams with an enormous initial slope $\left.\frac{\partial H_{c_2}}{\partial T}\right|_{T_c} > -270$ kOe/K (regime I for $H < 15$ kOe) followed by a quasi-linear temperature dependence down to 0 K with a slope $\left.\frac{\partial H_{c_2}}{\partial T}\right|_{T \to 0} \sim -98$ kOe/K (regime II for $H > 15$ kOe).

3.2 THERMOELECTRIC POWER (Q). — Figure 1 shows the temperature variation of Q for different applied fields. In low fields ($H \sim 1$ kOe), the striking features are the enormous value of Q (~ -24 μV/K) at T_c^+ just above T_c and the increase of its amplitude on cooling. The proportionality of Q to T that is usual for metal at low T is not found. If the normal state could persist at 0 K, Q would pass through a minimum below 0.8 K. Qualitatively, this behaviour implies the occurrence of a structure in the density of states with a characteristic energy $\lesssim 1$ K.

Another interesting result is that, in the regime I of the phase diagram, $\frac{\partial Q}{\partial T} > 0$ while in II, $\frac{\partial Q}{\partial T} < 0$. Furthermore, the field dependence of $Q(T_c^+)$ is quasi-linear in H with an extrapolation to zero at $H = 78$ kOe $\lesssim H_{c_2}(0)$ (insert b); the superconductivity is destroyed when $Q(T)$ becomes positive for $H > H_{c_2}(0)$. In $CeAl_3$ [6], positive thermopower has been observed at low temperatures just when ρ follows a T^2 dependence.

3.3 THERMAL CONDUCTIVITY (κ). — Figure 2 presents the thermal conductivity at two different fields $H = 1$ kOe and 75 kOe. For $H = 75$ kOe, κ follows a linear temperature dependence in the normal phase well below T_M^ρ.

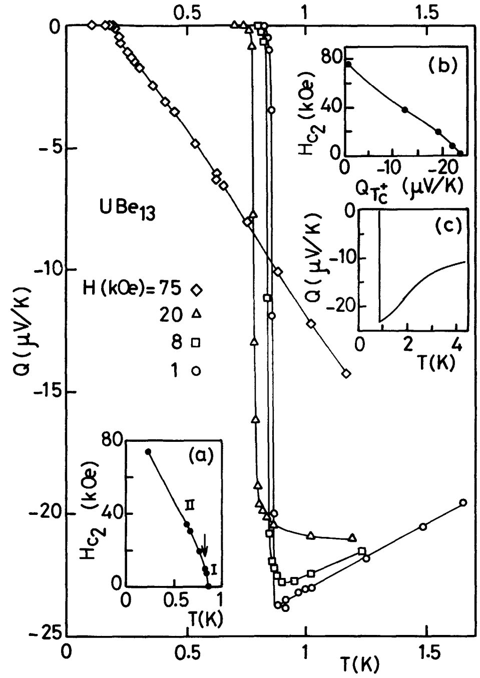

Fig. 1. — Thermoelectric power Q of UBe$_{13}$ as a function of T for $H = 1$, 8, 20 and 75 kOe. Insert (a) is the $H_{c_2}(T)$ phase diagram obtained by resistivity measurements, insert (b) shows a phase diagram drawn with $Q(T_c^+)$ as the variable and insert (c) is for $H = 1$ kOe, $Q(T)$ up to 4.2 K.

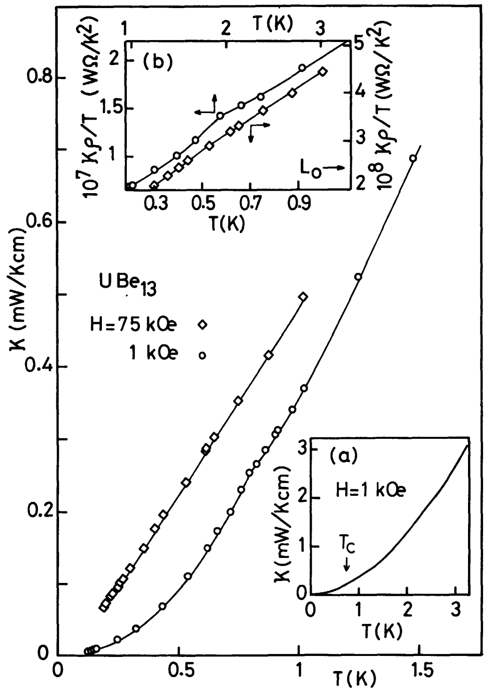

Fig. 2. — Thermal conductivity κ at $H = 1$ and 75 kOe. Insert (a) shows the variation of κ at $H = 1$ kOe up to 3.5 K, and insert (b) the quasilinear T dependence of the « Lorentz number » $\kappa\rho / T$.

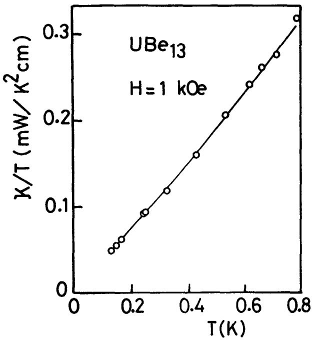

Fig. 3. — In the superconducting phase of UBe_{13}, κ/T as a function of T.

At 1 kOe, a kink in κ is observed at 0.79 K i.e. 65 ± 5 mK below $T_c(\rho)$. This discrepancy is probably due to the inhomogeneity of the sample. In the superconducting state almost up to T_c, κ_s has a T^2 dependence as shown in figure 3. The contribution of the phonons is difficult to establish with certainty. However, a high Debye temperature ($\theta_D \sim 840$ K) [7] and the imperfections of the lattice limiting the phonon mean free path favour a weak contribution below 1 K. Above 1 K, an experimental proof of the importance of the electronic contribution is already given by the fact that the maximum in ρ at $T_M^\rho \sim 2.5$ K is connected with a change in the thermal conductivity at $T = 2.2\ \text{K} \sim T_M^\rho$.

The measurements of κ and ρ permit a comparison of their product with the well-known Wiedemann-Franz law observed for elastic scattering processes of fermions :

$$\rho_0 \kappa_0 = L_0 T$$

with $L_0 \sim 2.45 \times 10^{-8}\ \text{W}\Omega\text{K}^{-2}$. As shown in the insert b of figure 2, $\kappa\rho/T$ shows quasi-linear temperature dependences in low field ($H \sim 1$ kOe) and high field ($H = 75$ kOe) : at 1 K, respective values are $6.8 \times 10^{-8}\ \text{W}\Omega\text{K}^{-2}$ and $4.5 \times 10^{-8}\ \text{W}\Omega\text{K}^{-2}$ for $H = 1$ kOe and 75 kOe.

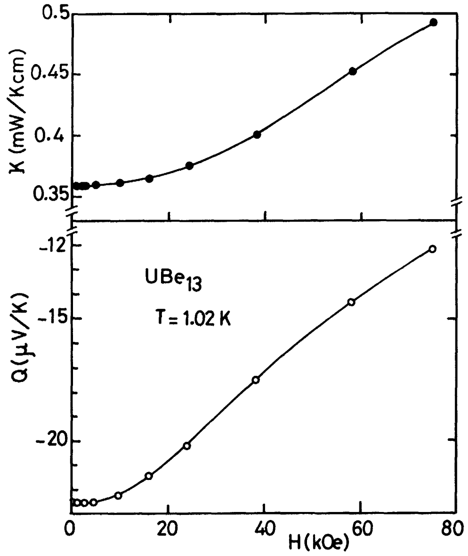

Fig. 4. — Field dependence of κ and Q at 1.02 K.

This unusual behaviour points out that, in UBe_{13}, the carriers of electricity and heat may differ or that, as it has been emphasized in magnetoresistivity experiments [8], below 100 kOe, there is a strong interplay between Fermi liquid and scattering disorder phenomena; when, at low temperatures, the ρ data is analysed with residual plus T^2 terms, different field dependence must be given to the two contributions [8].

Figures 4 and 5 reproduce the field dependence of κ and Q at 1.02 K and at 655 mK. In the normal phase, the low field variation of κ and Q is weak; a field sweep at 655 mK shows a quasi quadratic dependence in H below H_{c_2} and a linear variation above H_{c_2}.

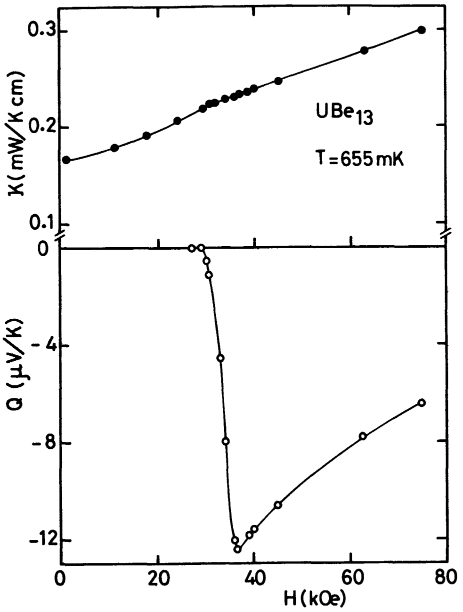

Fig. 5. — Field dependence of κ and Q at 655 mK.

4. Discussion.

The quasi-linear temperature dependence of the Lorentz number $\kappa\rho/T$, the possible occurrence at low field of a minimum in Q below T_c and the huge value of the resistivity indicate that, in UBe_{13}, the Fermi liquid regime is far from being achieved at T_c. In low fields, clearly a large disorder occurs; in regime I, the scattering disorder may reinforce the superconducting state as suggested for weakly localized, nearly magnetic systems [9].

In the superconducting state, the T^2 dependence of κ appears to be another demonstration of unusual superconducting pairing [10, 11]. Such a variation has been predicted for a polar-like state in odd pairing [12, 13]. However group-theoretical arguments seem to reject such a state [14, 15], although it may be allowed in the presence of spin anisotropy in the pairing interactions [13].

In many aspects, UBe_{13} is an unusual heavy fermion compound. The exotic heavy fermion properties are usually observed for compounds with a strong axial symmetry ($CeAl_3$, $CeCu_2Si_2$, UPt_3) and not for cubic materials like UBe_{13}. An interesting idea is that the hybridization matrix elements are extremely anisotropic and that the cubic symmetry can be spontaneously broken [7]. Thermal conductivity experiments must be performed below 100 mK to test the picture of strong anisotropy of the energy gap with BCS singlet pairing [7].

It is worthwhile to mention that the three well-known heavy fermion superconductors have quite different magnitudes of the resistivity at T_c^+ (or of their extrapolated value $\rho(0)$ at $T \rightarrow 0$), of the thermoelectric power at T_c^+ and of the ratio of T_c by T_M^ρ when a clear maximum can be observed in resistivity data (Table I). There is no doubt [17] that UPt_3 is a clean superconductor with an electronic mean free path ($l \sim$ 3 000 Å) higher than the coherence length ($\phi_0 \sim$ 100 Å) whereas crude estimates of $l \leqslant$ 30 Å and $\phi_0 \sim$ 50 Å in UBe_{13} classify it as a dirty superconductor [4].

Table I. — *Resistivity $\rho(T_c^+)$ and thermoelectric power $Q(T_c^+)$ just above T_c, ratio of the superconducting transition T_c to the temperature T_M^ρ of the maximum in resistivity and estimated residual resistivity.*

	$\rho(T_c^+)$ µΩ cm	$Q(T_c^+)$ µV/K	T_c/T_M^ρ	$\rho(0)$ µΩ cm	Ref.
UBe_{13}	~ 180	− 24	0.36	40	[1, 4, 8]
$CeCu_2Si_2$	~ 15	− 3	0.03	5	[16]
UPt_3	~ 1.5	+ 0.5		< 0.6	[17]

At the difference of this diversity, the thermal conductivity of the three heavy fermion compounds follows

$$\kappa_s = \alpha T^2 + \beta T \quad \text{for} \quad T < T_c .$$

The parameters α and β are reported in table II. For UBe_{13}, a quadratic term alone is observed. Renormalization by the thermal conductivity at T_c gives a dimensionless parameter $\alpha T_c^2/\kappa(T_c)$ almost equal to 1 for UBe_{13} and $CeCu_2Si_2$. This suggests a similarity between both compounds which has already been observed in the temperature dependence of their specific heats [18] (a T exponent upper than two has been found for UBe_{13} and $CeCu_2Si_2$ [18] while T plus T^2 terms have been reported for UPt_3 [19, 20]). In our opinion, the linear temperature term in κ observed in $CeCu_2Si_2$ is a parasitic effect coming from the difficulty of producing a homogeneous sample of the required size; the $CeCu_2Si_2$ reported results are on a dense unannealed sample with a low content of microcracks; the large broadening of the superconducting transition measured by susceptibility [16] suggests that a larger part of the sample stays in a normal state and that the T term has then a parasitic origin.

Table II. — *Coefficients α and β of the T^2 and T term of the thermal conductivity in the superconduction state.*

	α mW/K³ cm	β mW/K² cm	$\kappa(T_c)$ mW/K cm	$\dfrac{\alpha T_c^2}{\kappa(T_c)}$	T_c mK	Ref.
UBe_{13}	0.38	0	0.25	1.08	854	
$CeCu_2Si_2$	2.8	0.7	0.8	1.11	560	
UPt_3	19.5	0.55	3.1	1.70	520	[17]

However, the observation in UPt_3 of a T term in the specific heat well below T_c (at T = 146 mK ~ 0.28 T_c) [19] may suggest that here a normal component could exist in the superconducting phase. The interesting conclusion would be that the βT contribution in κ is an intrinsic effect due to the motion of the normal component. If the gap vanishes along lines on the Fermi surface, only T^2 contributions have been predicted [12]. The intrinsic character of the T terms in UPt_3 is still an open experimental question.

5. Conclusion.

Extensive measurements of transport properties of UBe_{13} show clearly the complexity of this unusual system. A simple result is the T^2 dependence of its thermal conductivity. Important differences exist between the two uranium superconductors UBe_{13} and UPt_3. The possible occurrence of normal component below T_c in UPt_3 is underlined.

Acknowledgments.

The authors thank particularly Dr. N. Phillips, Dr. S. von Molnar and Dr. C. M. Varma for their encouragements and stimulating discussions. They are indebted to Dr. H. F. Braun and P. Lejay for preparing the $CeCu_2Si_2$ and UPt_3 samples. Work at Los Alamos was performed under the auspices of the U.S. Department of Energy.

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