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Observation of an Unusual Magnetic Anomaly in the Superconducting Mixed State of Heavy-Fermion Compound UBe$_{13}$ by Precise dc Magnetization Measurements

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We have performed precise dc magnetization measurements for a single crystal of UBe$_{13}$ down to 0.14 K, up to 80 kOe. We observed a magnetic anomaly in the superconducting (SC) mixed state at a field, named H^{*}_{Mag} ($\sim$ 26 kOe, at 0.14 K), implying that UBe$_{13}$ has a magnetically unusual SC state. We studied the magnetization curves of UBe$_{13}$, assuming that the H^{*}_{Mag} anomaly originates from (1) and unusual SC diamagnetic response, or (2) a peculiarity of the normal-state magnetization due to vortices in the SC mixed state. The origin of the H^{*}_{Mag} anomaly is discussed.

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Unconventional superconductivity (USC), which cannot be explained in the framework of the Bardeen-Cooper-Schrieffer (BCS) theory, has been found in heavy-fermion compounds, high-T_{c} cuprates, and organic conductors. Among them, uranium heavy-fermion superconductors show an extremely exotic superconducting (SC) nature, as represented by UBe$_{13}$ [1], UPt$_3$ [2], URu$_2$Si$_2$ [3], UGe$_2$ [4], and URhGe [5]. In order to elucidate the SC mechanism of these novel superconductors, it is crucial to unravel the origin of various non-BCS behaviors, including anisotropic SC gap structures, unusual upper-critical field H_{c2} [6,7], a multi-SC diagram [8], and a manner of coexistence of magnetism and superconductivity [4,5,9,10].

UBe$_{13}$ has a cubic structure with space group O_h^6 (Fm$\bar{3}$c), and is one of the most exotic and mysterious superconductors ever found ($T_{\mathrm{c}} \sim 0.8$–0.9 K) [1]. There are two major problems in terms of study on SC properties of UBe$_{13}$: (i) SC symmetry, including the nodal SC gap structure and the parity of Cooper pairing in UBe$_{13}$ [11–14], (ii) an additional anomaly (multiple phase) in the SC state in thorium(Th)-doped and pure UBe$_{13}$.

Ott *et al.* discovered that U$_{1\text{-}x}$Th$_x$Be$_{13}$ shows a second transition at T_{c2} below a SC transition at T_{c1} only for 2–4% Th-doped samples [15], as well as the nonmonotonic x dependence of its SC transition temperature [16]. For this, whereas an ordering of antiferromagnetic (AF) spin-density wave (SDW) was proposed from ultrasonic studies [17], two coexisting SC order parameters in Th-doped and pure UBe$_{13}$ were also proposed [18]. Furthermore, Heffner *et al.* observed a weak magnetism below T_{c2} in U$_{1\text{-}x}$Th$_x$Be$_{13}$ ($0.019 < x < 0.045$) by zero-field μSR measurements [9,10]. It is still debated whether the weak magnetism originates from AF SDW [17] or an additional SC state with broken time-reversal symmetry such as a nonunitary state [19].

Kromer *et al.* found a line of anomaly $T_{\mathrm{L}}(x)$ which smoothly merges into $T_{c2}(x)$ on the T-x phase diagram of U$_{1\text{-}x}$Th$_x$Be$_{13}$ from thermal-expansion $\alpha(T)$ and specific-heat $C(T)$ measurements [20]. They also reported that a broad anomaly at B^* in isothermal specific-heat $C(B)$ corresponds to $\alpha(T)$ anomaly at T_{L}. They proposed that the anomaly at T_{L} is a precursor of the second transition at T_{c2}, and T_{L} and B^* originate from a short-range AF ordering [20]. However, the origin of $B^*(T)$ anomaly and the weak magnetism still remains unclear. Also, how is this anomaly related to the USC in UBe$_{13}$?

Our motivation for the present study is further understanding of the USC in UBe$_{13}$, regarding its magnetic properties. In this Letter, we report precise dc magnetization $M(H)$ measurements at very low temperature below T_c on a single crystal of UBe$_{13}$ for $H \| \langle 001 \rangle$.

A single crystal of UBe$_{13}$ was grown by an Al-flux method [21]. $M(H)$ curves were measured down to 0.14 K and up to 80 kOe with a field gradient of 500–900 Oe/cm by using a capacitive Faraday-force magnetometer [22] installed in a ^{3}He-^{4}He dilution refrigerator. Here, in order to subtract the slight magnetic-torque effect on our measurements, we also measured the field dependence of capacitance with *no field gradient* at each temperature.

Figure 1(a) shows $M(H)$ curves in the mixed state of the single crystal of UBe$_{13}$ for $H \| \langle 001 \rangle$ at 0.14 K. A clear irreversibility observed in low-field region below $\sim$20 kOe is due to vortex flux pinning in a type-II superconductor. We define H_{irr} as a field where the irreversibility vanishes completely. Just below H_{irr}, a peak effect is observed. Here, we note that H_{c2} of UBe$_{13}$ is almost the same as H_{irr} ($\lesssim H_{c2}$) [23].

In Fig. 1(a), we can see that the raw $M(H)$ curves bend around 20–30 kOe, $d^2M(H)/dH^2 > 0$, for both increasing [$M_{\mathrm{inc}}(H)$] and decreasing [$M_{\mathrm{dec}}(H)$] processes. The present results indicate that the origin of the anomaly in pure UBe$_{13}$ is *magnetic*.

The irreversibility of $M(H)$ curves becomes entirely small in high-field region above $\sim$30 kOe (Fig. 1) [24],

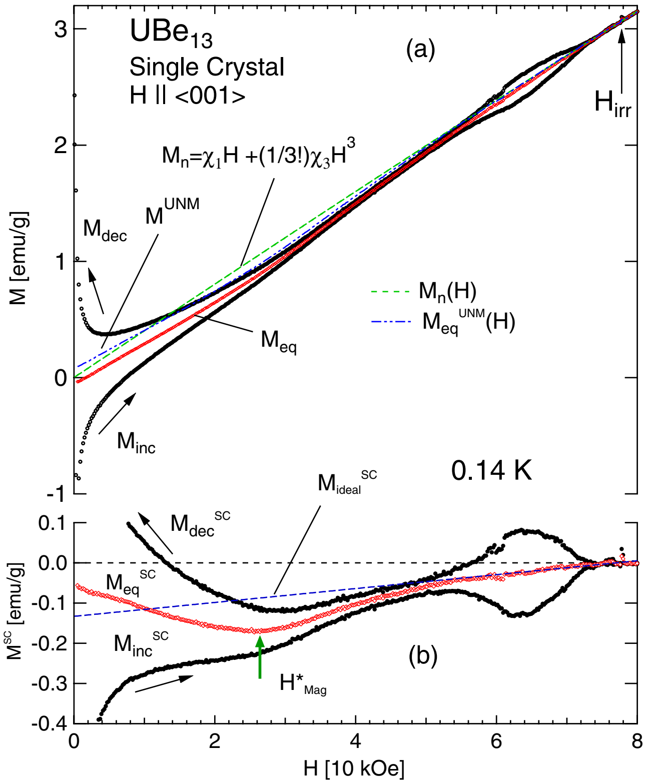

FIG. 1 (color online). (a) dc magnetization curves, $M_{\mathrm{inc}}(H)$, $M_{\mathrm{dec}}(H)$, and $M_{\mathrm{eq}}(H)$ of UBe_{13} for $H\|\langle 001\rangle$ at 0.14 K. Normal-state magnetization $M_n(H)$ is also plotted. $M_n^{\mathrm{UNM}}(H)$ is the unusual normal-state magnetization, assuming that the SC diamagnetic response is conventional (see text). (b) SC contribution to magnetization $M^{\mathrm{SC}}(H)$, and $M_{\mathrm{eq}}^{\mathrm{SC}}(H)$. $M_{\mathrm{ideal}}^{\mathrm{SC}}(H)$ is conventional SC diamagnetic response, which is estimated from SC condensation energy obtained by $C(T)$ (see text).

suggesting that the used single crystal is of high quality, because the flux-pinning centers are impurities or lattice defects in general. We also checked the large and sharp specific-heat jump at the SC transition for the same sample [25]. We then consider that the observed magnetic anomaly is an intrinsic phenomenon for a good-quality sample, which is related to the SC nature of UBe_{13}.

We approximately obtain thermal-equilibrium magnetization $M_{\mathrm{eq}}(H)$ by averaging increasing and decreasing processes: $M_{\mathrm{eq}} = (M_{\mathrm{inc}} + M_{\mathrm{dec}})/2$, as plotted in Fig. 1(a). The anomaly around ~26 kOe is observed both in $M_{\mathrm{inc}}(H)$ and $M_{\mathrm{dec}}(H)$, then we can bring out the anomaly also in $M_{\mathrm{eq}}(H)$. This $M_{\mathrm{eq}}(H)$ consists of normal-state magnetization $M_n(H)$ of vortices and SC diamagnetic response (SCDR) $M_{\mathrm{eq}}^{\mathrm{SC}}(H)$: $M_{\mathrm{eq}}(H) = M_n(H) + M_{\mathrm{eq}}^{\mathrm{SC}}(H)$. Therefore, it is natural to consider that the magnetic anomaly is originated from a peculiarity of $M_{\mathrm{eq}}^{\mathrm{SC}}(H)$ or $M_n(H)$. First, we report results of analysis, assumed that H_{Mag}^* anomaly is originated from an unusual SCDR. Although $M_n(H)$ above H_{c2} is almost linear, there is a very small contribution of nonlinear susceptibility in $M_n(H)$. We therefore fit $M_n(H)$ above H_{c2} by $M_n(H) = \chi_1 H + (1/3!)\chi_3 H^3$, and assume that $M_n(H)$ in the SC mixed state is also described by the same expression. By subtracting the contribution of $M_n(H)$, one can obtain $M_{\mathrm{eq}}^{\mathrm{SC}}(H)$.

Figure 1(b) shows SC contribution $M^{\mathrm{SC}}(H)$ and $M_{\mathrm{eq}}^{\mathrm{SC}}(H)$ of UBe_{13} at 0.14 K for $H\|\langle 001\rangle$. A broad minimum in $M_{\mathrm{eq}}^{\mathrm{SC}}(H)$ can be seen at around 26 kOe. We define H_{Mag}^* as a magnetic field where $M_{\mathrm{eq}}^{\mathrm{SC}}(H)$ shows the minimum; i.e., $|M_{\mathrm{eq}}^{\mathrm{SC}}(H)|$ reaches a maximum, which we denoted by upper arrow at ~26 kOe for 0.14 K in Fig. 1(b). As for $M_n^{\mathrm{UNM}}(H)$, and $M_{\mathrm{ideal}}^{\mathrm{SC}}(H)$, we will describe them in the following discussion.

Magnetic anomaly in the mixed state has been reported as anomalous magnetic-torque (AMT) effect from high-resolution torque measurements for a similar-quality sample by Schmiedeshoff *et al.* [26]. They observed the AMT effect also in the normal state at low-T and in high-H (~ 40–50 kOe) region. In our no-field-gradient measurements, we could not clearly observe it within the experimental error. Besides, since we observed the magnetic anomaly only in the SC state, its origin is different from the AMT effect [26]; while the AMT effect is related to a magnetic anisotropy, H_{Mag}^* is probably related to a change in terms of the absolute value of $M(H)$.

Figure 2 shows $M^{\mathrm{SC}}(H)$ and $M_{\mathrm{eq}}^{\mathrm{SC}}(H)$ of UBe_{13} for $H\|\langle 001\rangle$ at 0.18, 0.24, 0.37, and 0.50 K obtained in the same way as described above. We can see the anomaly around H_{Mag}^* denoted by upper arrows at each temperature, and it becomes distinct with cooling.

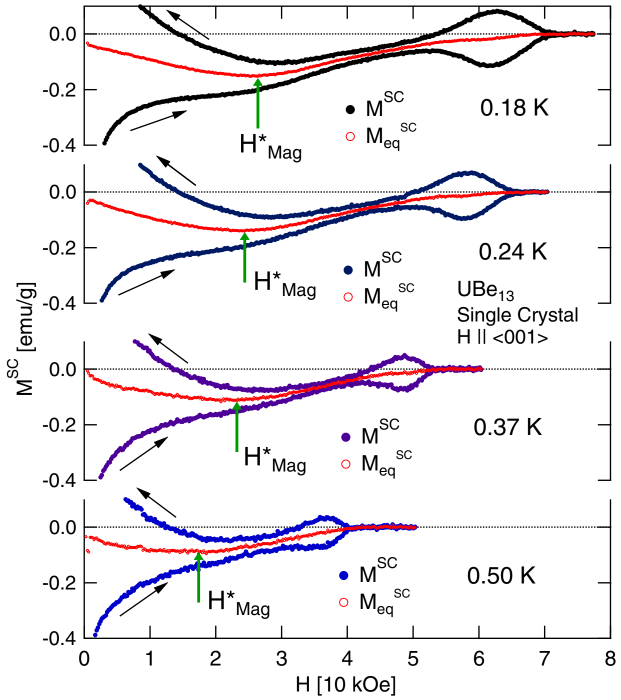

FIG. 2 (color online). SC diamagnetic responses, $M^{\mathrm{SC}}(H)$, and $M_{\mathrm{eq}}^{\mathrm{SC}}(H)$ of UBe_{13} for $H\|\langle 001\rangle$ at 0.18, 0.24, 0.37, and 0.50 K.

Figure 3 shows H-T SC phase diagram of UBe$_{13}$ with an image plot of $M_{\rm eq}^{\rm SC}(H)$ for $H||\langle 001\rangle$. $H_{\rm Mag}^{*}$ and $H_{\rm irr}$ ($\lesssim H_{c2}$) are also plotted. As seen in Fig. 3, the temperature dependence of $H_{\rm Mag}^{*}$ is similar to that of B^{*} [20,27], suggesting that the origins of these anomalies are the same. The light color in the image plot indicates a H-T region where $|M_{\rm eq}^{\rm SC}(H)|$ enhances. However, this phenomenon is extremely strange, because generally SC state excludes magnetic flux as well known, and SCDR decreases with increasing field in the SC mixed state. Then it is natural to consider that the unusual SCDR is reflecting a peculiarity of SC diamagnetic current or a presence of some orbital current below $H_{\rm Mag}^{*}$ by undefined reason, and then the SCDR below $H_{\rm Mag}^{*}$ becomes small *in appearance*. For example, such an effect might be caused by paramagnetic Meissner effect[28] or some strong flux-trapping effect below $H_{\rm Mag}^{*}$. In order to clarify this, a precise study of its sample-shape dependence is needed, because it has been reported that the paramagnetic Meissner effect occurs in very thick SC samples [28].

Another possibility is a presence of a magnetic-field-induced SC state in UBe$_{13}$, which is quite unlike BCS type-II superconductors; this might be caused by an increase of the superfluid density at around $H_{\rm Mag}^{*}$, or/and an increase of volume fraction of the SC state by some mechanism. The later case indicates that there is a non-SC part in the sample even below T_c. The volume fraction of the SC state would be dependent on the sample quality, then it will be needed to study the sample dependence on the behavior of $H_{\rm Mag}^{*}$ anomaly.

Is the estimation of $M_n(H)$ in the SC mixed state appropriate for UBe$_{13}$? In order to verify this, we examine whether SC condensation energy (SCCE) deduced from $M_{\rm eq}^{\rm SC}$ is quantitative or not. For this, it is useful to compare with a result obtained from specific-heat $C(T)$. Here, we define $H_c^{\rm Mag}$ and $H_c^{\rm SH}$ as thermodynamic critical fields obtained from $M_{\rm eq}^{\rm SC}(H)$ and $C(T)$, respectively. We obtain $H_c^{\rm Mag}$ by integrating $M_{\rm eq}^{\rm SC}(H): \frac{(H_c^{\rm Mag})^2}{8\pi} = -\int_0^{H_{c2}} M_{\rm eq}^{\rm SC}(H)dH$. On the other hand, we obtain SCCE also from $C(T): H_c^{\rm SH}(T)^2/8\pi = \int_T^{T_c} dT \int_T^{T_c} [C_{\rm SC}(T') - C_n(T')]/T' dT'$, where $C_{\rm SC}(T)$ and $C_n(T)$ are specific-heat in SC and normal states, respectively. Here, we assumed an entropy balance between the SC and the normal states according to Ref. [29].

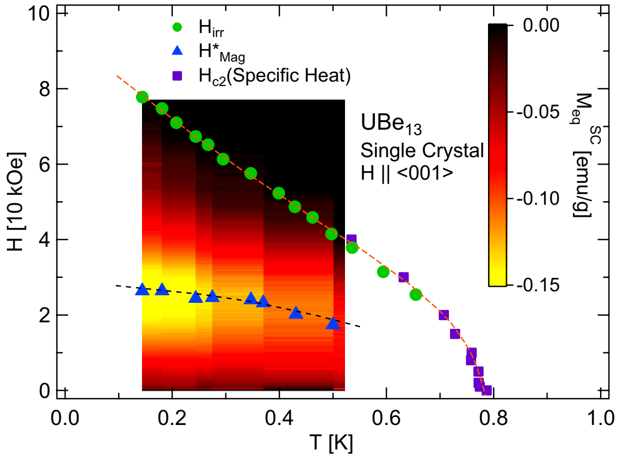

FIG. 3 (color online). SC phase diagram of UBe$_{13}$, including the results of $H_{\rm irr}(\lesssim H_{c2})$ and $H_{\rm Mag}^{*}$, with an image plot of $M_{\rm eq}^{\rm SC}(H)$ for $H||\langle 001\rangle$. Our previous specific-heat $C(T)$ results for $H||\langle 001\rangle$ on the same sample [the peak-top temperatures of $C(T)$ jump at SC transition] [25] are also plotted. Dashed lines are guide to the eye. The light color in the phase diagram indicates a H-T region where $|M_{\rm eq}^{\rm SC}(H)|$ becomes large.

Figure 4 shows the results of SCCE and $H_c^{\rm Mag}(T)$ obtained from $M_{\rm eq}^{\rm SC}$ curves for $H||\langle 001\rangle$, together with results for $H||\langle 110\rangle$ [30]. Dashed line indicates $H_c^{\rm SH}(T)$ obtained from $C(T)$ data in Ref. [25]. As seen in Fig. 4, there is no significant difference in SCCE and $H_c^{\rm Mag}(T)$ between for $H||\langle 001\rangle$ and for $H||\langle 110\rangle$. Since SCCE is a scalar quantity, SCCE should be isotropic. Namely, this isotropic behavior on $H_c^{\rm Mag}(T)$ is considered to be valid. Furthermore, $H_c^{\rm Mag}(T)$ roughly agrees with $H_c^{\rm SH}(T)$, suggesting that we cannot rule out the possibility of magnetic-field induced superconductivity.

We now discuss the origin of $H_{\rm Mag}^{*}$ anomaly, assuming that it is caused by an unusual normal-state magnetization (UNM) in the SC mixed state. We shall obtain the UNM in the SC mixed state by using the value of $H_c^{\rm SH}$ obtained from $C(T)$, assuming that the SCDR is *conventional* as in a BCS superconductor. We define $M_{\rm ideal}^{\rm SC}(H)$ as the conventional SCDR. Since we do not know a rigorous function of $M_{\rm ideal}^{\rm SC}(H)$, we roughly estimate $M_{\rm ideal}^{\rm SC}(H)$ by a straight linear magnetization $a + bH$ as shown in Fig. 1(b), so that the SC condensation energy becomes equal to that obtained from $C(T)$. Here, $a(<0)$, and $b(>0)$ are

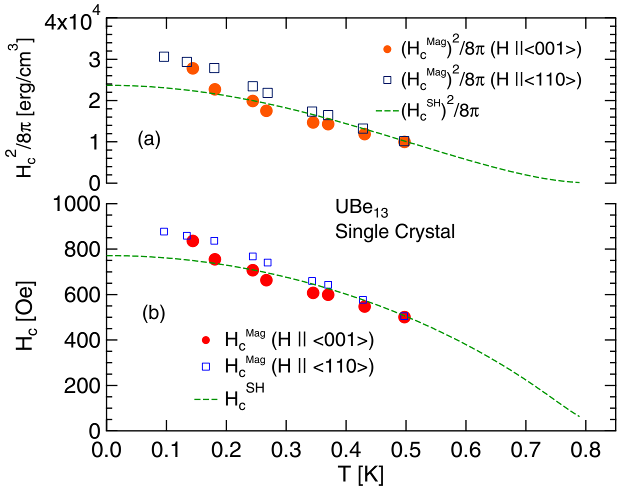

FIG. 4 (color online). (a) SC condensation energy, and (b) thermodynamic critical field of UBe$_{13}$ for $H||\langle 001\rangle$ (and $H||\langle 110\rangle$ [30]) obtained from $M_{\rm eq}^{\rm SC}(H)$ and $C(T)$ data [25].

constants, and are determined by an equation as below: $(H_c^{\mathrm{SH}})^2/8\pi \equiv -\int_0^{H_{c2}} M_{\mathrm{ideal}}^{\mathrm{SC}}(H)dH = -\int_0^{H_{c2}}(a+bH)dH = -[a+(b/2)H_{c2}]H_{c2}$. Besides, we do not consider the lower-critical field H_{c1}, because H_{c1} of UBe_{13} has been reported to be very small, $H_{c1}(T \sim 0) \sim 40$–50 Oe [31]. We may approximate $M_{\mathrm{ideal}}^{\mathrm{SC}}(H)$ in this way, because a type-II superconductor with a large Maki parameter κ_2 should exhibit an almost linear SCDR in sufficiently large magnetic field ($H_{c1} \ll H$), if the SCDR is conventional [32]; we have already known that UBe_{13} has a large value of the $\kappa_2 \sim 50$–100 as reported in our previous work [25]. Next, we conversely obtain the UNM, $M_n^{\mathrm{UNM}}(H)$ from $M_{\mathrm{ideal}}^{\mathrm{SC}}(H)$ and experimentally obtained $M_{\mathrm{eq}}(H)$; from $M_{\mathrm{eq}}(H) = M_n^{\mathrm{UNM}}(H) + M_{\mathrm{ideal}}^{\mathrm{SC}}(H)$, we obtain $M_n^{\mathrm{UNM}}(H) = M_{\mathrm{eq}}(H) - M_{\mathrm{ideal}}^{\mathrm{SC}}(H)$ [Fig. 1(a)].

As seen in Fig. 1(a), $M_n^{\mathrm{UNM}}(H)$ is not zero even in the vicinity of zero field. This implies a presence of ferromagnetic (FM) contribution, $M_0 \sim 0.1\ \mathrm{emu/g} \sim 0.4\ \mathrm{emu/cm^3} \sim 3 \times 10\ \mathrm{emu/mol}$ [33]. The FM moment per f.u. of UBe_{13} is converted as $m_0^{\mathrm{U}} \sim (3 \times 10\ \mathrm{emu/mol})/(N_{\mathrm{A}}\ \mathrm{mol}^{-1} \times \mu_{\mathrm{B}}\ \mathrm{erg/Oe}) \sim 5 \times 10^{-3}\ \mu_{\mathrm{B}}/\mathrm{U}$, where the μ_{B} and N_{A}, are the Bohr magneton and the Avogadro number, respectively. One of possible explanations of our results, regarding the assumption of UNM, is a presence of a weak FM moment of order $10^{-3}\ \mu_{\mathrm{B}}/\mathrm{U}$ below H_{Mag}^*, in the SC state of UBe_{13}.

Although a possibility of AF SDW has been discussed as an origin of the weak magnetism in $\mathrm{U}_{1\text{-}x}\mathrm{Th}_x\mathrm{Be}_{13}$ [10,17], our simple analysis indicates that the magnetic correlation below H_{Mag}^* is FM rather than AF. Considering the systematic studies of $\alpha(T)$ on pure and Th-doped UBe_{13} in Ref. [20], H_{Mag}^* anomaly observed in dc magnetization curves, may be originated from a short-range FM ordering with the weak magnetic moment. In fact, a FM correlation in UBe_{13} has been proposed from a scaling analysis on magnetoresistance measurements in its normal state by Andraka *et al.* [34]. If the FM correlation in UBe_{13} is reinforced only in its SC sate by some undefined reason, the FM instability might be deeply involved to the USC in UBe_{13}. The presence of such a FM correlation between f electrons in UBe_{13} is more natural than that of an AF correlation to form a parallel-spin Cooper pairing, which is suggested from the $C(T)$ results [11,25,29]. It is intriguing to investigate the FM fluctuation in UBe_{13} by another experiment, focusing on the weak magnetism below H_{Mag}^*.

At present, we cannot rule out a possibility of an uncompensated small moment due to geometry and size effects on an AF (short-range) ordering in vortices. Besides, we cannot completely exclude an extrinsic origin for its small magnitude. Then, it is worth considering possibilities of nonferromagnetic origin for this anomaly. For example, a short-range ordering of multipole degrees of freedom might yield such a small field-induced moment in addition to the H_{Mag}^* anomaly. To confirm this scenario, it is crucial to clarify the crystalline-electric-field ground state in UBe_{13}. Alternatively, a change of Fermi surface (density of states) around H_{Mag}^* might cause an increase of the susceptibility of normal state, as $d^2M(H)/dH^2 > 0$. In any case, we stress that the variation of magnetization, $d^2M(H)/dH^2 > 0$ around H_{Mag}^*, itself is an intrinsic experimental fact, no matter what the origin of this anomaly is. In order to clarify its origin, further studies such as microscopic measurements and its sample-dependence study will be needed.

In conclusion, we have performed precise low-T dc magnetization measurements on a single crystal of UBe_{13}, and observed the unusual magnetic anomaly at H_{Mag}^* in the SC mixed state. We suggest that magnetic field H_{Mag}^* is an energy scale which characterizes the unusual magnetic properties on the SC state of UBe_{13}.

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