

# Strong Coupling Effects on the Upper Critical Field of the Heavy-Fermion Superconductor $UBe_{13}$

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*The upper critical field  $H_{c2}(T)$  of a high quality single crystal of  $UBe_{13}$  is studied with very low noise resistivity measurements. It shows a large but finite slope of  $-45 T/K$ , an unusual temperature dependence with an inflexion point at  $T/T_c \sim 0.5$  and a large saturated limit for  $T \rightarrow 0$  of  $H_{c2}(0) = 14 T$ . The complete temperature dependence of  $H_{c2}(T)$  can be described by a simple model of strong coupling superconductor, assuming a full Pauli limitation and the occurrence of a non-uniform superconducting state (FFLO state) at low temperature.*

## 1. INTRODUCTION

$UBe_{13}$  was one of the first heavy fermion systems in which superconductivity was discovered.<sup>1,2</sup> However, there has been little progress in the understanding of the physics of this material, due to the complexity of its normal state and the difficulty of crystal growth. One of the major problems is that the Fermi liquid behaviour is not well established at the superconducting transition temperature ( $T_c \sim 900$  mK):

— the entropy balance of the specific heat ( $C$ )<sup>3,4</sup> reveals that  $C/T$  has to increase, instead of remaining constant, at low temperature.

— the resistivity follows the characteristic behaviour expected for a strongly correlated Fermi liquid, i.e.  $\rho = \rho_0 + AT^2$ , only at temperatures below  $T_c$  (high magnetic fields ( $>8$  T) are required in order to remove superconductivity and observe this behaviour).<sup>5</sup>

In addition, in the normal-state,  $\text{UBe}_{13}$  displays also a very strong negative magnetoresistance<sup>5,6</sup> which is uncharacteristic of other heavy fermion superconductors and of still unknown origin. As for the superconducting state, it was recognised early on that  $\text{UBe}_{13}$  is in the strong coupling regime, as the height of the specific heat jump is large ( $\Delta C/C \sim 2.5$ ).<sup>1</sup> This difficulty, combined with the complexity of the normal phase and anomalous impurity effects, has hindered a quantitative analysis of the physical properties of superconducting state (specific heat,<sup>1,3,7</sup> London penetration depth,<sup>8</sup> NMR relaxation time  $T_1$ <sup>9</sup>).

This is made clear when one considers that the power law behaviours observed for the temperature dependence of the specific heat and the London penetration depth point toward points of zeros in the gap function, whereas NMR measurements of  $1/T_1$  point toward lines of zeros, showing the lack of convincing evidence for the actual gap topology. The parity of the pairing is even more unclear. Knight shift measured by muon spin resonance ( $\mu$  SR) gives different results (as to a change at the superconducting transition) which depend on the sample or the muon energy.<sup>9</sup>

In this paper, we focus on the most extensively studied properties of  $\text{UBe}_{13}$ : the upper critical field  $H_{c2}$ , which has a temperature dependence quite unlike that of other heavy-fermion superconductors. Close to  $T_c$ , the upper critical field slope  $\partial H_{c2}/\partial T|_{T_c}$  is very large (estimates in the literature<sup>5,6,11-15</sup> range from  $-25$  T/K up to infinity) and the most detailed measurements agree on a value around  $-42$  T/K.<sup>11</sup> This is usually interpreted as resulting from the quasiparticles in  $\text{UBe}_{13}$  having a very large effective mass  $m^*$  (the Sommerfeld coefficient just above  $T_c$  is about one thousand times that of copper). But  $H_{c2}(T)$  does not follow at all the BCS behaviour for an “orbital limit.” The curvature of  $H_{c2}(T)$  close to  $T_c$  is strongly negative and the measured value of the critical field at zero temperature  $H_{c2}(0)$  ( $\sim 10$ – $14$  T) is much lower than the value deduced from the slope at  $T_c$  ( $\sim 30$  T). However,  $H_{c2}(0)$  greatly exceeds the Clogston Pauli limit ( $H_c^p = 1.84 T_c$ ), suggesting either a p-wave pairing or a strong spin-orbit coupling.<sup>16</sup> The spin-orbit coupling could be intrinsic, due to the f-electron nature of the quasiparticles, or extrinsic, through scattering processes on magnetic impurities.<sup>16</sup>

Another peculiarity is that an inflexion point can be observed in  $H_{c2}(T)$  on the best samples, at roughly  $T_c/2$ , which has been interpreted in many ways. They rely either on the normal-state properties (magnetoresistance or low-temperature specific heat),<sup>6</sup> on field-dependent interactions,<sup>12,17</sup> on multiple superconducting phases<sup>18</sup> or on possible magnetic ordering.<sup>13</sup>

We show here for the first time that taking properly into account the strong-coupling effects yields a complete quantitative understanding of  $H_{c2}(T)$  in  $\text{UBe}_{13}$ . One can explain at the same time the different curvatures observed in  $H_{c2}(T)$  and the value of  $H_{c2}(0)$ . This leads to a prediction that a non-uniform superconducting state (Fulde-Ferrell-Larkin-Ochnikov or FFLO state)<sup>19,20</sup> appears below 0.4 K in our sample.

After a brief description of the various experimental details and results, we develop a simple model of strong-coupling superconductivity, following references.<sup>21,22</sup> For simplicity, we assume s-wave pairing. Actually, the exact symmetry of the pairing which is not yet known for  $\text{UBe}_{13}$  is not very important for the calculation of  $H_{c2}(T)$ . We calculate the orbital and Pauli limits taking into account the possibility of an FFLO state. The calculations are made in the clean limit, as it has been already argued that this should be the case for  $\text{UBe}_{13}$ <sup>12</sup> (the mean free path should exceed 600 Å whereas the coherence length is of the order of 50 Å). These results are then compared to experiment.

## 2. EXPERIMENTAL DETAILS

### 2.1. Resistivity

Two different apparatus, both in dilution refrigerators, were used to perform the resistivity experiments. The first one (in Grenoble) has an 8 T compensated magnet. We used a bridge technique to measure the resistance with a helium-cooled transformer achieving a sensitivity of  $0.1 \text{ nV}/\sqrt{\text{Hz}}$ . This proved useful for the measurement of the initial slope of  $H_{c2}$  where a current density of the order of  $10 \mu\text{A}/\text{mm}^2$  was used in order not to broaden the transition. The second one (in Darmstadt) has an 17.5 T compensated magnet and the resistance was measured with a standard lock-in technique (the noise level here is  $3 \text{ nV}/\sqrt{\text{Hz}}$ ). No significant difference was found in the thermometry between Darmstadt and Grenoble. The match between the two experiments at 369 mK (and 8 T) is better than 1 mK. In Grenoble, the superconducting transition was measured at constant field. In Darmstadt, it was also measured at constant field varying the temperature above 250 mK, and at constant temperature varying the field below 250 mK. In all cases,  $T_c(H)$  (or  $H_{c2}(T)$ ) was determined with an

onset criterion, namely, the crossing point between the linear extrapolation of the normal-state resistivity just above the transition and the linear extrapolation of the resistive transition.

Electrical contacts were made with Indium soldered directly on the sample. The change of resistance due to the superconducting transition of the contacts was of the order 0.1% in zero field. The shape factor was typically  $10 \text{ cm}^{-1}$ , and the current was flowing along a principal crystallographic axis.

## 2.2. Specific Heat

Specific heat was measured in Grenoble using a heat pulse technique<sup>12, 23</sup> in the same cryostat used for the resistivity experiments. Care was taken to have long relaxation time constants ( $\sim 30$  seconds) so that thermal equilibrium was achieved inside the sample despite the low thermal diffusivity of  $\text{UBe}_{13}$  in the superconducting state. The sample was cooled down to 20 mK, but the measurements are assumed to be reliable only above 50 mK, when the relaxation of the heat inside the sample started to be exponential.

## 2.3. Sample Quality

A bar shaped single crystal ( $1 \times 1 \times 2 \text{ mm}^3$ ) was grown by the method described in.<sup>15</sup> Sample quality was checked by specific heat ( $C$ ) at zero field and by resistivity in high field, in order to get the residual resistivity  $\rho_0$ .

Figure 1 displays the  $C/T$  versus  $T$  curve. The superconducting transition width is 50 mK, with a midpoint at  $T_c = 903 \text{ mK}$ , and no finite residual  $C/T$  term was detectable down to 50 mK. Below this temperature,  $C/T$  increases slightly on cooling, but as indicated above, this could be due to technical problems such as non equilibrium heat distribution inside the sample, or hyperfine contributions. We also found a usual value for  $C/T$  in the normal-state ( $850 \text{ mJ/K}^2 \text{ mol}$  at 1 K). If the value of both  $T_c$  and the transition width are used as criteria of sample quality, this single crystal is superior to those previously used by different groups.<sup>5, 13, 24</sup>

The resistivity is known to be another sensitive method to check the sample quality, as  $\rho_0$  reflects mainly the defects of the crystal. The problem in the case of  $\text{UBe}_{13}$  is that, due to its superconductivity, one needs very high magnetic fields to measure the residual resistivity at very low temperature. We have found  $\rho_0 \sim 10 \mu\Omega \cdot \text{cm}$  ( $T = 5 \text{ mK}$  and  $H = 15 \text{ T}$ ). This has to be compared to  $\rho \sim 150 \mu\Omega \cdot \text{cm}$  at  $T = 1 \text{ K}$  and  $H = 0 \text{ T}$ . The resistive transition did not broaden very much with magnetic field:  $\Delta T_c/T_c = 2\%$  at zero field and  $\Delta H_{c2}/H_{c2} = 7\%$  at 14 T, where  $\Delta T_c$  and

$\Delta H_{c2}$  are obtained via the usual criteria  $\Delta T_c = T_c(90\%) - T_c(10\%)$  and  $\Delta H_{c2} = H_{c2}(90\%) - H_{c2}(10\%)$ . Again, this indicates that our  $\text{UBe}_{13}$  single crystal is of a high quality.

## 2.4. Results

A surprising result in our  $C/T$  versus  $T$  data (Fig. 1) is that the entropy balance requires only a linear extrapolation of the normal state  $C/T$  (noted  $C_n/T$ ) to  $T=0$  in order to be fulfilled. Results published previously in the literature indicated, as mentioned above, a strong upturn at low temperature in the normal phase extrapolated  $C_n/T$ .<sup>3,4</sup> One possible explanation is that the previous results were somewhat blurred due to some nonintrinsic contributions at very low temperature. In any case we find that  $C_n/T = \gamma - aT$  ( $\gamma = 1 \text{ J/K}^2 \text{ mol}$  and  $a = 0.15 \text{ J/K}^3 \text{ mol}$ ) which is not expected for a Fermi liquid, but exists also in other heavy fermion materials.<sup>25</sup> This emphasises (as the temperature dependence of the resistivity...) that a simple Fermi liquid regime is not yet reached at  $T_c$ . For our sample, we find a specific heat jump at  $T_c$  of  $\Delta C/C_n = 2.65$  (and  $\Delta C/C_n = 2.2$  if we normalise to the Sommerfeld coefficient extrapolated at

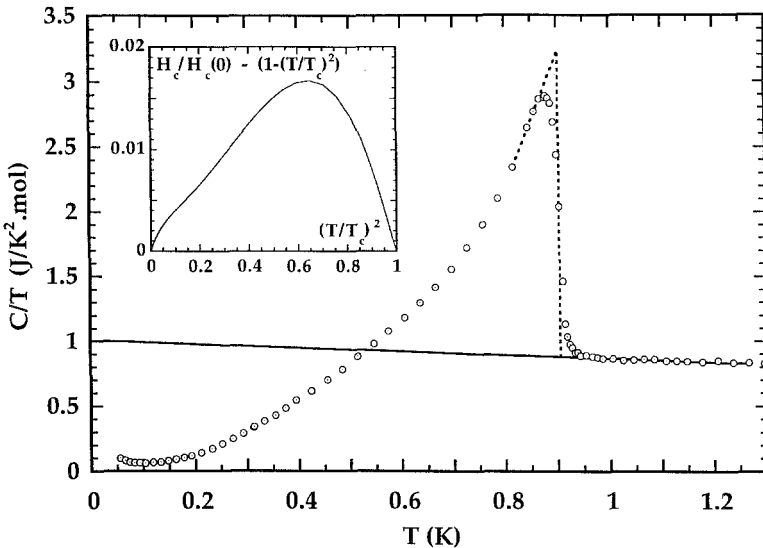


Fig. 1. Specific heat versus temperature for our annealed  $\text{UBe}_{13}$  single crystal at zero magnetic field. The full line corresponds to the normal phase extrapolation fulfilling the entropy balance. The dashed line is the ideal transition deduced from equal entropy construction. Inset: deviation of the thermodynamic critical field from the parabolic behaviour (see text).

zero temperature). A roughly  $T^3$  temperature dependence is observed below 0.5 K, in good agreement with previous results.

The inset of Fig. 1 shows the usual plot which classifies the superconductors (strong or weak coupling) with respect to their thermodynamic critical field  $H_c$ . We have plotted the deviation function  $D(T)$  from the parabolic behaviour, i.e.:

$$D(T) = \frac{H_c}{H_c(0)} - \left(1 - \left(\frac{T}{T_c}\right)^2\right)$$

$H_c$  being calculated from the specific heat taking the idealised jump  $\Delta C$  at  $T_c$  (as deduced from an equal-entropy construction). In this representation, the weak-coupling regime is characterized by a negative deviation and the strong-coupling regime by a positive one. As expected from the value of the specific heat jump at  $T_c$ ,  $\text{UBe}_{13}$  is found to be strongly coupled.  $\text{UBe}_{13}$  is very peculiar, as all the other heavy fermion superconductors show weak-coupling behaviour, i.e. the specific-heat jump is smaller than the BCS value and the temperature dependence of  $C(T)$  is reasonably well fitted by weak coupling models.<sup>23</sup> However, if one keeps in mind that all of these materials have strong electronic correlations with very large effective mass enhancement, i.e. very small electronic energy scale, the expected behaviour is that of strong coupling type.

The upper critical field  $H_{c2}(T)$  is shown in figure 2 and the inset of this figure focuses on the low-field region. The main results are:

(i) The upper critical field slope at  $T_c$  is found to be large but finite, i.e.:

$$\left. \frac{\partial H_{c2}}{\partial T} \right|_{T_c} = -50 \pm 10 \text{ T/K}$$

The uncertainty is large for several reasons. The first one is that  $H_{c2}$  has a very strong curvature close to  $T_c$ , so that only the very lowest field data points could be used to determine the slope at  $T_c$ . The second one is that the slope itself is large so that, at low field, the shift of the transition is typically 0.2 mK for 100 Gauss (our typical field step) and has to be compared to the resistive transition width in low field (20 mK). These two effects cause a large uncertainty in each point.

(ii) We observe a saturation of the upper critical field at very low temperature. The value is  $H_{c2}(0) = (14 \pm 0.5) \text{ T}$ . In this case, the uncertainty is given directly by the transition width in high field. This saturation effect was not seen before in high quality polycrystals, where the value found for  $H_{c2}(0)$  was close to ours ( $\sim 13 \text{ T}$ ),<sup>12</sup> but where the authors used

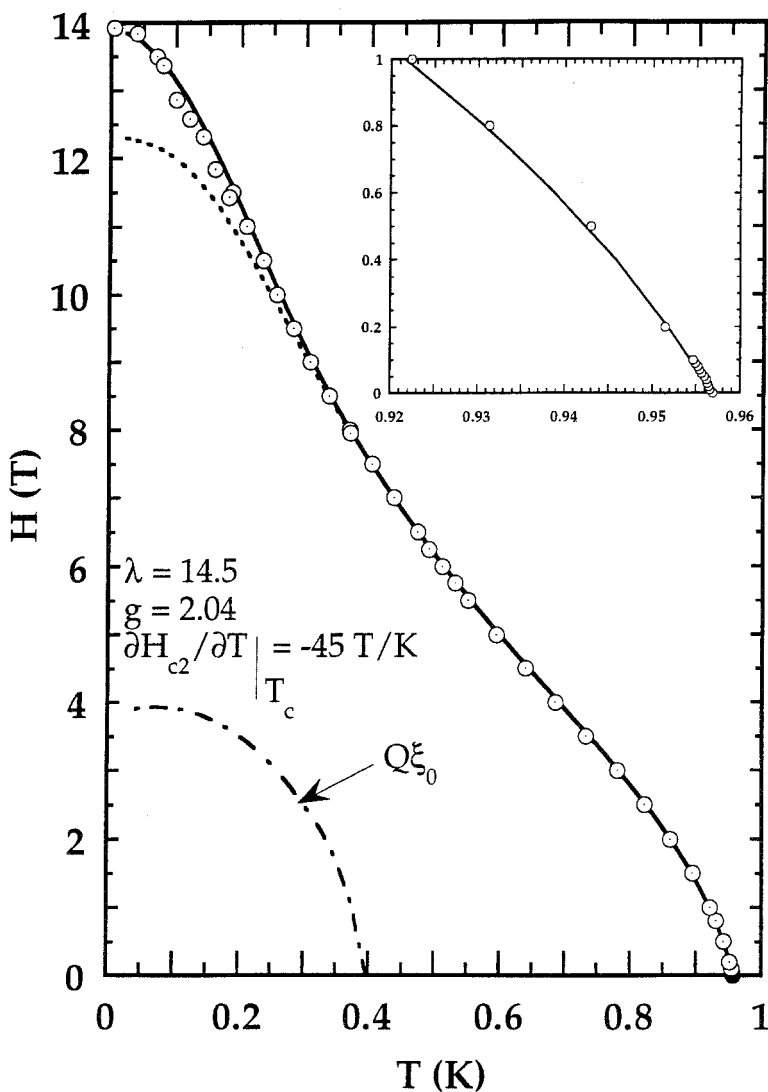


Fig. 2. Upper critical field  $H_{c2}(T)$  of our sample of  $\text{UBe}_{13}$  measured by resistivity. The full line corresponds to the best fit from our model (see text), including the FFLO state. The value of parameters used for the fit are also reported on the graph. The dashed line is the fit without the FFLO state. The dash dotted line is the dimensionless value of the modulation vector in the FFLO state. Inset: the low-field region. Note that curvature starts already at very low field ( $\sim 0.2$  T).

the mid point of the resistive transition to determine  $T_c(H)$ . For single crystals, the values already published were lower ( $< 11$  T) either with<sup>13</sup> or without the saturation.<sup>5</sup> Finally, these results seem to depend on the way one extracts  $T_c(H)$  or  $H_{c2}(T)$  from the raw data: the saturation appears if one takes an onset-like criterion in order to correct for the field broadening of the transition. This is especially important in case of large broadening.

(iii) We observe a change of curvature at 6 T, this change being smooth rather than abrupt.

Again, this behaviour resembles that of high-quality polycrystals,<sup>12, 13, 18</sup> but differs from that of other single crystals.<sup>5</sup>

We conclude that, if the quality of the sample is increased,  $H_{c2}(T)$  saturates as  $T \rightarrow 0$ , the value of  $H_{c2}(0)$  is increased (and saturates), the change of curvature near  $T_c$  is more pronounced, but the slope of the critical field at  $T_c$  remains finite, no matter whether a single crystal or a polycrystalline sample is used.

### 3. THEORETICAL MODEL

In the following, we describe (to our knowledge) the first analysis of the FFLO state for a strong coupling superconductor. The temperature interval for the existence of a FFLO state is maximum for intermediate coupling (it is approximately 20% larger for a coupling parameter  $\lambda \cong 2$ ).

We use the model of superconductors investigated in<sup>21</sup> where the system of electrons coupled with a one-phonon mode has been considered. More precisely the spectral density of interaction (Eliashberg function) is taken in the form:

$$\alpha^2(\omega) F(\omega) = \left( \frac{\lambda \Omega}{2} \right) \delta(\omega - \Omega) \quad (1)$$

where  $\lambda$ ,  $\Omega$  are used in their usual sense and  $\omega$  is the frequency.

In the case of heavy-fermion superconductors like  $\text{UBe}_{13}$  or  $\text{URu}_2\text{Si}_2$ , it is often assumed that the coupling of electrons is mediated by magnetic excitations rather than phonon like. However, we believe that the model<sup>21</sup> can be useful for a general understanding of the critical-field behaviour and FFLO-state formation, in the case of strong-coupling superconductivity, regardless of the pairing mechanism.

The system of linear equations for the gap function  $\Delta$  in the presence of a magnetic field has the form<sup>21, 22</sup> (in standard notation):

$$\Delta(i\tilde{\omega}_n) = \frac{\pi T}{\Omega} \sum_{\omega_m} \lambda(\omega_n - \omega_m) \chi(\tilde{\omega}_m) \Delta(i\tilde{\omega}_m) \quad (2)$$



where  $T$  is the temperature and  $\omega_n$ ,  $\tilde{\omega}_n$  and  $\lambda(\omega)$  are defined by:

$$\begin{aligned} \tilde{\omega}_n &= \omega_n + \pi T \sum_m \lambda(\omega_n - \omega_m) \operatorname{sgn}(\omega_n) \\ \lambda(\omega_n - \omega_m) &= \frac{\lambda \Omega^2}{\Omega^2 + (\omega_n - \omega_m)^2}, \quad \omega_n = \pi T(2n + 1) \end{aligned} \quad (3)$$

The function  $\chi(\tilde{\omega}_n)$  is actually the eigenfunction of the operator introduced by Helfand and Werthamer<sup>26</sup> for calculations of the upper critical field. Generalising the approach<sup>26</sup> by taking into account the possibility of the formation of the FFLO state, i.e. introducing a modulation with the wave vector  $Q$  of the gap function along the magnetic field  $H$ , we obtain the following expression for  $\chi(\tilde{\omega}_n)$

$$\chi(\tilde{\omega}_n) = \int_0^\infty dx \frac{\beta \exp(-\beta x)}{\sqrt{\tilde{Q}^2 + x}} \tan^{-1} \left( \frac{\sqrt{\tilde{Q}^2 + x}}{|\tilde{\omega}_n| + i\mu_B H \operatorname{sgn}(\tilde{\omega}_n)} \right) \quad (4)$$

where  $\beta = (2\Omega^2 c / \hbar e H v_F^2)$  and  $\tilde{Q}$  is the dimensionless wave vector of FFLO state  $\tilde{Q} = (\hbar v_F Q / 2\Omega)$ .

The upper critical field is determined as the field where the system of equations (2) has a nontrivial solution.

In the case of purely paramagnetic limitation (neglecting the orbital effect) the expression for  $\chi(\tilde{\omega}_n)$  becomes:

$$\chi(\tilde{\omega}_n) = \frac{2\Omega}{Qv_F} \tan^{-1} \left( \frac{Qv_F/2}{|\tilde{\omega}_n| + i\mu_B H \operatorname{sgn}(\tilde{\omega}_n)} \right) \quad (5)$$

The condition that the set of equations (2) has a solution is:

$$\det \| 1 - \pi T \lambda(\omega_n - \omega_m) \chi(\tilde{\omega}_m) \| = 0 \quad (6)$$

This gives an equation for the upper critical field suitable for numerical calculations (analytical expressions valid for the limit  $\lambda \gg 1$  are given in the appendix). Our numerical procedure has a good convergence except at very low temperature: for  $T < 0.2 T_c$ , the size of the matrix was increased from  $60 \times 60$  up to  $180 \times 180$ . Further increase of the matrix size did not change the upper critical field by more than 1%. The result for the pure orbital and the pure Pauli limit (including the FFLO state) are presented in figure 3 for different values of  $\lambda$ . Note that the positive curvature of the orbital critical field in the case of large  $\lambda$  has been already found by the calculations of reference.<sup>21</sup> Following reference,<sup>21</sup> the physical reason for this effect is that near  $T_c$ , in the case of strong coupling, there are many thermally

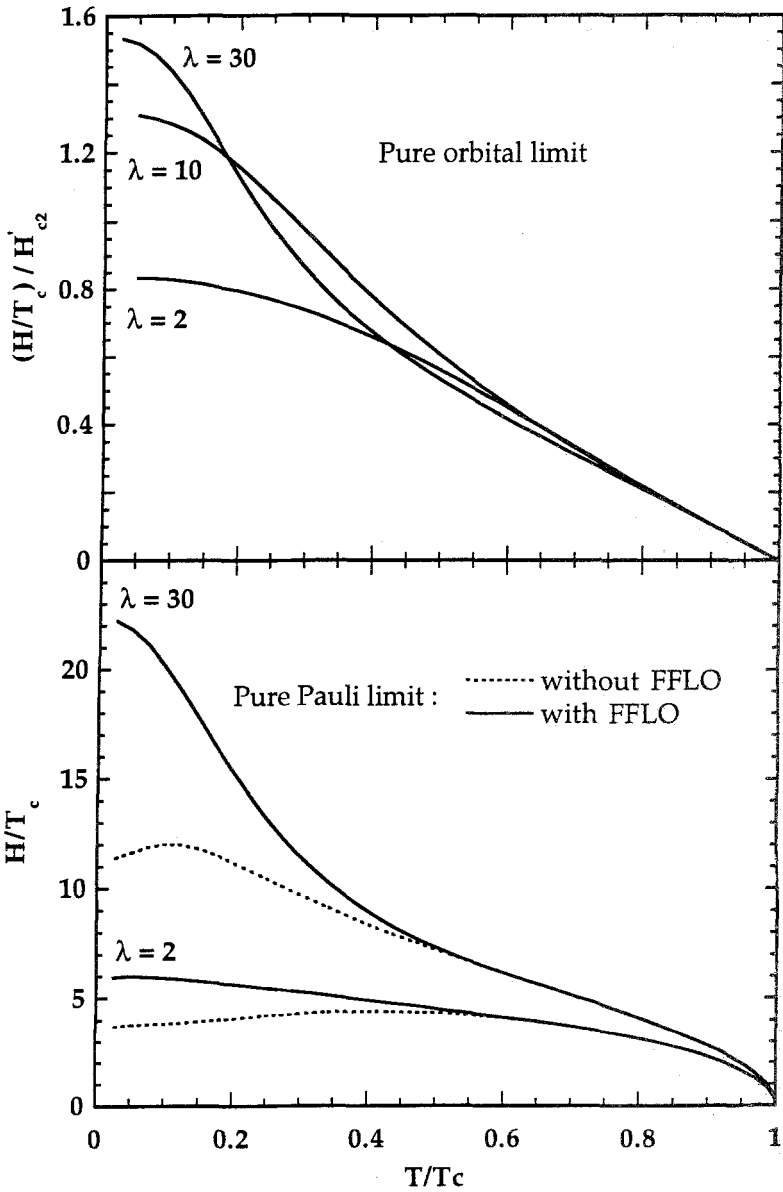


Fig. 3. Top: orbital upper critical field calculated for three different values of the strong coupling parameter  $\lambda$ , normalised to a slope at  $T_c$  equal to 1. Bottom: Pauli critical field for two different values of  $\lambda$ , calculated with the FFLO state (full line) or without (dashed line).

activated phonons (or, in general, excitations responsible for the pairing). This is due to the fact that  $T_c \geq \Omega$ , contrary to the weak coupling BCS case where  $T_c \ll \Omega$ . These thermal excitations depress the critical temperature compared to the case of pairing due to virtual excitations only ( $T_c^0 \sim \lambda\Omega$ ). At low temperature ( $T < \Omega$ ), these thermal excitations disappear and the properties of the superconductor (upper critical field, superconducting gap, etc...) are determined by the bare  $T_c^0$ . This reasoning is completely applicable for the Pauli limit to explain both the positive curvature and the violation of the Clogston limit.

In Fig. 3, the purely paramagnetic limitations with and without the FFLO state is presented for  $\lambda = 2$  and  $\lambda = 30$ . Note that the temperature  $T^*$  at which the FFLO state appears is somewhat different from the BCS case ( $T^* = 0.56 T_c$ ). The maximum  $T^*$  is reached for  $\lambda \cong 2$  ( $T^* = 0.66 T_c$ ), whereas for large  $\lambda$  the ratio  $T^*/T_c$  starts to decrease. At the same time, the relative increase of the critical field below  $T^*$  due to the FFLO state is more pronounced for larger values of  $\lambda$ . In the absence of the FFLO state, the superconducting transition should be of first order, but we did not calculate the corresponding transition line. We can just guess that, as in the BCS case, this first order line lies below the FFLO line.

#### 4. DISCUSSION

In Fig. 2, our data are fitted using the formulae of the previous section. This fit clearly produces a good description of the data, with three adjustable parameters of whom two may be directly compared with experiment. These parameters are:

(i) The slope of  $H_{c2}$  at  $T_c$ , which determines the orbital limit. The value of  $-45$  T/K is well within the error bars of our experimental determination, and the inset of figure 2 shows that it is fully compatible with the data.

(ii) The gyromagnetic ratio of the quasiparticles  $g$ , which determines the Pauli limit. We got slightly better results with  $g = 2.04$  than with the free electron value  $g = 2$ . However the difference is probably not significant, due to the crudeness of the model: we used a very simple spectrum of interactions, an  $s$ -type of pairing and a spherical Fermi surface. The reasons are, among others, that there is no experimental measurement of the Fermi surface (quantum oscillations of the heavy quasiparticles have not been clearly seen), and the nature of the pairing interaction is unknown. In addition, proposals of unconventional superconductivity in  $\text{UBe}_{13}$  are of a speculative type, due to a number of contradictions remaining among experimental results. Another point is that there is no reason to find a  $g$

value exactly equal to 2 for these heavy quasi particles (of 5f symmetries), in fact it is even surprising to find a value so close to the free-electron value.

(iii) The strength of the coupling, parametrized by  $\lambda$ . In a BCS-like formalism,  $\lambda \sim N(0) V$ , where  $V$  is the coupling energy and  $N(0)$  the density of states. The value we find for  $\lambda$  ( $\sim 14.5$ ) is very large, bearing in mind that, for classical strong-coupling superconductors,  $\lambda$  values are of the order of 1 (in lead, for example). However one should also take into account that in heavy-fermion materials, the mass enhancement is huge due to strong electronic correlations so that a much larger strong-coupling parameter appears plausible. The value of  $\lambda$ , which is indeed the only really free parameter of the fit, controls both the Clogston limit (and thus  $H_{c2}(0)$  as the orbital limit is much larger) and the curvature of  $H_{c2}(T)$  at finite temperature. In other words, if we would take a smaller value of  $\lambda$  while reducing the  $g$  factor (in order to maintain the value of  $H_{c2}(0)$ ), we would miss the general behaviour of the critical field, i.e. both the inflection point at 6 T and the strong curvature close to  $T_c$ .

Let us now comment on some consequences of our results. Perhaps, the most remarkable one is that the temperature dependence of  $H_{c2}$  is almost completely understood in terms of Pauli limitation. This was of course suggested by the strong curvature close to  $T_c$ , but it was impossible to understand<sup>6</sup> without taking properly into account the strong coupling effects. In particular, we see now that the fact that  $H_{c2}(T)$  strongly exceeds the classical Clogston limit is no proof of  $p$ -wave pairing, and no spin-orbit scattering has to be involved to explain this violation. Note that in URu<sub>2</sub>Si<sub>2</sub> also, for  $H$  parallel to the tetragonal  $c$  axis,  $g \sim 2$  has been found<sup>27</sup> from the same kind of analysis of  $H_{c2}(T)$ . In both compounds, we therefore expect a full reduction of the Knight shift well below  $T_c$ , as the Pauli limitation can be clearly seen in our data. In other words, the actual contradiction among NMR or  $\mu$  SR experiments<sup>9, 10</sup> might come from sample-quality problems (like surface defects) or technical difficulties (absolute magnitude of the Knight shift) rather than being related to intrinsic properties of the material.

Another consequence is the prediction, in this clean Pauli-limited superconductor, of the appearance of a FFLO state below 0.4 K. To our knowledge, it is the first time where a quantitative fit has been made showing the existence of such a state. It had been guessed<sup>28</sup> that UBe<sub>13</sub> could be a good candidate for a FFLO state. Indeed, strong coupling is favourable in this case for the appearance of the FFLO state but, concerning the inflexion point close to  $T_c \sim 0.5$  K, we know now that

the FFLO state appears only at a lower temperature. Note that this state can only exist in clean superconductors and could be sensitive to impurities. This may explain why the value of  $H_{c2}(0)$  has increased with the sample quality.

We emphasise that, although as  $T_c \rightarrow 0$  the orbital limit is roughly four times larger than the Pauli limit, it has still a strong effect on the FFLO state as it reduces the temperature  $T^*$  of the appearance of the state from  $T/T_c \sim 0.62$  to  $T/T_c \sim 0.42$ . The size of the  $H_{c2}$  enhancement due to the FFLO state is also reduced. As a consequence, the first-order transition line that should exist below  $H_{c2}$  for  $T < 0.4$  K has to be very close to the second-order line of  $H_{c2}(T)$ . The former will be therefore experimentally rather difficult to detect, since the transition widths could be larger than the difference between the 2 lines.

Compared to other systems where there has been evidence for the FFLO state, the case of  $\text{UBe}_{13}$  is much stronger (cf.  $\text{URu}_2\text{Si}_2$ ),<sup>27</sup> with the temperature  $T^*$  well defined. In  $\text{UPd}_2\text{Al}_3$ <sup>29</sup> or  $\text{CeRu}_2$ ,<sup>30</sup> an irreversibility line observed below  $H_{c2}(T)$  has first been considered as a possible FFLO transition line, but is now rather interpreted as due to peak effect. A new theoretical approach explains the irreversibility phenomena with a generalised FFLO scenario.<sup>31</sup> But one should note that this line, which is observed almost up to  $0.8 T_c$ , is not understandable in the models currently proposed for the FFLO state. Worse, in these materials, the orbital limit is quite close to the Pauli limit. Then, according to the current models, the temperature  $T^*$  of appearance of the FFLO state has to decrease strongly compared to the pure Pauli case.<sup>32, 33</sup>

At least one puzzling question remains concerning our fit, and this is related to the large value of  $\lambda$ . If the interactions were mediated by phonons, such a coupling strength would most likely lead to a lattice instability. On the other hand, if the interactions are mediated by a magnetic background, one may guess that  $\text{UBe}_{13}$  would show a magnetic instability. The origin of the strong-coupling interactions is as mysterious as is the origin of the mass enhancement. But it is plausible that part of the damping effects of the quasi particles near  $T_c$  (which are the key reasons for strong-coupling effects) are coming from the proximity of  $T_c$  and the Fermi temperature  $T_F$ : we recall that the Fermi liquid behaviour of the normal phase is not yet well established at  $T_c$  in  $\text{UBe}_{13}$ , due to this proximity. Another problem is that, at least in a “classical” strong-coupling theory, the specific-heat jump at  $T_c$  is related to the value of  $\lambda$ . In  $\text{UBe}_{13}$ , it is essentially the same as in lead, although a factor of 10 is found between their respective  $\lambda$  values. Whether this is due to the origin of the coupling (in a “classical” strong-coupling theory, the value of the Sommerfeld coefficient is also related to  $\lambda$ ) or to unconventional pairing remains to be understood.

## 5. CONCLUSION

We have presented new data on the upper critical field of a high-quality single crystal of  $\text{UBe}_{13}$ . Overall, they are in good agreement with previously published data, with a subtle difference on  $H_{c2}(0)$ , being slightly higher in the present measurements. We have confirmed that the initial slope is large but finite ( $\sim -45$  T/K). We have proposed a simple model which takes into account strong-coupling effects as inferred from specific-heat data. This model can explain quantitatively the very peculiar behaviour of  $H_{c2}$ , with a full Pauli limit (gyromagnetic ratio  $\sim 2$ ) and a very large value of the coupling parameter. We also predict that a non-uniform superconducting state (FFLO state) appears in  $\text{UBe}_{13}$  below 0.4 K for very pure samples. The nature of the strong-coupling however remains an open question.

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## APPENDIX. PARAMAGNETIC CRITICAL FIELD IN THE LIMIT $\lambda \gg 1$

In the limit of very large  $\lambda$  in the Eq. (2) we may restrict ourselves to:  $\Delta(\pi T) = \Delta^*(-\pi T)$ , all other  $\Delta(\omega_n)$  being zero. In such a case, the equation for the paramagnetic critical field becomes very simple:

$$\frac{\Omega}{\pi T} - \chi_1 - \chi_1^* + \frac{\pi T}{\Omega} \left( 1 - \left( \frac{\Omega}{2\pi T} \right)^2 \right) \chi_1 \chi_1^* = 0$$

where  $\chi_1 = \chi(\tilde{\omega}_n = \pi T(\lambda + 1))$  (A1)

The critical temperature in this approximation is  $T_c = (\Omega/2\pi) \sqrt{\lambda}$  which is rather close to the exact asymptotic limit ( $\lambda \gg 1$ ):  $T_c = 0.18\Omega \sqrt{\lambda}$ .<sup>34</sup>

Without FFLO state the paramagnetic critical field for  $T \gg \Omega$  is given by the expression:

$$\frac{\mu_B H}{\pi T_c} = \frac{T_c}{T} \sqrt{1 - \left( \frac{T}{T_c} \right)^4} \quad (\text{A2})$$

Note the strong positive curvature of the critical field. The temperature  $T^*$  below which the FFLO state appears is  $T^* = T_c(2/\lambda)^{1/4}$ .

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