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TRANSPORT PROPERTIES OF HEAVY FERMION SYSTEMS

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Abstract : Taking three examples of different ground states (non magnetic, magnetic and superconducting), the powerful tool of transport measurements to detect the different regimes of heavy fermion compounds is shown. A special emphasis is given on magnetic field effect (ie the action of a f polarization) and on the interplay between disorder and lattice properties.

Introduction

The terminology of heavy fermion (HF) is associated to the low characteristic electronic temperature arising from the apparent weak delocalization of highly localized particles by their resonant coupling with itinerant electrons ; above a few degrees Kelvin the full entropy of each f electron is recovered [1,2]. A large variety of ground state may appear. $CeAl_3$ is an HF system which stays in a normal state without the occurrence of long range magnetic ordering [3], CeB_6 is magnetically ordered at $T_N \sim 2.3$ K without the appearance of any superconductivity [4] and UBe_{13} becomes superconducting below 0.9 K without long range magnetic ordering [5]. We will describe mostly transport measurements realized on these three examples and their links with thermodynamic data such as specific heat. We will focus particularly on the action of a magnetic field (H) on HF properties. The simple idea is that, in very high fields, the single particle behavior of HF systems must be slowly recovered ie the mass can still be high by renormalization effects but the interactions between heavy particles have been collapsed.

This scheme implies the notion that subtle details (crystal symmetry and related anisotropies of magnetic coupling and electronic properties) define the different ground state. Common properties emerge when the disorder is large, ie in term of entropy S for S > 0.5 Rlog2. Coherence effects appear for $S \lesssim 0.15$ Rlog2 ; in non magnetic ground state, they correspond to Fermi liquid (FL) properties and in magnetic ground state to decoupling between spin waves and FL properties.

Schematically, intermediate valence (IV) materials correspond to strong mixing of the f particles with the itinerant electrons. This leads to a collapse of crystal field effects and to a drop of interactions between particles [6]. For IV compounds, interband scattering between heavy and light particles seems the process which dominates the thermal dependence of the thermoelectric power TEP [7] ie at low temperature the inverse of the relaxation time τ is proportional to the large dressed density of states $N_f(E_F)$ of the f electrons at the Fermi level. According to the Mott's formula [8], the TEP, Q, is linear in temperature as in common metal, but proportional to the factor $(\partial \ln N_f/\partial E)_{E_F}$ which can take very large positive or negative values depending on the exact location of the many body resonance near E_F. In this scheme, the corresponding term of the electronic specific heat and of the resistivity are linear in temperature with a proportionality constant equals to the density of states N_f of the heavy particles. At a temperature T_m, which approximatively corresponds to the width W of the many body resonance, the TEP shows a maximum. At a first approximation, T_m scales the density of states N_f. This crude picture explains rather well the TEP data of Ce and Yb IV compounds [7,9]. Coherence effects appear clearly in T^2 resistivity law ; this assumes a collapse of the reservoir making the interband transitions. The same mechanism will appear in HF compounds with new additions of crystal field effects and at low temperature of interactions between particles [10].

1. $\underline{CeAl_3}$

For instance, three characteristic regimes appear in the absolute TEP of $CeAl_3$ measured over almost four temperature decades (Fig. 1). At a first approximation the contribution of the electron-phonon scattering can be neglected in this compound. Due to the large sensitivity of the TEP to the energy-dependence of the relaxation time in the vicinity of the Fermi level, each characteristic temperature is associated with a TEP peak. In a rare earth HF system, the local character of the 4f atom seems preserved, notably the crystal field effects, but the d-f hybridization leads to a narrow many body resonance at the Fermi level reminiscent of the single impurity Kondo effect. For $CeAl_3$ typical width of the d-f Kondo resonance is $T_K \sim 5$ K, as deduced by neutron scattering experiment. The high temperature TEP anomaly centered at ~ 60 K is observed in the temperature region of the -lnT variation of the resistivity. This high temperature anomaly has been ascribed to an interplay of crystal field and Kondo effects, while the low temperature negative TEP peak at ~ 4 K is reminiscent of the Kondo resonance of each ion [10].

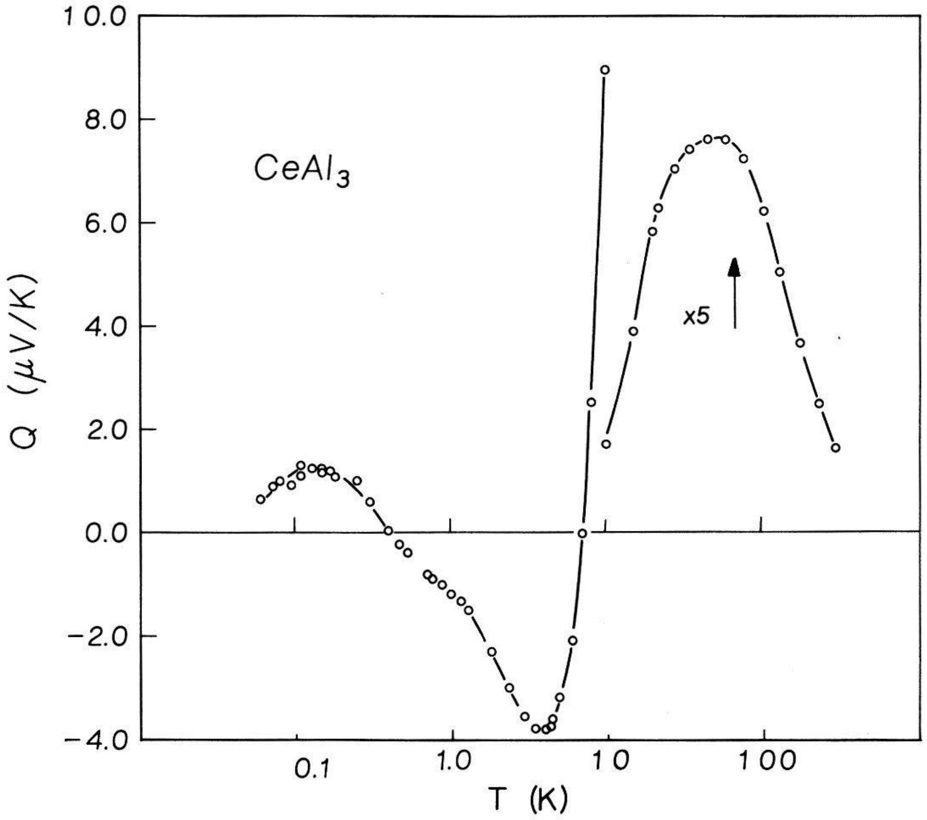

Fig. 1: Thermoelectric power Q of $CeAl_3$ over almost 4 decades of temperatures.

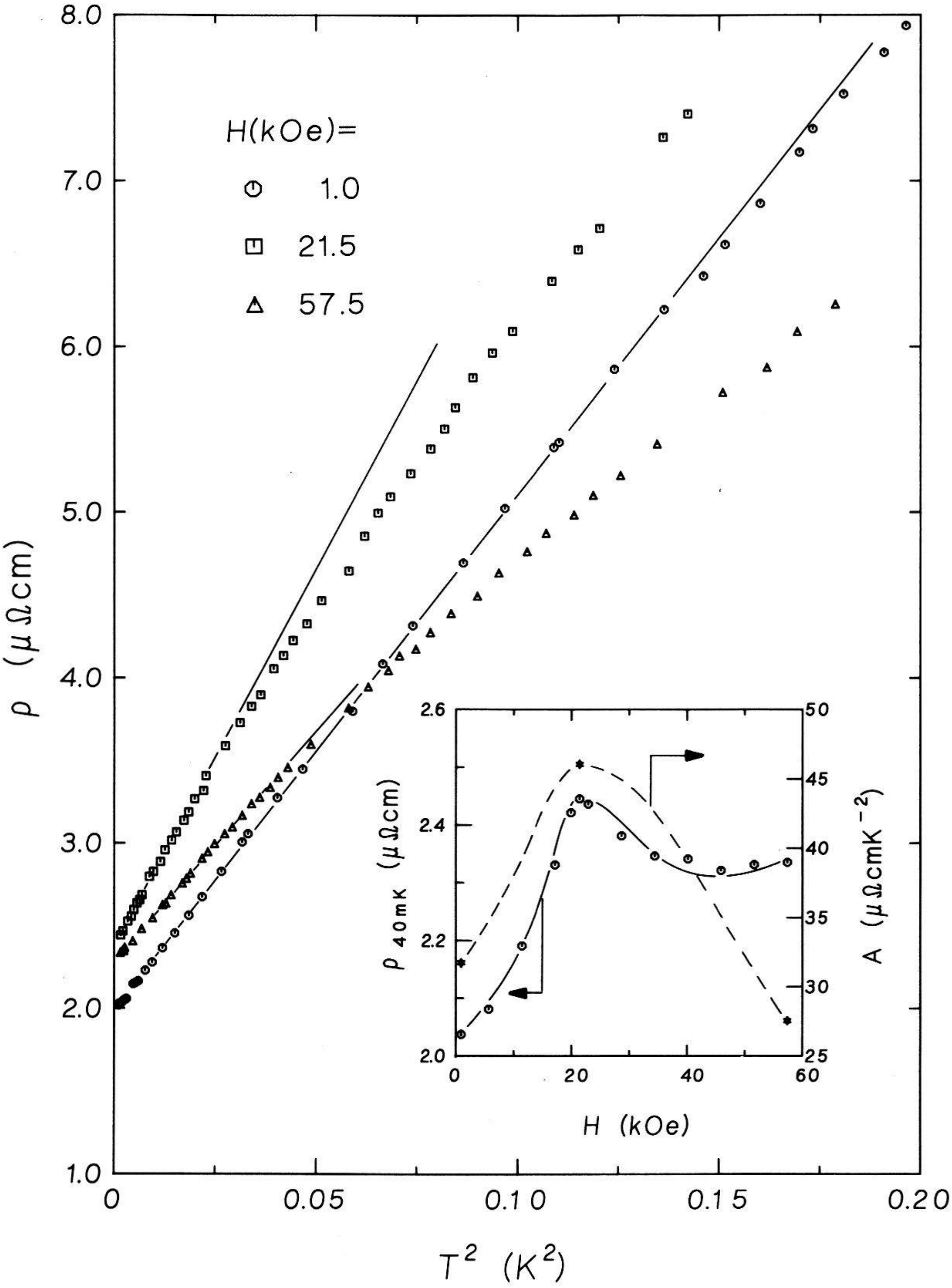

Fig. 2: Resistivity (ρ) of $CeAl_3$ at very low temperature for three different fields. Insert : field variation of the residual resistivity ρ_0 and of the A coefficient of the resistivity law : $\rho(H) = \rho_0 + AT^2$.

At still lower temperature, Bloch waves develop in the lattice and a coherence effect is observed in the resistivity. At a temperature $T^* \cong 0.3$ K, the system enters in a strongly interacting Fermi liquid as shown for instance by the simultaneous thermodynamic measurements [10] of the entropy and its pressure derivative. Again, near this temperature T^*, a TEP peak appears. T^* describes the interaction between heavy particles and is about an order of magnitude smaller than T_K which describes the coupling of 4f electrons with the Fermi sea. Furthermore the Kondo resonance can be viewed as an almost uncorrelated band with interband transitions. Such transitions may be induced by uncorrelated magnetic 4f moments, ie spatial fluctuations of the d-f hybridization when all the sites are non-equivalent [11]. Consequently, in the temperature window $T^* < T < T_K$, the TEP and the resistivity are expected to be linear in T. For our $CeAl_3$ sample, Q is linear in T between 1 and 4 K while the resistivity is proportional to T to better than 1 % between 0.6 and 1.4 K.

It should be noted that this last temperature range is rather narrow but it corresponds to a strong increase of the resistivity from about 12 to 36 μΩcm. Even wider temperature region for the linear T resistivity regime has been observed in $CeCu_2Si_2$ [12].

Let us now focus on the very low temperature transport properties when the system achieves the coherent Fermi liquid regime. At $T < T^*$ the electron-electron interactions dominates leading to a relaxation time $\tau \sim T^{-2}$ according to Fermi statistics. Fig. 2 shows that the resistivity of $CeAl_3$ follows the well known T^2 law up to T^* when the measurements are performed at zero magnetic field (H = 1 kOe in our experience to destroy the superconductivity of the indium contact). Under magnetic field however, the T^2 law is still obeyed but over smaller temperature range, and the insert of Fig. 2 shows that the residual resistivity as well as the T^2 coefficient A take maximum values at a field $H^* \sim 21$ kOe. This field H^* can be regarded as the field analogy of T^* and coincides with the field maximum of the effective mass ($m^* \sim \sqrt{A}$) as deduced from specific heat measurements. Applying a field of the order of H^* will quench in part the interaction between heavy particles.

Usually, the resistivity is written according to the Matthiessen's rule $\rho = \rho_0 + AT^2$ with the assumption that the residual ρ_0 and AT^2 contributions are due to independent scattering events. However, the similar field behavior of ρ_0 and A asks questions about such a rule. Large field variations of ρ_0 has already been measured in UBe_{13} and UPt_3 [13].

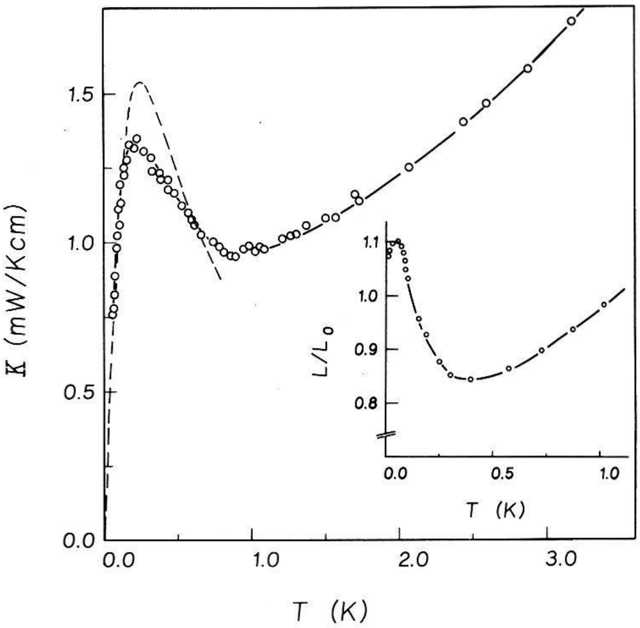

Fig. 3: Thermal conductivity of $CeAl_3$ as a function of the temperature. Dashed line : see text. Insert : experimental Lorentz number.

For $CeAl_3$, more experimental informations can be found by the analysis of the thermal conductivity κ reported in Fig. 3. By defining a Lorentz number $L = \kappa\rho/T$ it appears that, above 1 K, L is larger than $L = (\pi^2/3)(k_B/e)^2$ the free electron Lorentz number. A possible explanation for this departure is that the phonon contribution becomes important at high temperature. The same observation was made for other HF compounds (UBe_{13} and CeB_6) [10]. Below 1 K, the phonon contribution to κ is negligible, and the thermal conductivity increases on cooling down to 200 mK where it reaches a maximum. Using elementary kinetic theory, one may deduce a thermal conductivity $\kappa \sim T^{-1}$ since the specific heat C and κ are directly related by the expression: $\kappa = \frac{1}{3} C v_F \tau$ where v_F is the Fermi velocity. An usual thermal conductivity $\kappa \sim T$ due to the limitation of the electronic mean free path by static impurities, is recovered only below 50 mK.

Assuming the validity of the Wiedemann-Franz law $\kappa\rho = L_0 T$ and the resistivity as $\rho = \rho_0 + AT^2$, the thermal conductivity κ is calculated (dashed line in Fig. 3) using the experimental values $\rho_0 = 1.985\ \mu\Omega cm$ and $A = 31.7\ \mu\Omega cmK^{-2}$. This is equivalent to write $\kappa^{-1} = \kappa_{ie}^{-1} + \kappa_{ee}^{-1}$ where $\kappa_{ie} = L_0 T/\rho_0$ is due to electron-impurity scattering and $\kappa_{ee} = L_0/AT$ to electron-electron scattering. The deviation of the experimental from the calculated curve reflects that the scattering is partly inelastic ($L \neq L_0$) and that above 0.35 K the AT^2 contribution overestimates the resistivity. However, assuming $\rho_0 \to 0$ for an ideal crystal, the divergence of $\kappa_{ee} \sim T^{-1}$ as $T \to 0$ is questionable and suggests that impurity and electron scattering are not independent. It would then be interesting to compare thermal conductivity data for samples of lower residual resistivity.

The insert of Fig. 3 shows that the Lorentz number L goes through a minimum at T $\sim$ 0.5 K. An extrapolation to T = 0 K of the data taken above 0.5 K gives $L \cong 0.75\ L_0$; this deviation from L_0 emphasizes the importance of inelastic processes. It is worthwhile to mention that in the coherent regime of light and heavy particles (basically the Baber mechanism), L equals to 0.65 L_0 [14].

Qualitatively, below 0.5 K, the increase of L is due to the entrance in a regime where the contribution of static impurities to the scattering increases as T decreases ($\rho_0 > AT^2$) : L should tend to L_0 when $T \to 0$ K. However the interesting feature is that L reaches a weak maximum $L_M = 1.1\ L_0$ at 50 mK and then a value 1.07 L_0 at the lowest temperature. This last maximum seems much larger than the experimental accuracy of about 2 %. Its origin is still unknown ; it may be due to spurious effects of a low content of parasitic phases.

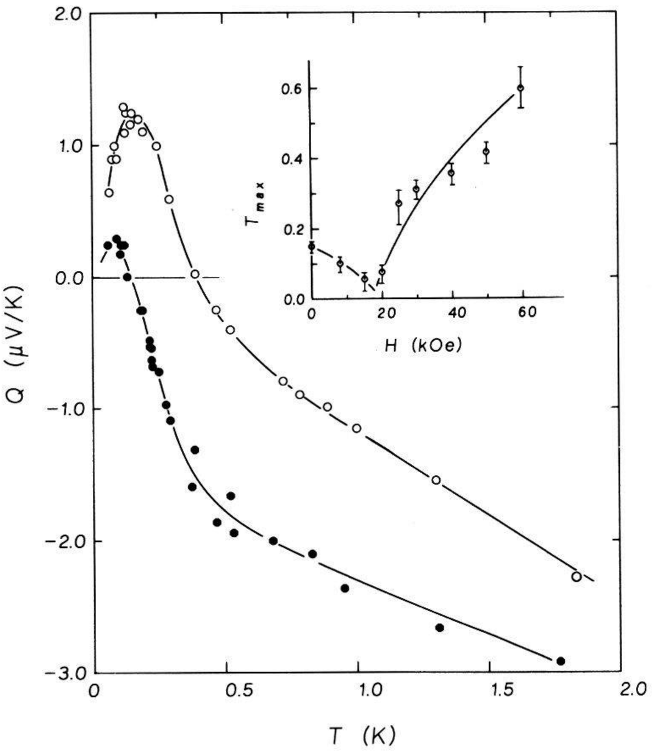

Fig. 4: Thermoelectric power of two samples of $CeAl_3$. Insert : field dependence of the temperature maximum of Q.

The low temperature TEP of two samples of $CeAl_3$ with different residual resistivity are represented in Fig. 4. For sample I, the resistivity $\rho = \rho_0 + At^2$ with $\rho_0 = 5\ \mu\Omega cm$ and $A = 30\ \mu\Omega cm$ and the TEP data (open symbols) are those of Fig. 1. The dotted points in Fig. 4 correspond to sample II which resistivity ($\rho_0 = 1.985\ \mu\Omega cm$) and thermal conductivity have been discussed before.

For sample I, the zero of Q at 0.4 K near T^* coincides with the maximum of C/T suggesting a $(\partial N/\partial E)_{E_F}$ dependence of Q. Assuming that the temperature variation of C/T mimics the energy dependence of the density of states, one evaluates, from the Mott's formula after normalisation by the factor $AT^2/(\rho_0 + AT^2)$, a value of $Q \sim 1.5\ \mu V/K$ at $T = 0.1\ K < T^*$, in good agreement with the experiment [9]. Below T^*, this evaluation is however suspect since the dressed particles have d and f character. The cerium ions behave coherently, no auxiliary energy reservoir coming from uncorrelated 4f moments can be invoked for inducing d-f transitions. Since in a first approximation, the resistivity and the thermal conductivity can be decomposed in an impurity term and an electron term, the Nordheim-Gorter rule should be qualitatively obeyed for the TEP. This should justify the normalisation factor $AT^2/(\rho_0 + AT^2)$. However, for sample II, such a rule predicts a larger TEP peak than in sample I, while the experimental result is just the reverse. Additional difficulties arise for sample II as the linear TEP observed above 1 K do not extrapolate to $Q = 0$ for $T \to 0$.

In a magnetic field, the temperature T_{max} of the maximum of Q is reported in the insert of Fig. 3 for sample I. The result for sample II is qualitatively similar. At low field T_{max} decreases, reproducing the field enhancement of the effective mass. This is compensated in high field by the usual Zeeman splitting of the band. As for the resistivity a crossover field $H^* = 20$ kOe appears. Lastly, it is noteworthy that high magnetic field induces positive TEP.

Our data show that the transport properties ρ, κ and Q of the archetype non magnetic compound $CeAl_3$ reveal characteristic features at the entrance of the strongly interacting electron regime. Such effects are mainly observed at magnetic field lower than H^*. With a strong analogy with (H,T) phase diagram of magnetic systems, characteristic field (H^*) and temperature (T^*) appear. These crossover parameters are found as well in strongly localized Fermi liquid theory [15] than in spin fluctuation model [16]. Field enhancement of γ has been predicted in the first scheme. The field coincidence of maxima for a term like the residual resistivity characteristic of the disorder and for the A coefficient of the T^2 law characteristic of FL properties of the lattice deserves a particular attention. This suggests the existence of a correlation length changing with the strength of A.

2. CeB_6

This compound is interesting since its number n_e of itinerant electrons is just equal to the number n_m of magnetic carriers. Two magnetic phase transitions occur at T_m = 3.3 K and T_N = 2.3 K [17]. In the antiferromagnetic phase I ($T < T_N$) the ordered moment ($\mu \sim 0.28\ \mu_B$ at T = 1.5 K) is much lower than the expected value for the Γ_8 quartet ground state ($\mu \sim 1.56\ \mu_B$). For $T_N < T <$ Tm, an antiquadrupolar phase II with no spontaneous moment has been observed [18]. At low temperature, the electronic specific heat coefficient $\gamma \cong$ 220 mJ/K^2mole is not so huge for a HF system since the main part of the entropy has dropped at the magnetic ordering. Recently the field dependence of $\gamma(H)$ has been studied [18]. The interesting result is that $\gamma(H)$ shows a large maximum at H ~ 15 kOe, ie at the transition from the antiferromagnetic phase I to the quadrupolar type phase II. In this field transition, the behavior of CeB_6 appears quite similar to the previous situation found for $CeAl_3$.

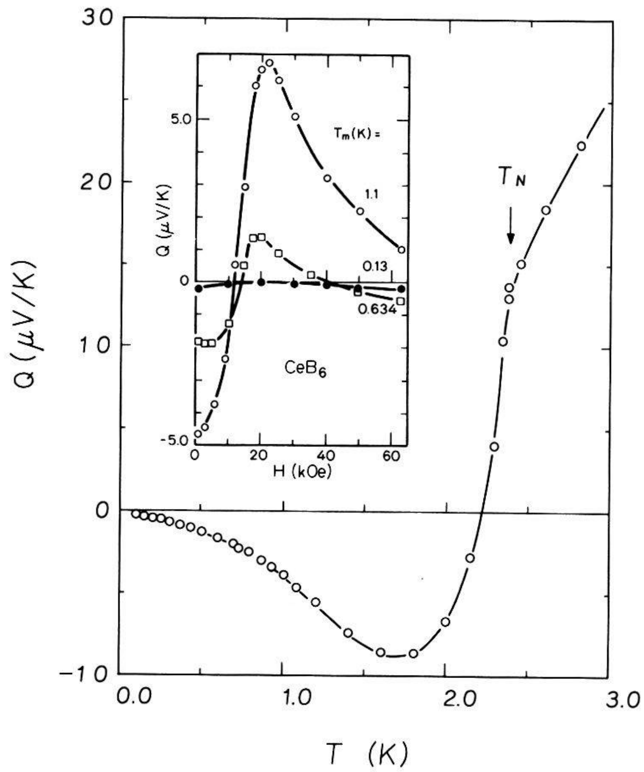

Fig. 5: Temperature dependence of the TEP (Q) of CeB_6 at H = 0. Insert : at constant temperature field variation of Q.

Fig. 5 shows the low temperature TEP of CeB_6 and its field dependence at different temperature. Let us emphasize the kink of Q at T_N, the change to negative value only below T_N and the large hump near 1.7 K with a non-linear extrapolation at $T \to 0$. The kink of Q at T_N may be correlated with $n_e = n_m$ and with the low value of the ordered moment. By comparison no TEP anomaly is detected at the antiferromagnetic transition of compounds like $CeAl_2$ where the almost full 4f moment is ordered.

According the discussion of $CeAl_3$, the negative peak of Q is associated with the presence of renormalized single particles band while the negative curvature at $T \to 0$ could originate in a coherence effect. In a temperature window $T^* < T < T_K$, Q is expected to be dominated by a contribution proportional to $(\partial \ln N_f/\partial E)_{E_F}$ attributed to interband transitions. The insert of Fig. 5 shows this to be the case. The field dependence of the electronic specific heat term $\gamma \sim N_f$ corresponds quite well with the field behavior of the TEP. In particular the maximum of $\gamma(H)$ at the magnetic phase transition coincides with the zero of Q(H). All happens as if the Fermi level in a function of the magnetic field moves across a maximum of the density of states. Furthermore, assuming $\kappa^{-1} \sim \rho \sim N_f$, this crude two band picture also explains the weak minimum observed in the field dependence of the thermal conductivity κ near H ~ 15 kOe [19]. The behaviors of ρ and κ are dominated by the strong scattering of antiferromagnetic magnons. Below T_N, the heat appears to be carried by fermions (large γT term) which are scattered by magnons. A T^{-2} increase of κ on cooling is observed up to T ~ 850 mK [10,13]. At very low temperature the T^2 term observed in the resistivity is mainly intrinsic resulting from energy and momentum conservation in collisions between heavy and light fermions [20]. Microscopically, the direct observation of spin waves in magnetic HF compounds is needed as well as the study of the evolution of the magnetic coupling through T_N.

3. UBe_{13}

The aim to report new measurements of the thermal conductivity of UBe_{13} which becomes superconducting below $T_c \sim 0.9$ K [5] is to underline the importance to reach large value of T_c/T for probing a scaling law. A power law who seems obeyed down a given temperature may describe poorly the experiments when a new attempt is made to increase the T_c/T ratio.

UBe_{13} is an illustrating case of HF superconductors since the pair condensation at T_c occurs far above the regime where T^2 resistivity law is observed and even above the temperature where a minima of the TEP would appear [22]. Just above T_c, the scattering is enormous : $\rho(T_c^+) \sim 100\ \mu\Omega cm$. Furthermore the scattering disorder seems correlated with the characteristics of the superconducting state. In low field, when the residual resistivity ρ_0 is large, the derivative of the upper critical field is high $\frac{\partial H_{c2}}{\partial T}\Big|_{T_c} > 200$ kOe/K whereas, in high field, the linear field decrease of $H_{c_2}(T)$ is associated with a $\sqrt{H}$ decrease of ρ_0. At $H_{c_2}(0)$, a regime change of ρ occurs clearly [23].

Previous thermal conductivity κ measurements down to 0.15 K show a T^2 dependence below T_c [23]. Recently, extensive specific heat and thermal conductivity measurements were performed down to 60 mK [24]. The good quality of the sample was checked in ρ, TEP and susceptibility measurements. Above 150 mK, published data are confirmed [5] : for specific heat C, γ = 800 $mJmole^{-1}K^{-2}$ at T_c, the high specific heat jump $\Delta C/\gamma T_c \sim 2.3$ and $C \sim T^{2.9}$ below T_c. Lowering the temperature down to 60 mK shows a non vanishing $\gamma_0 \sim$ 110 $mJmole^{-1}K^{-2}$ for an extrapolation to 0 K correlated with a linear temperature αT variation of κ. This new low temperature regime has been also found in NMR data [25] : the inverse of the nuclear relaxation time follows a T^3 power law down to 150 mK but recovers a Korringa dependence below 150 mK. Fig. 6 shows the thermal conductivity κ of UBe_{13} below 200 mK : no fit can be made in T^2 as well as in $T+T^2$. It must be underlined that $\gamma_0/\gamma(T_c)$ is of the order of $\alpha T_c/\kappa(T_c)$. Furthermore a subtraction of αT to κ leads to a quasi linear dependence of $\ln((\kappa-\alpha T)/\kappa(T_c^+))$ over three order of magnitude of κ.

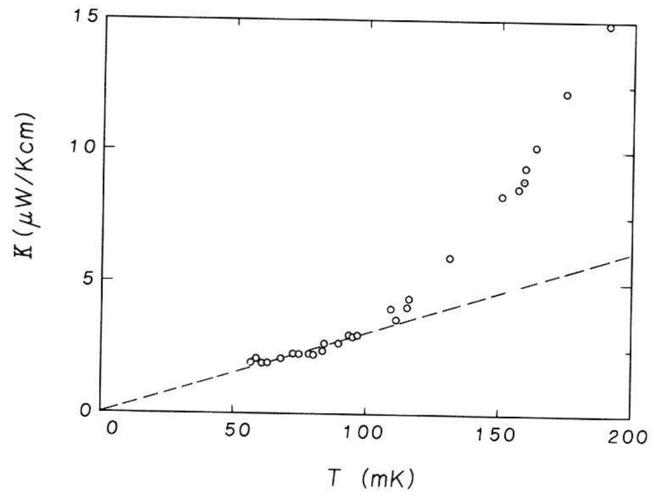

Fig. 6: Thermal conductivity of of UBe_{13} far below its superconducting transition T_c 0.9 K. Dashed line: linear temperature αT dependence of κ.

It is clear that experimental efforts must be performed on the sample quality and on the achievement of high T_c/T ratio for probing the intrinsic power law found in this exotic superconductor. The first claims of T^3 and T^2 law dependence of C and κ appear erroneous [5-22]. Thus, the exact nature of the pairing is still an open question. Let us lastly note that a γ strength of 100 $mJmole^{-1}K^{-2}$ is a typical value found in ordered HF compounds.

Conclusions

Spectroscopic technics will give new microscopic informations on the nature of the ground states. However transport measurements are a nice and sensitive tool to study HF properties. They are particularly useful for probing the different energy scales. In the normal non magnetic phase like $CeAl_3$, interband scattering characteristic of baths of heavy and light particles is observed down to a certain amount of entropy ($S > 0.15$ RLog2) while a strongly interacting FL emerges for $S < 0.15$ RLog2 ; the other striking point is the definition of field and temperature crossover parameter to the FL properties with the initial field enhancement of the effective mass. In the magnetic ordered phases like CeB_6 at low enough temperature the collective spin wave excitations appear decoupled from FL properties.

For superconductivity phases, we have underlined the necessity to take a pure experimental point of view of reaching high T_c/T values and we have shown the detection of residual linear temperature terms in C and κ ; similar results were first found for another HF superconductor UPt_3 [26]. The interplay between disorder properties and HF properties has been shown. Exhaustive measurements and analysis in the same spirit of localization phenomena will be fruitful in this perspective notably to precise the correlation length involved in HF problems. Finally, by comparison to the large amount of experimental works, few theoretical attempts have been made to describe the real conditions of the experiments.

Acknowledgments

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