Pressure Dependence of the Upper Critical Field of the Heavy Fermion Superconductor UBe_{13}

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We report measurements under pressure of the upper critical field of the heavy fermion superconductor UBe_{13}. An interpretation in the framework of a simple strong coupling model is achieved consistently with only one arbitrary parameter: the strong coupling constant λ. We find that UBe_{13} is in an extreme strong coupling regime and that the variation of λ with pressure explains the pressure dependence of the thermodynamic properties of both the normal and the superconducting phases. It reveals a strong interplay between the mass renormalization and the pairing mechanisms, yielding the first quantitative indication of a nonphonon mediated pairing in a superconductor. [S0031-9007(98)08084-3]

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A characteristic feature of heavy fermion (HF) intermetallic compounds is the occurrence at low temperature ($\approx$10 K) of a very large renormalization of the mass of the charge carriers: up to several hundred times the mass of a free electron. There is no complete understanding of this huge mass renormalization, but the consensus is that it arises from strong electronic correlations, which also produce different kinds of magnetic excitations. It has also long been suggested that the pairing mechanism responsible for superconductivity in HF may differ from the usual electron-phonon interaction, and is believed to involve the magnetic properties of the normal phase. But as well as for high-T_c cuprates, real quantitative results on this point are still lacking. We propose here a new approach to this challenging question, taking advantage of the extreme strong coupling regime met in the HF superconductor UBe_{13}: our measurements and analysis of the upper critical field H_{c2} of UBe_{13} under pressure reveal a direct link between the mechanisms of mass renormalization and superconductivity in this system, yielding the first quantitative indication of a nonphonon mediated mechanism in a superconductor.

The normal phase of UBe_{13} already presents striking features. The quasiparticles have a record effective mass (renormalization by a factor of 1000 has been suggested [1,2]), and coherence in the lattice occurs only at very low temperature: the resistivity presents a maximum at 2.5 K [1]. Therefore, and as opposed to all other HF superconductors, UBe_{13} has the very unusual feature to be in an ill defined Fermi liquid regime when superconductivity appears at $T_c \approx 1$ K [1,3]. With regard to the superconducting state, it is clearly in a strong coupling regime, as indicated by the relative jump of the specific heat at T_c: of the order of 3, much larger than the BCS weak coupling value of 1.43 [4]. Quite curiously, the specific heat has long been the only superconducting property analyzed in a strong coupling scheme [4,5]. It is only very recently that strong coupling effects were quantitatively discussed on H_{c2} [6], providing a new but straightforward interpretation of its peculiar behavior.

The temperature dependence of H_{c2} in UBe_{13} at zero pressure (see the curve $P = 0$ kbar of Fig. 1) is very puzzling. First, H_{c2} shows a very strong negative curvature near T_c. This observation suggests a strong paramagnetic limitation of the upper critical field (i.e., the effect of the field on the spin of the quasiparticles) [2]. The difficulty for a quantitative fit comes both from the value of H_{c2} at $T = 0$ in UBe_{13} [more than 7 times greater than the Clogston paramagnetic limit: $H^P(T = 0) = \sqrt{2}\,\Delta(T = 0)/g\mu_B \approx 1.85 k_B T_c$, where Δ is the energy gap, g the gyromagnetic ratio, and μ_B the Bohr magneton] and from

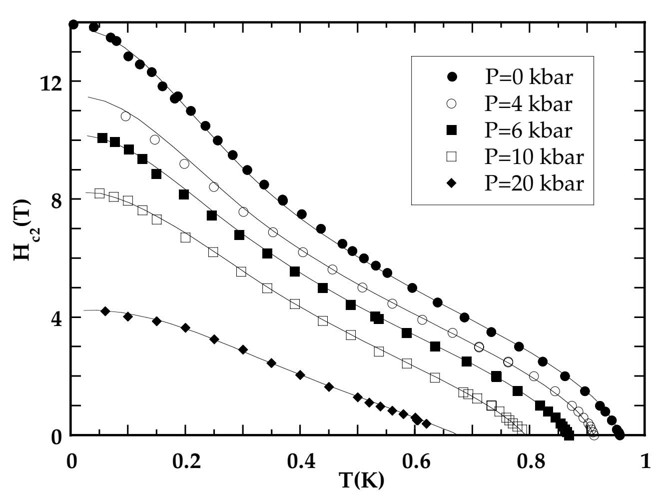

FIG. 1. Upper critical field of UBe_{13} under various pressures. Both the strong curvature near T_c and the anomalous increase in high field gradually disappear with increasing pressure. Full lines are the best fits of our strong coupling model.

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the shape of $H_{c2}(T)$, which displays, on the best samples, an unusual upward curvature at a temperature around $T_c/2$. Tentative explanations have relied on two superconducting order parameters with weak Pauli limiting [7], on additional magnetic phase transitions [8,9], or even on a field dependence of the normal or superconducting state parameters [10–13]. Also they cannot be completely dismissed, none of these phenomenological interpretations have found a firm basis in other measurements or theoretical developments. It was shown recently [6] that such a behavior of H_{c2} is predicted directly by the Eliashberg equations for an extreme strong coupling regime, without any additional hypothesis (see also the discussion below). A prime interest of these new pressure measurements of H_{c2} is to probe if this straightforward interpretation can consistently explain $H_{c2}(T)$ when the strong coupling regime is changed. As we shall see, they also lead to a much deeper insight into the connection between normal phase properties and superconductivity in UBe_{13}.

Experimental procedure and setup.—$H_{c2}(T)$ has been measured by monitoring the resistive transition. To determine the critical temperature at fixed magnetic field or the critical magnetic field at fixed temperature, a "junction" criterion is used (i.e., the determination of the crossing point between the extrapolated resistivity of the normal phase and the extrapolated linear part of the transition). We use a 12 T compensated magnet, which enables reliable thermometry under field. The pressure is applied in a piston-cylinder cell, with a liquid mixture of alcohol as transmitting medium. It is measured by the known pressure dependence of T_c in UBe_{13} [14], which has been checked in our sample against the superconducting transition of a tin manometer at $P = 6$ kbar and $P = 20$ kbar. Hydrostaticity of the cell is controlled by measuring the broadening of the transition: 2.5 mK/kbar. For the highest pressures of 10 and 20 kbar, the low field transition is blurred by another transition of (we guess) uranium filaments in our sample, whose superconducting transition temperature is known to increase under pressure but is rapidly suppressed by a magnetic field [15]. Apart from this problem, a special effort is made on the accuracy of the determination of the initial slope at T_c, and on the saturating value at $T \to 0$. For this purpose, the current density was kept low. Temperature or field sweeps were performed, overlapping in a range of intermediate fields and temperatures where the results were tested to be consistent.

Sample quality and results.—The evolution of H_{c2} under pressure is reported in Fig. 1. The sample quality has been improved since the pioneering measurements of Ref. [16] (which lacked a little of data for a quantitative analysis) so that the agreement between the two sets of data is satisfying. Concerning our data, those at zero pressure (from [6]) were measured on a high quality single crystal. The data under pressure were taken on the polycrystalline sample of [12]. The T_c of this last sample at zero pressure is slightly higher than that of the single crystal (0.97 K instead of 0.957 K) but with a transition width $\Delta T_c \approx 28$ mK instead of 18 mK, which remains, nevertheless, good for UBe_{13}. Both samples have a residual resistivity in high field of about 10 $\mu\Omega$ cm, which also attests to their good quality, and we can safely assume that they are in the clean limit [1,12]. As expected, in the range of fields where the polycrystal has been measured (up to 12 T), the H_{c2} of the two samples match when the same onset criterion is used (UBe_{13} is cubic). So at zero pressure, we have reported here the results on the single crystal which are more precise and extend up to larger fields.

Apart from the known variation of T_c under pressure, the striking feature seen on Fig. 1 is the general evolution of the shape of $H_{c2}(T)$: the peculiar behavior observed at zero pressure gradually disappears with increasing pressure. Let us discuss this evolution from the point of view of a strong coupling scheme. We will not reproduce here the calculations presented in [6] but rather concentrate on the physical effects and the parameters entering the model, which are all directly probed by the pressure measurements. In the absence of a precise description of the quasiparticles in the normal state and with no knowledge of the type and spectrum of the interactions responsible for the pairing, we have used a simple Einstein spectrum for the density of interactions of the form $\alpha^2F(\omega) = (\lambda\Omega/2)\delta(\omega - \Omega)$ [17], where λ is the parameter giving the strength of the pairing and Ω corresponds to the characteristic frequency of the excitations responsible for this pairing. Because large values of λ are used in our model, we have neglected the possible influence of the Coulomb repulsion (characterized by a single scalar parameter μ^*), an hypothesis that we will justify more quantitatively later.

From a general point of view, the strong coupling regime essentially adds two effects on H_{c2} with respect to a weak coupling scenario. The first one is an enhancement of the orbital limitation due to the mass renormalization of the quasiparticles induced by the interactions responsible for the pairing. This renormalization already exists in the normal state and is controlled by λ through the relation: $m^*/m_{\mathrm{band}} = \lambda + 1$. m_{band} is the band mass of the quasiparticles, which means in our case the mass of the quasiparticles renormalized by all the interactions which do not participate in the pairing potential. The strong coupling effects on the orbital limitation could be mimicked by a weak coupling scenario if the full renormalized value m^* of the mass is used in the Helfand-Werthamer expressions instead of m_{band} [18,19].

The second type of effects are thermal effects, and they are completely specific to the strong coupling regime. Indeed, the fact that T_c can be quite close to the characteristic energy Ω of the excitations responsible for the pairing leads to a reinforcement of the superconducting properties at temperatures much lower than Ω where the pair-breaking thermal excitations have disappeared. It affects the orbital limitation but has the most dramatic effects on the Pauli limit [17]. The latter is enhanced at low temperature, compared to the BCS prediction, due to the increase of the ratio $\Delta(T = 0)/k_B T_c$. There is another factor which raises further the Pauli limitation in the strong coupling regime: in a clean Pauli limited superconductor, an elaborate calculation of H_{c2} shows that at low temperature, the transition to the superconducting phase is made into a modulated phase called the Fulde-Ferrel-Larkin-Ovchinnikov (FFLO) phase. It yields a reinforcement of H_{c2} compared to the calculation for a uniform state which is at most 5% in the weak coupling regime [20], but is much larger in the strong coupling regime. The appearance of this phase below a temperature $T_{\rm FFLO}$ of the order of 0.4 K at zero pressure is calculated without any additional free parameters by optimization of H_{c2} with respect to the modulation vector, and is responsible for the upward curvature of $H_{c2}(T)$ around $T_c/2$ [6].

The microscopic calculations [6] of $H_{c2}(T)$ have been performed for a simple s-wave state, probably not the most appropriate for UBe_{13}, but actually the symmetry of the pairing has little influence on H_{c2} (only the amplitude of the paramagnetic effect may be strongly affected in case of odd-parity pairing). λ is the only truly arbitrary parameter of the fit: Ω is calculated from the values of λ and T_c, where μ^* has been fixed at 0. The gyromagnetic ratio g for the paramagnetic limitation has been kept close to the free electron value ($g = 2$) and the measured initial slope $(\partial H_{c2}/\partial T)_{T=T_c}$, essentially proportional to m^{*2} for a clean superconductor, determines the orbital limitation. The best fits of H_{c2} for all pressures are displayed in Fig. 1, and their parameters are reported in Table I, together with $T_{\rm FFLO}$, the temperature of the appearance of the FFLO phase.

At first glance, a remarkable feature of the model is that it explains the evolution of the shape of $H_{c2}(T)$ with a value of $g \approx 2$ (the free electron value) remaining almost pressure independent: this supports that H_{c2} in UBe_{13} is mainly controlled by the Pauli limitation with strong coupling effects. In particular, the gradual disappearance of the upturn is naturally explained by the decrease of $T_{\rm FFLO}$ due to the fact that the strong coupling parameter is decreased under pressure, and that the orbital limitation becomes more dominant at high pressures (see the decrease of the initial slope). It appears that the balance between the orbital and paramagnetic limitation is reversed in the pressure range of our experiments. We stress that, for each fit, λ is the only real free parameter if one considers that the value of g has to be about 2, and that the initial slope is controlled by the measurements close to T_c: So λ is basically fixed by the value of $H_{c2}(0)$ (in order to provide the necessary increase of the Pauli limitation) and agreement of the fit with the experiment in such a range of values of λ and of the initial slope is very likely meaningful. But further insight can be gained by analyzing the pressure variation of the parameters, and most remarkably that of λ.

TABLE I. Parameters of the fits of Fig. 1. The gyromagnetic ratio g is almost constant which supports our interpretation of dominant Pauli limitation; pressure in kbar and $(\partial H/\partial T)_{T=T_c}$ in T/K.

P	λ	g	$(\partial H/\partial T)_{T=T_c}$	$T_{\rm FFLO}/T_c$
0	15	2.1	−55	0.45
4	13	2.2	−42	0.42
6	12.5	2.25	−32	0.37
10	11	2.2	−21	0.26
20	6.5	2.2	−8.5	0.1

Pressure dependence of the parameters.—Popular wisdom has it that in a classical electron-phonon interaction scheme, a value of λ as large as reported in Table I would be impossible because a lattice instability should then occur before superconductivity could appear. But in heavy fermion systems, mass renormalizations of more than 100 times the free electron mass do exist, most likely due to magnetic interactions and in any case not to electron-phonon coupling. Obviously in most cases, these interactions do not favor superconductivity: few HF are superconducting. UBe_{13} is also necessarily a special case, because all other HF superconductors cannot be in a similar extreme strong coupling regime. This is shown, for example, by the very conventional behavior of their upper critical field, so that the pairing mechanism usually has to be different from the mass renormalization mechanism. The singularity of UBe_{13} is also reflected in the fact that the Fermi liquid regime is so ill defined at T_c in this compound, which is not proof of a strong coupling regime, but is compatible with it.

In this respect, the decrease of λ under pressure is consistent with the restoration of a well defined Fermi-liquid regime at T_c as observed in transport measurements [21]. More quantitatively, $\lambda(P)$ [mainly determined by $H_{c2}(0)$] gives the pressure dependence of m^* through the relation: $m^*(P)/m_{\rm band}(P) = \lambda(P) + 1$. If one further assumes that $m_{\rm band}(P)$ varies little in this pressure range, then $[\lambda(P) + 1]/[\lambda(0) + 1]$ is a measure of $m^*(P)/m^*(0)$. It is seen in Fig. 2 (solid squares) that this yields a decrease of m^* of about 50% between $P = 0$ and 20 kbar. A second independent estimate of $m^*(P)/m^*(0)$ can be extracted from the fit of the initial slope $(\partial H_{c2}/\partial T)_{T=T_c}$: m^* decreases by more than 60% in the same pressure range. This depends little on the model used to fit H_{c2}: it is essentially dictated by the experimental values of $(\partial H_{c2}/\partial T)_{T=T_c}$ (proportional to m^{*2}). This is nicely confirmed by a third measurement of m^*: the specific heat Sommerfeld coefficient γ, whose variation has been measured up to 9 kbar [22]. The agreement of the three independent estimates of $m^*(P)/m^*(0)$ can be seen on Fig. 2. The consistency of these results, on top of giving confidence in the validity of our model, also shows

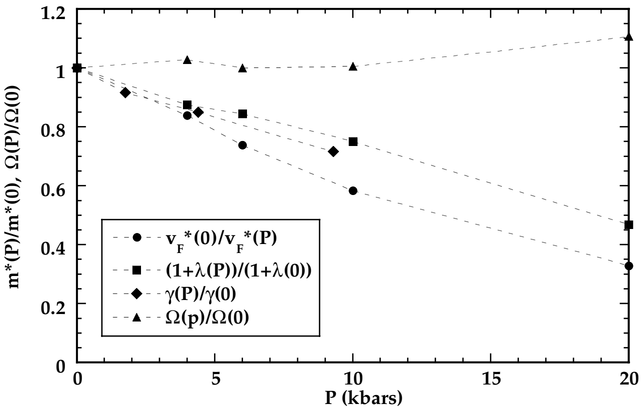

FIG. 2. $m^*(P)/m(0)$ obtained from $\lambda(P)$ (full squares); $(\partial H_{c2}/\partial T)_{T=T_c}$ (full circles); and the Sommerfeld constant $\gamma(P)$ [22] (full diamond). Full triangles: $\Omega(P)/\Omega(0)$. Dashed lines are guides to the eye.

that in UBe_{13} the mechanism responsible for the pairing in the superconducting state accounts for a large part of the mass renormalization in the normal state: about a factor of 16 at zero pressure. It gives a basis to the extreme strong coupling regime found in this compound and also yields a first quantitative experimental argument against electron-phonon coupling, because a mass renormalization of a factor of 16 by such a mechanism is extremely unlikely, especially with a Debye temperature of the order of 640 K [23].

Microscopically, λ is given by $\lambda = 2\int d\omega\, \alpha^2 F(\omega)/\omega$. So a large value of λ indicates an anomalous shift of the density of interactions at low frequencies, whereas higher frequencies are more favorable for an optimum value of T_c at a given spectral weight [19,24]. Indeed, we find an absolute value of $\Omega(0) \approx 1.5$ K, which is very small and close to T_c. $\Omega(P)/\Omega(0)$ is shown in Fig. 2. So $\mu*$, which is usually of the order of 0.1–0.2, might be much smaller in UBe_{13}: μ^* is some frequency integral of the Coulomb repulsion up to a cutoff proportional to Ω, and in any case, it is quite negligible compared to the value of λ. We also find that $\Omega(P) \approx$ const. It means that the whole variation of T_c stems from that of λ, whereas in ordinary superconductors, a variation of both $\lambda(P)$ and $\mu^*(P)$ is necessary to account for $T_c(P)$, and Ω is also generally found to only slightly increase with pressure. In this respect, $\Omega(P)$ cannot be directly related to the resistivity maximum (at 2.5 K at $P = 0$), which is the smallest energy scale known in UBe_{13} [1], because this maximum increases much faster with pressure [21]. At present, a microscopic identification of Ω seems hopeless due to the lack of neutron studies in this compound.

In conclusion, we have shown that it is possible to account for the whole (P, T) dependence of H_{c2} in UBe_{13} with an extreme strong coupling regime and only one arbitrary parameter: $\lambda(P)$. The variation of $\lambda(P)$ is consistent with the thermodynamics of the normal and of the superconducting phases. It shows that the pairing mechanism in UBe_{13} is also responsible for a large factor of the mass renormalization of the quasiparticles, which is too large to be reasonably attributed to the electron-phonon interaction. To our knowledge, this is the first quantitative indication of a nonphonon mediated pairing mechanism in any superconductor, making UBe_{13} a privileged candidate for further microscopic studies.

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