Magnetization in the Superconducting Mixed State of the Heavy-Fermion Compound UBe$_{13}$

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Abstract.
Static dc magnetization measurements in the superconducting mixed state of a single crystal UBe$_{13}$ along the [110] axis were performed by means of a capacitance Faraday-force method down to 0.24 K. Below the upper critical field $B_{c2}$, not only a peak effect but also an additional broad anomaly have been observed in magnetization curves. We report superconducting phase diagram of UBe$_{13}$ obtained by our magnetization measurements, including these anomalies.

1. Introduction
The heavy-fermion compound UBe$_{13}$ (cubic, $O_h^6$(Fm$ar{3}$c)) has attracted much attention since the discovery of non-BCS type superconductivity (SC) 27 years ago [1]. The SC transition of this compound takes place at about 0.85 K with a large specific-heat jump $\Delta C$, which is the same order of the electronic specific heat $C \sim 1$ J/Kmol observed just above $T_c$. This indicates that the heavy electrons form the Cooper pairs which condense into the SC state. In order to understand the unconventional SC of UBe$_{13}$, it is very important to clarify the symmetry of the Cooper pairs and investigate the SC phase diagram precisely. First, as for the SC symmetry, an early specific-heat study made by Ott et al. [2] has suggested a $p$-wave Anderson-Brinkman-Morel (ABM) state from $T^3$ behavior of the specific heat well below $T_c$, because it is similar to the behavior observed in the ABM state of $^3$He superfluidity. NMR spin-relaxation rates [3] and ultrasonic velocities [4] have also suggested that the SC gap has a nodal structure. These results indicate that there is a line node in the SC gap, whereas the ABM state has point nodes. In addition, it remains also unclear whether the spin state of a Cooper pair is singlet or triplet. A recent $\mu^+$-Knight shift study [5] in $B||[001]$ has suggested that the spin susceptibility decreases below $T_c$, indicating a spin-singlet pairing or a spin-triplet pairing with a strong spin-orbit coupling. On the other hand, a recent NMR study [6] has suggested that the Knight shift for $B||[001]$ shows no decrease below $T_c$, which indicates the spin-triplet pairing. In this way, no clear experimental consensus has been obtained about not only the SC gap structure but also the Cooper pairing state in UBe$_{13}$. Second, it is noted that the SC phase diagram of UBe$_{13}$ is quite
different from that of the conventional BCS superconductors. One of the unusual properties is the upper critical field curve $B_{c2}(T)$ that exhibits a clear upturn at low temperatures [7]. Moreover, low-$T$ specific-heat and thermal-expansion measurements by Kromer et al. show a new anomaly at $T_L$ below $T_c$ for pure UBe$_{13}$ in zero field. In field scans of $C(B, T = \text{const.})$, this anomaly corresponds to $B^*(T)$. They have proposed that this anomaly is a precursor of another phase transition observed in the U$_{1-x}$Th$_x$Be$_{13}$ system at $T_{c2}$ [8], because it seems that $T_L(x)$ smoothly merges into $T_{c2}(x)$ on a $T$-$x$ phase diagram [9, 10].

In order to obtain further information about the unusual SC of UBe$_{13}$, we have performed low-temperature dc magnetization measurements in the SC mixed state of UBe$_{13}$ using a high-quality single crystal.

2. Experimental Procedure
A single crystal of UBe$_{13}$ was grown by an Al-flux method, and its weight was 6.6 mg. The SC transition temperature is about 0.8 K, which is defined by the peak-top temperature of the specific-heat jump for zero field. The dc magnetization measurements along $[110]$ were performed by using a capacitive Faraday-force magnetometer in a $^3$He-$^4$He dilution refrigerator [11] at temperatures down to 0.24 K and in fields up to 6.9 T. Magnetization processes were measured in a field gradient of 500-900 Oe/cm applied in addition to the uniform magnetic field.

3. Results and Discussion
Figure 1 shows the magnetization process of UBe$_{13}$ observed for $B||[110]$ at 0.24 K up to 6.9 T. Below $\sim 2$ T, a clear irreversibility attributed to a vortex flux pinning is observed. On the other hand, the irreversibility decreases with increasing field above $\sim 2$ T. The smallness of this hysteresis in high field indicates a high-quality of the used sample, since flux-pinning centers are impurities or lattice defects in general. Here, we define $B_{c2}$ as the field where the irreversibility vanishes completely. The linear component above $B_{c2}$ is the normal state magnetization $M_n$. Just below $B_{c2}$, a peak effect, which is occasionally seen in type-II superconductors, is observed. We define the onset of the peak as $B_{\text{peak}}$. Figure 2 shows hysteresis magnetization curves $\Delta M$ measured at various temperatures: $\Delta M = (M_{\text{dec}} - M_{\text{inc}})/2$, where $M_{\text{dec}}$ and $M_{\text{inc}}$ are the magnetization of decreasing- and increasing-field processes, respectively.

Figure 1. Magnetization curves of the single crystalline UBe$_{13}$ at 0.24 K ($B||[110]$). The upper critical field $B_{c2}$ is pointed by the arrow. $B_{\text{peak}}$ denotes the onset of the peak effect.

Figure 2. The hysteresis magnetization $\Delta M$ of UBe$_{13}$ in various temperatures ($B||[110]$). Down and up arrows indicate $B_{\text{peak}}$ and $B_{c2}$, respectively.
increases, the peak becomes indistinctive due to the interference with a large hysteresis in the low-field region. The peak effect has been observed also in other heavy-fermion superconductors such as UPt₃ and CeCoIn₅ [12, 13]. In particular, UPt₃ shows the peak effect just below B_c₂, which is similar to the behavior of UBe₁₃.

Since the hysteresis of the magnetization is sufficiently small in the high-field region, we can approximately obtain the thermal equilibrium magnetization M_eq in the mixed state by averaging the increasing- and decreasing-field processes. The M_eq consists of the normal state component M_n and the diamagnetic magnetization component M_eq^SC: M_eq = M_n + M_eq^SC. We estimated M_n in the mixed state by extrapolating the normal state magnetization above B_c₂ to the origin, assuming the linear region: M_n = \chi_n B. Figure 3 shows the M_eq^SC obtained at 0.24 K and 0.59 K. For the M_eq^SC at each temperatures, there are two bends below B_c₂ (at \sim 4 T and \sim 5 T, for 0.24 K). First, we note that M_eq^SC bends at a field clearly lower than B_peak. This anomaly is broad but distinct. We define this broad anomaly as B_M^*. At nearly the same field \sim 4 T, the specific heat and thermal expansion measurements [9, 10] have suggested that there is an anomaly at B^* below B_c₂, as mentioned in Introduction. Ac-susceptibility (ac-\chi) also shows an anomaly at \sim 4 T for T \sim 0.25 K [14]; we call this anomaly B_{ac}^* in order to distinguish it from B^* obtained by Kromer et al. Second, we note another bend observed in M_eq^SC near B_peak (at \sim 5 T for 0.24 K). This behavior, however, does not necessarily indicate an occurrence of a phase transition: if there is a slight deference in the magnitude of the magnetization peaks between increasing- and decreasing-field processes, then the bend may appear, except for a phase transition. Therefore, this bend at B_peak in M_eq^SC may not be intrinsic.

Figure 4 represents the obtained SC phase diagram of UBe₁₃ for B || [110], including our specific-heat results and the previous studies [9, 10, 14].

As seen in Figure 4, B_M^* gets closer to B_peak above \sim 0.5 K. In fact, above \sim 0.6 K, we could
not distinguish $B^*_M$ from $B_{peak}$. Therefore, this disagreement may be caused by the difficulty of distinguishing $B^*_M$ from $B_{peak}$ above $\sim 0.5$ K, and $B^*_{ac}$ above $\sim 0.5$ K may correspond to $B_{peak}$. Anyway, all works have suggested that there is at least an anomaly in the SC phase diagram in the same field range $B \lesssim 4.5$ T.

On the other hand, the scale of $B_{c2}$ obtained from our measurements disagree with that in [9, 10] and [14]. Here, we can guess that there is little anisotropy of $B_{c2}$, because of the lack of anisotropy at least between $B||[100]$ and $B||[110]$, which has been suggested from the ac-$\chi$ measurements [14]. In particular, although $B^*_M$ is almost consistent with $B^*$ obtained by Kromer et al., the scale of the $B_{c2}(T)$ curve obtained for the present sample is obviously smaller than that of refs. [9] and [10]. If $B^*_M$ and $B^*$ are due to the same origin, the above comparison suggests that the scale of $B^*$ is independent of that of $B_{c2}$. This may indicate that the anomaly at $B^*$ originates from some phenomenon which is not basically associated with the SC. As for the origin of $B^*$, the following two cases may be considered: (1) a some magnetic correlation which coexists with the SC [9]; (2) a flux-pinning mechanism [14]. If $B^*$ is orginated from the case (1), the scale of $B^*$ may not necessarily depend on that of $B_{c2}$. In contrast, if $B^*$ is orginated from the case (2), temperature dependence of $B^*$ would follow $B_{c2}(T)$ as seen in the behavior of $B_{peak}(T)$. Therefore, the proposal that $B^*$ is orginated from the magnetic correlation coexisting with the SC might be constant with our results. In order to uncover the relationship between $B^*$ and the unusual SC of UBe$_{13}$, further studies such as a check of the sample-quality dependence will be needed.

4. Summary
The superconducting phase diagram of a single-crystalline UBe$_{13}$ for $B||[110]$ has been obtained by means of the static dc magnetization measurements down to 0.24 K. In addition to the peak effect just below the upper critical field $B_{c2}$, we found a broad anomaly at $B^*_M$, which is clearly below $B_{peak}$. In contrast to the disagreement of the $B_{c2}$ curves, the observed $B^*_M$ values are nearly comparable to the fields where specific heat and thermal expansion show an anomaly.

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