Thermodynamic features in the $H$-$T$ plane of superconducting UBe$_{13}$

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We present specific-heat $C$ measurements of a high-quality polycrystal of UBe$_{13}$ for $0.2 \leq T \leq 1.2$ K and $0 \leq H \leq 80$ kG. Field-sweep data, $C(H)$ at fixed $T$, reveal two broad peaks in the superconducting state for $T < 0.6$ K. The low-field feature occurs at $H \approx 20$ kG, independent of temperature. Conventional temperature scans, $C(T)$ at fixed $H$, presumably smear this feature beyond recognition.

The spin degrees of freedom play an essential role in heavy-fermion superconductivity. In UPt$_3$, not only may spin fluctuations provide the pairing mechanism, but the existence of antiferromagnetic order below $T_c$ (with a magnetic moment $\sim 0.02\mu_B$) allows the possibility of multiple superconducting phases. Similarly, recent magnetic x-ray and neutron-scattering studies of UR$_2$-Si$_2$ establish the microscopic coexistence of antiferromagnetism (with a magnetic moment $\sim 0.04\mu_B$) and superconductivity. Magnetic-susceptibility and heat-capacity measurements on thoriated CeCu$_2$Si$_2$ show a significant bulk ordered moment coexisting with the superconductivity in that heavy-fermion system.

The situation for UBe$_{13}$ is less settled. When doped with thorium, magnetic correlations arise and for Th substitution between 2% and 4% two transitions are observed in the low-temperature specific heat. For pure UBe$_{13}$, magnetostriction data indicate the onset of antiferromagnetic order at $T_N \approx 8.8$ K $\sim 10T_c$, but there is no direct evidence to date for magnetic ordering below $T_c$. On the basis of muon spin resonance and low-field measurements of superconducting UBe$_{13}$, Heffner et al. rule out either magnetism or a second phase transition at low fields below $T_c$. In contrast, Rauchschwalbe suggests two superconducting order parameters for UBe$_{13}$ from an analysis of a break in the upper critical-field slope at $H \sim 20$ kG. We report here multiple thermodynamic signatures in pure UBe$_{13}$ for $T/T_c < 0.6$ at $H \approx 20$ kG.

The key difference in our heat-capacity measurements is that we fix temperature $T$ and sweep magnetic field $H$, in contrast to the conventional approach of fixing $H$ and varying $T$. Varying the magnetic field introduces the technical complexity of recalibrating the thermometer attached to the sample at every one of typically 8 magnetic-field points. It provides, however, sensitivity to thermodynamic features when the phase boundary of interest is essentially parallel to the temperature axis in the $H$-$T$ plane, features which would be smeared beyond recognition in a temperature sweep at fixed $H$. The specific heat $C(H)$ at fixed $T$ has proved successful in revealing sharp features in the $H$-$T$ plane of UPt$_3$, even in a sample with one broad peak in $C/T$ vs $T$ at any $H$. Furthermore, the functional form of $C(H)$ for $T < T_c$ provides information about the nature of the excitation spectrum in the superconducting state.

The UBe$_{13}$ sample used in our work is an arc-melted, high-purity polycrystal of mass 0.08 g. Preparation specifics have been provided elsewhere. In zero magnetic field, the superconducting transition temperature $T_c = 0.925$ K, the Sommerfeld constant $\gamma = 1.0$ $\text{J/mol K}$, and the specific-heat jump at $T_c$, $\Delta C/C = 1.8$ (see Fig. 1). The experiments were performed using standard heat-pulse techniques in a helium dilution refrigerator for $0.2 \leq T \leq 1.2$ K and $0 \leq H \leq 80$ kG. As discussed above, the thermometer was a carbon chip whose magnetoresistance made it necessary to recalibrate it at every magnetic-field point during field sweeps. The heater was made of Au/Cr and was field independent to better than 0.03% over the entire magnetic-field range. The nuclear Zeeman contribution to the specific heat from Be (non-negligible for $H > 40$ kG at the lowest temperatures) has been subtracted from all the data.

We plot in Fig. 2 a series of specific-heat field sweeps for $T < T_c (H = 0)$. At the higher temperatures it is possible to determine the upper critical field, delineated by the essentially field-independent response of $C(H)$ in the normal state. $H_{c2}$ moves out of our magnetic-field window, however, by $T = 0.4$ K. At the lower temperatures ($T \leq 0.6$ K), $C(H)$ rises with increasing magnetic field as the vortices supply quasiparticles, dropping to the normal-state value of the specific heat with the approach...
FIG. 2. Specific heat $C$ of UBe$_{13}$ as a function of magnetic field $H$ at a series of temperatures $T < T_c (H=0)$. $C(H)$ is essentially independent of $H$ in the normal state. Two peaks appear in the superconducting state for $T < 0.6 \text{ K}$, most clearly distinguished at $T = 0.5 \text{ K}$.

to $H_c$. The quadratic form of $C(H)$ as $H \rightarrow 0$ apparent from $T = 0.2$ and $0.3 \text{ K}$ gives way to a more linear dependence on magnetic field for larger $T/T_c (H=0)$. At corresponding values of $T/T_c (H=0)$ in UPt$_3$ (Ref. 11) $C(H)$ is also proportional to $H$, suggesting a similar excitation spectrum.

The UBe$_{13}$ data of Fig. 2 reveal an additional feature in $C(H)$ at an intermediate field, $H = 20 \text{ kG}$, for $T < 0.6 \text{ K}$. This secondary maximum can be observed most clearly at $T = 0.5 \text{ K}$, but it can be discerned as well at the three lower temperatures. We plot in Fig. 3 a putative phase diagram for UBe$_{13}$ in the $H$-$T$ plane based on the structure

do the upper critical field, taken as the midpoint of the sharp rise in $C(H)$ at large $H$. The open circles mark the position of the partially buried peak at lower field as a function of $T$.

Although the origin of multiple thermodynamic features in magnetic field and temperature for UBe$_{13}$ is not known, we can make some general observations regarding Fig. 3. First, it is clear from the nearly temperature-independent behavior of the lower boundary that cuts in $C(H,T)$ along $H$, as reported in this experiment, are required to see it. Second, the position of the lower boundary at $H = 20 \text{ kG}$ coincides with the change in slope of the upper critical-field curves reported previously by at least three groups.\textsuperscript{10,13,14} Such a kink is consistent with

our $H_c$ data of Fig. 3; additional points would be required to define it conclusively on the basis of specific-heat data. The boundary at $H = 20 \text{ kG}$ also coincides with an anomaly in the magnetostriction observed\textsuperscript{8} at $T = 0.6 \text{ K}$. Third, the appearance of two features below $T = 0.6 \text{ K}$ is at the same temperature posited for the emergence of a second superconducting order parameter in the Rauchschwalbe analysis\textsuperscript{16} of $H_c$ data. However, that proposed phase diagram predicts a lower boundary at $H = 70 \text{ kG}$, in contrast to the calorimetric features at $H = 20 \text{ kG}$.

If there are indeed two distinct phases, then the question remains as to their nature, particularly in view of the precedents in other heavy-fermion compounds of the microscopic interplay of magnetism and superconductivity. The work of Heffner \textit{et al.}\textsuperscript{9} indicates that the lower phase cannot have a significant local magnetic moment ($\mu < 0.01\mu_B$). Many alternatives still exist, however, given the likely higher-order pairing in UBe$_{13}$. The possible role of quadrupolar coupling\textsuperscript{15} may also predispose the system to a magnetic-field-induced crossover. The effects of symmetry-breaking fields other than the magnetic field, such as uniaxial stress,\textsuperscript{16} may be able to help narrow the possibilities.

In summary, specific-heat measurements as a function of $H$ at fixed $T$ reveal both the transition into the normal state at $H_c(T)$ and a secondary maximum at $H = 20 \text{ kG}$ for $T < 0.6 \text{ K}$. The lower-field feature occurs at a kink in the slope of $H_c(T)$ and suggests the possibility of distinct phases in superconducting UBe$_{13}$.

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