Implications of $T_c$ variation in UBe$_{13}$ for a possible Fulde-Ferrell-Larkin-Ovchinnikov phase

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We measure the heat capacity of UBe$_{13}$ with an unusually low $T_c$ for a polycrystal. We find an upturn in the upper critical field $H_{c2}(T)$ below about $T_c/2$, much as for higher-$T_c$ samples. Comparing the critical fields in our sample and in samples with higher $T_c$’s shows that the low-temperature limit of $H_{c2}$ is proportional to $T_c(H=0)$, as expected if the upturn comes from an FFLO phase and strong coupling.

Although discovered in 1983, the heavy-fermion superconductor UBe$_{13}$ still has many incompletely understood properties. These include the splitting of the superconducting transition upon doping the U sites with thorium, and the power law temperature dependences of various quantities in the superconducting phase. Another issue is the temperature dependence of the upper critical field $H_{c2}(T)$. Unique among heavy fermion superconductors, UBe$_{13}$ shows a clear upturn in $H_{c2}(T)$ at about half the superconducting transition temperature, with $H_{c2}(T=0)$ far exceeding the Clogston paramagnetic limit $T_c$. Indeed, the only other material with similar $H_{c2}$ behavior is the recently discovered UGe$_2$, which superconducts under pressure.

The two most detailed explanations of the upturn involve a mixture of two representations in the order parameter and an enhancement of the paramagnetic limit of $H_{c2}(T=0)$ by strong coupling. Here we show that the behavior of $H_{c2}(T)$ in samples of different $T_c$ is consistent with the latter explanation. Thomas et al. fit $H_{c2}(T)$ with three adjustable parameters: the slope $dH_{c2}/dT(T_c)$, which probes the orbital field limit; the gyromagnetic ratio $g$ of the quasiparticles; and the strong coupling parameter $\lambda$. Matching the upturn requires the further assumption of a Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state.

Several limits influence the low-temperature critical field. The orbital limit, $H_{c2}^{orb}(T) = \frac{\sqrt{\Delta^2(T)}}{g\mu_B}$, determines the critical field in most superconductors. The huge effective masses in heavy fermion superconductors make spin effects far more important than usual, although the orbital influence continues to dominate very close to $T_c$. A second field is the paramagnetic limit $H_{c2}^{p}$, also known as the Clogston or Pauli limit. For spin singlet pairing, the superconducting condensation energy competes with the magnetic energy tending to align the quasiparticle spins. In a single-particle excitation model, this leads to an abrupt quenching of superconductivity when the applied field reaches the Clogston limit $H_{c2}^{p}(T) = \frac{\sqrt{T\Delta(T)}}{g\mu_B}$, where $\Delta$ is the superconducting energy gap and $\mu_B$ is the Bohr magneton. Yet the Clogston limit is not the final word on spin limitations of $H_{c2}$. At high magnetic fields and $T$ less than about $T_c/2$, a Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state may appear in clean superconductors. $H_{c2}^{p}$ is derived in a single excitation picture, but multiple excitations interact by reducing the energy gap. In the FFLO state pairs are broken over an entire portion of the Fermi surface, with the resulting quasiparticles aligned by spin. The pairing over only part of the Fermi surface leads to an anisotropic energy gap with planar nodes perpendicular to the field. The energy difference between the spin-up and spin-down branches of the Fermi surface gives a finite center-of-mass pairing $Q$. Further energy considerations show that in fact the order parameter amplitude undergoes a real-space modulation. Since the FFLO state has partial spin alignment, its critical field $H_{c2}^{FFLO}$ exceeds $H_{c2}^{p}$.

These low-temperature critical field behaviors have different dependence on the sample’s transition temperature. The Clogston limit is proportional to the energy gap, leading to a proportionality to the zero-field transition temperature as well. The strong coupling parameter enters here through its effects on the gap. Similarly, the energetics of the FFLO state scale with the energy gap, and hence with $T_c$, so $H_{c2}^{FFLO}(t = T/T_c)$ should also be proportional to $T_c$. On the other hand, $H_{c2}^{orb}$ depends on the coherence length $\xi$, and its scaling with $T_c$ is not straightforward.

Fortunately, another unusual feature of UBe$_{13}$ allows us to test this scaling. Reported values for the onset of the superconducting transition vary by more than 200 mK, with high-quality crystals (as judged by the transition width) at both extremes of the range. Langhammer et al. suggest that two types

![Image](http://example.com/figure1.png)

**FIG. 1:** Heat capacity of our UBe$_{13}$ sample. The main figure is with no applied field, the inset at 8 Tesla.
of UBe$_{13}$ exist: L(low)-type, with $T_c$ around 750 mK; and H(high)-type, with $T_c$ from 850 to 950 mK [9]. Furthermore, polycrystals are supposedly all H-type, while single crystals fall into both categories. Here we show the $H_{c2}$ upturn in a polycrystal with a particularly low $T_c$. The only sample with such a low transition temperature previously studied had not shown the $H_{c2}$ upturn [8]. Comparing the shape of the upturn in samples of different $T_c$ supports the FFLO interpretation of the upturn.

Our sample was prepared by arc melting in an argon atmosphere and subsequent annealing at 1400$^\circ$C for 1000 hours in a beryllium atmosphere [9]. It is polycrystalline with a typical grain size of 100 $\mu$m, as seen in a Philips CM-30 transmission electron microscope. We use a relaxation method to measure heat capacity $C(T)$, shown in Figure 3 from 100 mK to 900 mK. The superconducting transition width is small, about 50 mK, speaking to a high sample quality despite the low $T_c$.

The inset of Figure 1 shows heat capacity in our highest magnetic field, 8 Tesla, measured on a small piece cut from the same sample as that for the main figure. This piece is less than 20 microns thick and less than 1 mm in lateral dimensions. Because of its small size, the background heat capacity is significant but the transition remains clearly visible. The piece was made thin for irradiation with high-energy uranium ions to study the influence of lattice disorder on the superconducting phases [10]. Although the irradiation shows no measurable effects up to a density of $10^{15}$ tracks per square meter, we performed various measurements as a function of applied field while searching for matching effects at fields where the defect density equals the vortex density. The results presented here are from the irradiated sample which we studied in the most detail, with track density $5 \times 10^{13}$/m$^2$. This sample’s zero-field $T_c$ and transition width are identical to those of the unirradiated samples.

Figure 2 combines our own data with previous measurements on a higher-$T_c$ sample [8]. Since the criterion for identifying $T_c$ can change the curve shape of $H_{c2}(T)$ [9], both our data and the high-$T_c$ comparison curve use $T_c$ as the transition midpoint of heat capacity measurements. The transition widths of the two samples are also comparable. Both curves display the characteristic upturn in $H_{c2}$. The temperature of the upturn tracks the zero-field $T_c$, remaining near $T_c/2$. The shift we observe in the temperature of the upturn with $T_c$ is consistent with an FFLO explanation.

Tc variation include aluminum impurities, and variations in the beryllium content, which must be carefully monitored during the preparation process [9]. Previously only single crystals had been reported with such low transition temperatures, and explanations focused on differences between single crystal and polycrystal preparation. However, this seems to be a consequence of reporting $T_c$ as the onset rather than midpoint of the transition.

![FIG. 2: Upper critical field as a function of temperature for our sample (diamonds) and from Reference 8 (circles).](image)
In our sample, with its narrow transition, midpoints and onsets for our sample and for those from several previously published works. Except as otherwise noted, we extracted these numbers from heat capacity graphs in those papers. Resistive measurements generally yield slightly higher values, but the large span of \(T_c\)’s remains. We note that the transition midpoints, for both single crystals and polycrystals, are widely distributed between 725 mK and 930 mK. Since previous polycrystalline samples with low \(T_c\)’s also had wide transitions, the \(T_c\) onsets were all near 900 mK. In our sample, with its narrow transition, \(T_c\) is clearly much lower. Since the samples with the narrowest transitions do not have the highest onset temperatures, identifying the midpoint temperatures is reasonable.

We find an upturn in \(H_{c2}(T)\) below about \(T_c/2\), a feature previously seen only in UBe\(_{13}\) samples with \(T_c\) around 900 mK. Comparing the shape of \(H_{c2}(T)\) for samples with different \(T_c\)’s meshes well with interpreting the upturn as an FFLO state in a strong coupled superconductor. As \(T_c\) changes the paramagnetic limit for critical field scales with \(T_c\), which we find for \(H_{c2}\) at low temperatures. The orbital limit depends on the superconducting coherence length and changes with \(T_c\) in a less straightforward way, in agreement with our measurements near \(T_c\). Observing the critical field upturn in a low \(T_c\) sample does raise a question about the role of disorder in causing the wide \(T_c\) range. If the upturn indicates an FFLO phase, which requires a clean limit, then the upturn should be suppressed in low \(T_c\) samples, as it is in thorium-doped UBe\(_{13}\). The resolution probably depends upon a better understanding of the \(T_c\) variation itself. Our measurements on a polycrystalline UBe\(_{13}\) sample with \(T_c = 770\) mK and our compilation of past work present evidence for the existence of only one type of UBe\(_{13}\) with a range in \(T_c\) between 725 mK and 930 mK.

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In Table I we list both midpoints and onsets for our sample and for those from several previously published works. Except as otherwise noted, we extracted these numbers from heat capacity graphs in those papers. Resistive measurements generally yield slightly higher values, but the large span of \(T_c\)’s remains. We note that the transition midpoints, for both single crystals and polycrystals, are widely distributed between 725 mK and 930 mK. Since previous polycrystalline samples with low \(T_c\)’s also had wide transitions, the \(T_c\) onsets were all near 900 mK. In our sample, with its narrow transition, \(T_c\) is clearly much lower. Since the samples with the narrowest transitions do not have the highest onset temperatures, identifying the midpoint temperatures is reasonable.

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### Table I: Transition temperatures of UBe\(_{13}\) measured by heat capacity for various samples. Both the midpoint and onset of the transition are given.

| Reference | Single crystals | | Midpoint (mK) | Onset (mK) | Width (mK) |
|-----------|----------------|---|-------------|-------------|------------|
| [15]      |                |   | 725         | 770         | 90         |
| [16]      |                |   | 745         | 780         | 70         |
| [8]       |                |   | 744         | 768         | 48         |
| [1]       |                |   | 790         | 880         | 180        |
| [17]      |                |   | 860         | –           | < 96       |
| [5, 8]    |                |   | 900         | 920         | 40         |
| [13]      |                |   | 907         | 950         | 86         |
| Polycrystals |            | | 765         | 790         | 50         |
| This work |                | | 830         | 910         | 160        |
| [19]      |                | | 830         | 885         | 110        |
| [20]      |                | | 885         | 945         | 120        |
| [14]      |                | | 915         | 980         | 130        |
| [17]      |                | | 919         | –           | < 40       |
| [21]      |                | | 930         | 970         | 80         |

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