Anomalous Upper Critical Field in the Heavy-Fermion Superconductor UBe$_{13}$ Studied by DC Magnetization Measurements

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Abstract. We have studied anomalous upper critical field $B_{c2}(T)$ of a single crystal of UBe$_{13}$ by DC magnetization measurements down to 0.14 K and up to 8 T. We have observed a broad anomaly and a peak effect below $B_{c2}$ in magnetization curves. Although a possibility of the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state has been proposed as an origin of the unusual upper critical field by previous works, our results have shown that thermal equilibrium magnetization curves reveal no step-like behavior, which could be observed at 1st-order phase transition from a superconducting mixed state to the FFLO state. We have also investigated anisotropy of $B_{c2}$ for $B \parallel <001>$ and $B \parallel <110>$, and found that $B_{c2}$ for $B \parallel <001>$ is larger than $B_{c2}$ for $B \parallel <110>$ below $\sim 0.5$ K. These results might indicate that the anomalous upward curvature of $B_{c2}$ in UBe$_{13}$ is related to anisotropy of $B_{c2}$ rather than the FFLO state.

1. Introduction

The heavy-fermion system UBe$_{13}$ is one of the most exotic superconducting materials in uranium compounds. After the discovery of this unconventional superconductivity by Ott et al. [1], it has been shown that this material has an anisotropic superconducting gap structure by specific heat $C(T)$ [2], nuclear magnetic resonance (NMR) spin-relaxation rate $1/T_1$ [3], and ultrasonic attenuation $\alpha(T)$ [4], and magnetic field penetration depth $\lambda(T)$ [5]. However, in fact, no consensus has been obtained about the superconducting gap structure of UBe$_{13}$. As for the parity of Cooper pairing, it has been said that Cooper pairing state of UBe$_{13}$ is triplet because Knight shift does not decrease below $T_c$ [6]. However, a recent muon spin rotation ($\mu^+\text{SR}$) study for $B \parallel <001>$ has shown that Knight shift decreases below $T_c$, and recently Tou et al. have reported that NMR Knight shift exhibits an anisotropic behavior below $T_A$, which is lower than $T_c$. Therefore, the parity of superconductivity in UBe$_{13}$ remains unclear even now.

UBe$_{13}$ reveals an anomalous upper critical field curvature $B_{c2}$ which can not be explained in a framework of the Bardeen-Cooper-Schrieffer (BCS) theory. First, the $B_{c2}$ of UBe$_{13}$ is much larger than the Pauli limit $\mu_0H_P(T=0)$ of 1.84 $T_c$ T ($\sim 1.66$ T for $T_c = 0.9$ K) [9]. On the other hand, the value of $B_{c2}$ for UBe$_{13}$ estimated experimentally by extrapolation to $T = 0$ is 8-13 T. Second,
$B_{c2}(T)$ of UBe$_{13}$ shows an upturn-like behavior at around $T_c/2$. So far, several explanations for this unusual $B_{c2}$ curve have been proposed, including two-coexisting superconducting order parameters [10], a recovering of coherent Fermi-liquid state in the normal state [11], and the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state [12]. In particular, the FFLO state, in which a Cooper pair has a non-zero momentum $q$, is generally possible in a singlet superconductor with a strong paramagnetic effect [13]. Therefore, it is noteworthy to clarify whether the unusual upward curvature of UBe$_{13}$ is due to the appearance of the FFLO state or not. In addition, anisotropy of $B_{c2}$ in UBe$_{13}$ is also important to clarify the superconducting symmetry and still in discussion [14, 15, 16].

In the present work, we have performed low temperature DC magnetization measurements for a single crystal of UBe$_{13}$ in order to elucidate the origin of anomalous upper critical field $B_{c2}$ of UBe$_{13}$. We have also investigated anisotropy of $H_{c2}$ for $B \parallel 001 >$ and $B \parallel 110 >$.

2. Experimental Procedures

A single crystal of UBe$_{13}$ was grown by an Al-flux method reported in [17]. Previous low-temperature DC magnetization [18] and specific-heat measurements [16] were measured for the same single crystal. The superconducting transition temperature of the sample is 0.81 K from specific-heat measurement in zero field obtained by entropy conservation analysis [16]. The static DC magnetization measurements were carried out by using a capacitive Faraday-force magnetometer installed in a $^3$He-$^4$He dilution refrigerator [19] down to $\sim 0.14$ K. The measurements have been done in a field gradient of 5-9 T/m applied in addition to the uniform magnetic field up to $\sim 8$ T. Since the dimension of the used sample is $1.8 \times 1.9 \times 0.8$ mm$^3$, distribution of the magnetic field due to the field gradient is estimated to be less than $\sim 18$ mT in a field gradient of 9 T/m.

3. Results and Discussion

Figure 1 shows the magnetization curves of UBe$_{13}$ in the superconducting state at 0.18 K for $B \parallel 001 >$. Up and down arrows denote increasing and decreasing processes, respectively. An irreversibility observed in low-field region below $\sim 2$ T is due to a vortex flux pinning, which is typically seen in type-II superconductors. $B_{c2}(= \mu_0 H_{c2})$ can be defined as a field where the hysteresis of magnetization vanishes completely. Almost linear component of above $B_{c2}$ is the normal state magnetization; there is a small contribution of non-linear susceptibility, $1/3! \chi_3 B^3$, as well as that of linear susceptibility, $\chi_1 B$. Assuming that the normal-state susceptibility in the superconducting mixed state due to vortices is the same as that in the normal state above $B_{c2}$, we obtain the contribution of superconducting diamagnetic magnetization by subtracting the normal-state magnetization. Figure 2 shows a diamagnetic magnetization curves in the mixed state at 0.18 K for $B \parallel 001 >$. Since the hysteresis of magnetization is small in our sample, we can obtain a thermal equilibrium magnetization $M_\text{eq}^{\text{SC}}$ (Red Marker) by averaging increasing and decreasing processes. We find that the thermal equilibrium magnetization process reveals a minimum in the mixed state. In other words, the absolute value of superconducting diamagnetization exhibits a maximum in the mixed state. In this work, we define $B_{\text{Mag}}^{*}$ as the field where the diamagnetic response shows the maximum. The temperature dependence of $B_{\text{Mag}}^{*}$ is similar to that of $B^*(T)$, which is observed in a magnetic field dependence of the specific heat and temperature dependence of thermal expansion [20]. This magnetization curve is extremely anomalous because an absolute value of magnetization in a type-II superconductor exhibit a maximum at lower critical field $B_{c1}$ and then decreases with increasing magnetic field monotonically in general. We have observed this maximum of diamagnetic response also for $B \parallel 110 >$, and $B_{\text{Mag}}^{*}$ for $B \parallel 110 >$ is larger than that for $B \parallel 001 >$. Further study for this broad anomaly will be reported in another paper.
Next, we remark about a possibility of the FFLO state in UBe$_{13}$. In a type-II singlet superconductor with a strong paramagnetic effect, the FFLO state could occur in high-field region of superconducting phase. In this case, its superconducting diamagnetic curve should exhibit a step-like behavior due to a 1st-order phase transition from the mixed state to the FFLO state [21]. However, we could not observe such a step-like behavior in the magnetization curves. Therefore, the results of our magnetization measurements do not support a possibility of the FFLO state in UBe$_{13}$. The FFLO state due to a strong paramagnetic effect occurs usually for even-parity Cooper pairs which are formed by anti-parallel spins. Therefore, our results are consistent with the possibility of an odd-parity (triplet) superconductor in which Cooper pairs have parallel-spin parings.

Another key to solve the unusual superconductivity in UBe$_{13}$ is anisotropy of $B_{c2}$. Figure 3 (a) and (b) shows magnetization curves at 0.50 K and 0.18 K for $B || <001>$ and $B || <110>$ in high-field regions, respectively. The $B_{c2}$ is almost isotropic within the experimental error at $\sim 0.5$ K. This is consistent with the results of our previous specific-heat measurements [15, 16]. However, at 0.18 K, we have found an anisotropic $B_{c2}$ in UBe$_{13}$ between $B_{c2}$ for $B || <001>$ and that for $B || <110>$. This anisotropy begins to appear below 0.4-0.5 K, and becomes larger with decreasing temperature. Figure 4 shows the temperature dependence of the difference between $B_{c2}^{<001>}$ and $B_{c2}^{<110>}$. Here, we define $\Delta B_{c2}$ as $B_{c2}^{<001>} - B_{c2}^{<110>}$. This anisotropy of $B_{c2}$ is sufficiently larger than the error of the magnetic field due to distribution of gradient field. This anisotropy might be associated with the upturn of the $B_{c2}$ in UBe$_{13}$, which is observed at around $T_c/2$. It is needed to understand why the upper critical field of UBe$_{13}$ is anisotropic only at low-temperature in order to elucidate the origin of anomalous upward upper critical field and its superconducting symmetry.

4. Conclusion
We have performed static DC magnetization measurements for a single crystal of UBe$_{13}$ and observed the broad anomaly in the superconducting mixed state and the peak effect just below...
$B_{c2}$. The thermal equilibrium magnetization reveals no step-like behavior in the superconducting phase. On the other hand, the anisotropy of $B_{c2}$ for between $B \parallel <001>$ and $B \parallel <110>$ has been observed only below $\sim 0.5$ K, where $B_{c2}<001>$ is larger than $B_{c2}<110>$. These facts indicate that the upward curvature of $B_{c2}(T)$ in UBe$_{13}$ is probably originated from the low-temperature anisotropy of $B_{c2}$ rather than the occurrence of the FFLO state.

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