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Quasiparticle excitations and evidence for superconducting double transitions in monocrystalline $U_{0.97}Th_{0.03}Be_{13}$

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Superconducting (SC) gap symmetry and magnetic response of cubic $U_{0.97}Th_{0.03}Be_{13}$ are studied by means of high-precision heat-capacity and dc magnetization measurements using a single crystal, in order to address the long-standing question of its second phase transition at T_{c2} in the SC state below T_{c1}. The absence (presence) of an anomaly at T_{c2} in the field-cooling (zero-field-cooling) magnetization indicates that this transition is between two different SC states. There is a qualitative difference in the field variation of the transition temperatures; $T_{c2}(H)$ is isotropic, whereas $T_{c1}(H)$ exhibits a weak anisotropy between the [001] and [111] directions. In the low-temperature phase below $T_{c2}(H)$, the angle-resolved heat capacity $C(T,H,\phi)$ reveals that the gap is fully opened over the Fermi surface, narrowing down the possible gap symmetry.

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The nature of superconductivity in heavy-fermion compounds is of primary importance because an unconventional pairing mechanism is generally expected to occur due to strong electron correlation between heavy quasiparticles. The discovery of heavy-fermion superconductivity in UBe_{13} [1] triggered exploration of an unconventional pairing mechanism in $5f$ actinide compounds, and subsequently two uranium compounds, UPt_3 [2] and URu_2Si_2 [3,4], were found to show superconductivity. These U-based heavy-fermion superconductors have attracted considerable interest because of their unusual superconducting (SC) and normal-state properties. Among these, superconductivity in UBe_{13} is highly enigmatic; it emerges from a strongly non-Fermi-liquid state with a large resistivity ($\rho \sim 150\ \mu\Omega\,\text{cm}$). Also unusual is the temperature variation of the upper critical field H_{c2}: an enormous initial slope $-(dH_{c2}/dT)_{T_c} \sim 42$ T/K and an apparent absence of a Pauli paramagnetic limiting at low temperatures [5]. Extensive studies have been made to elucidate the SC gap symmetry [6,7], with an expectation of an odd-parity pairing in this compound [8–11]. Recently, it has been found quite unexpectedly that nodal quasiparticle excitations in UBe_{13} are absent as revealed by low-T angle-resolved heat-capacity measurements for a single crystalline sample [12].

A long-standing mystery regarding UBe_{13} is the occurrence of a second phase transition in the SC state when a small amount of Th is substituted for U [Fig. 1(a)] [13,14]. It has been reported that there exist four phases (A, B, C, and D) in its SC state, according to the previous μSR [15] and thermal-expansion [16] experiments using polycrystalline samples. The SC transition temperature T_c is nonmonotonic as a function of the Th concentration x in $U_{1-x}Th_xBe_{13}$, and exhibits a sharp minimum near $x = 0.02$. Further doping of Th results in an increase of the bulk SC transition temperature (T_{c1}), reaching a local maximum at $x \sim 0.03$ [13]. Below T_{c1}, another phase transition accompanied by a large heat-capacity jump occurs at T_{c2} in a narrow range of $0.019 < x < 0.045$ [14,15]. Interestingly, only for this x region, weak magnetic correlations have been observed in zero-field μSR measurements [15]. The previous thermal-expansion study [16] claimed that the low-temperature ("T_L") anomaly appearing below T_c for $0 \leqslant x < 0.02$, which can be connected to the "B^* anomaly" observed in pure UBe_{13} [16–18], is a precursor of the transition at T_{c2}. Up to the present, the true nature of the transition at T_{c2} remains controversial [19,20].

Two different scenarios have been discussed so far on the T_{c2} transition: (i) an additional SC transition that breaks time-reversal symmetry [21], and (ii) the occurrence of an antiferromagnetic ordering that coexists with the SC state [22,23]. Indeed, although it has been reported that the NMR spin-relaxation rate [6], heat capacity [24], and muon Knight shift [25] show unusual temperature dependence in the SC state, little is known concerning the gap structure in $U_{1-x}Th_xBe_{13}$ due to the lack of information about the *anisotropy* of its quasiparticle excitations in magnetic fields.

In order to resolve the controversy regarding the second transition at T_{c2}, and to uncover its gap symmetry, in this Rapid Communication we report the results of high-precision heat-capacity and dc magnetization measurements on $U_{0.97}Th_{0.03}Be_{13}$. Single-crystalline $U_{0.97}Th_{0.03}Be_{13}$ samples were obtained using a tetra-arc furnace; the ingot was remelted several times and then quenched. By this procedure, we have succeeded in obtaining small monocrystalline samples with no additional heat treatment as confirmed by sharp x-ray Laue spots in Fig. 1(b). Heat capacity (C) was measured at low temperatures down to 60 mK by means of a standard quasiadiabatic heat-pulse method in a ^{3}He-^{4}He dilution refrigerator. Field-orientation dependences $C(H,\phi)$ were obtained under rotating magnetic fields in the $(1\bar{1}0)$ crystal plane that

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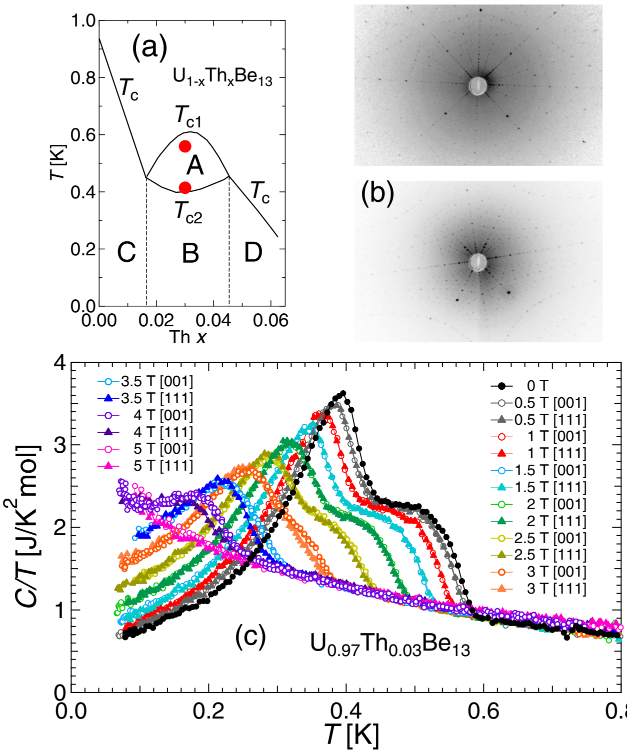

FIG. 1. (a) T-x phase diagram of $U_{1-x}Th_xBe_{13}$ [15,16], where the solid lines are based on Ref. [16]. There are four phases (A, B, C, and D) in its SC state, according to the previous μSR [15] and thermal-expansion [16] studies. The red circles indicate the transition temperatures at zero field for the sample used in the present experiment ($x = 0.03$). Here T_{c1} and T_{c2} are determined from $C(T)$ by considering the entropy conservation at each transition. (b) Laue x-ray photographs for the cubic fourfold (100) (upper panel) and twofold $(1\bar{1}0)$ (lower panel) planes. (c) $C(T)/T$ of $U_{0.97}Th_{0.03}Be_{13}$ at zero and in magnetic fields up to 5 T, measured every 0.5 T step for two directions $H \parallel [001]$ (circles) and $H \parallel [111]$ (triangles).

includes the [001], [111], and [110] axes, using a 5 T × 3 T vector magnet. We define the angle ϕ measured from the [001] direction. dc magnetization measurements were performed along the $[1\bar{1}0]$ axis down to $T \sim 0.28$ K for the same single crystal using a capacitive Faraday magnetometer [26] installed in a ^{3}He refrigerator. A magnetic-field gradient of 9 T/m was applied to the sample, independently of the central field at the sample position.

Figure 1(c) shows $C(T)/T$ curves measured at zero and various fields up to 5 T applied along the [001] and [111] axes. At zero field, two prominent jumps occur at $T_{c1} \sim 0.56$ K and $T_{c2} \sim 0.41$ K, where the transition temperatures [red circles in Fig. 1(a)] are determined by transforming the broadened transitions into idealized sharp ones by an equal-areas construction. The results are in agreement with the previous reports [13,14]. With increasing field, both transitions shift to lower temperature, getting closer to each other [27,28]. Above 3.5 T, the two transitions become very close to each other and are difficult to resolve separately. There is a notable feature in the anisotropy of $C(T)$ in magnetic fields. At low fields below ~1.75 T, the shifts of the two transition temperatures are almost isotropic. At higher fields above 2.5 T, however,

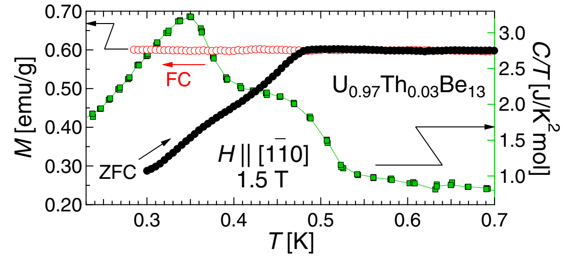

FIG. 2. Temperature dependence of the dc magnetization $M(T)$ measured at 1.5 T for $H||[1\bar{1}0]$. The data of $C(T)/T$ measured in the same magnetic field are also plotted for comparison.

the $T_{c1}(H)$ becomes slightly anisotropic, $T_{c1}(H||[001]) > T_{c1}(H||[111])$, while $T_{c2}(H)$ remains isotropic. In general, an anisotropy of $T_c(H)$ and H_{c2} results from those of SC gap function and/or Fermi velocity. If the double transitions come from two inhomogeneous SC states with the same gap symmetry, they should show the same anisotropic (or isotropic) field response. Our experimental results exclude such an extrinsic possibility. Thus the difference between field anisotropy in $T_{c1}(H)$ and $T_{c2}(H)$ is an essential effect which strongly suggests that the order parameters of these two phases have qualitatively different field-orientation dependences.

A key question, then, is whether the second transition at T_{c2} is a SC transition into a different gap symmetry. To address this question, we performed precise dc magnetization $[M(T)]$ measurements across the double transitions. Figure 2 shows the temperature dependence of $M(T)$ measured at 1.5 T together with the $C(T)/T$ data for the same field on the same sample. FC and ZFC denote the data taken in the field-cooling and zero-field-cooling protocols, respectively. The FC-ZFC branching occurs below ~0.5 K close to T_{c1} at this field, indicating the appearance of bulk superconductivity. We find a small but distinct kink in the ZFC data near T_{c2}, while no such anomaly can be seen in the FC curve. This fact implies a substantial change in the vortex pinning strength at this temperature, consistent with the previous vortex creep measurements [29,30]. Regarding the possible origin of the enhanced flux pinning in the low-T phase, we find no signatures that can be ascribed to a magnetic transition in the FC curve near T_{c2}. Our magnetization data, therefore, strongly suggest that the transition at T_{c2} is of a kind such that the SC order parameter changes. Indeed, it has been argued that such an enhancement of the vortex pinning occurs in a SC state with broken time-reversal symmetry [30]. This conclusion is also consistent with the previous neutron scattering measurements [31] which show no evidence for magnetic ordering in $U_{0.965}Th_{0.035}Be_{13}$ down to 0.15 K.

Next we examine the magnetic-field dependence of the heat capacity and its anisotropy in more detail, whose behavior in low fields reflects quasiparticle excitations in the SC state and provides a hint for the gap symmetry [32–34]. Figure 3(a) shows $C(H)/T$ measured at T =0.12, 0.18, 0.24, 0.30, 0.36, and 0.40 K for the cubic [001] and [111] directions, and the inset shows the enlarged $C(H)/T$ plot obtained at 0.08 K. Note that $C(H)$ below 1 T is quite linear to H at the lowest temperature of 0.08 K. This behavior is in striking contrast with a convex upward H dependence expected for nodal

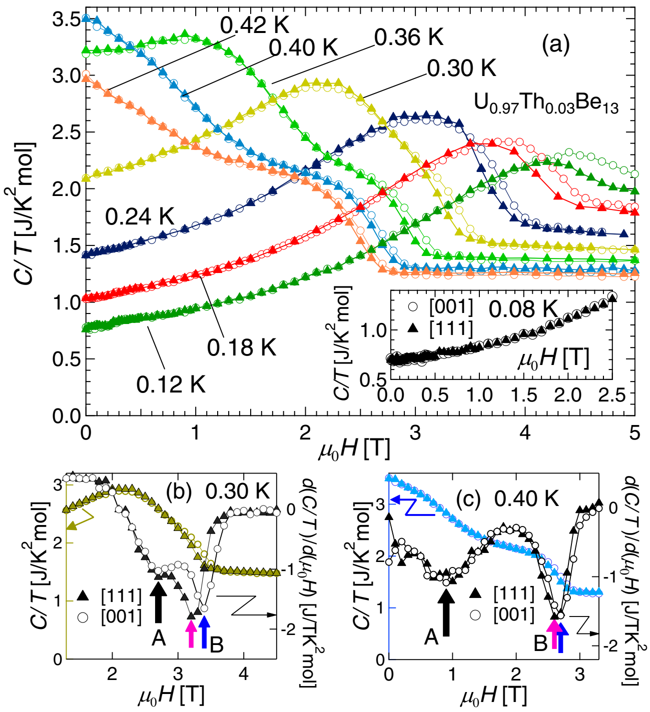

FIG. 3. (a) Magnetic-field dependence of $C(H)/T$ up to 5 T for $H||[001]$ (circles) and $H||[111]$ (triangles) measured at $T = 0.12$, 0.18, 0.24, 0.30, 0.36, 0.40, and 0.42 K. The inset shows the $C(H)/T$ in low magnetic fields measured at the base temperature of $T = 0.08$ K. $C(H)/T$ and its differential as a function of magnetic field around the double transitions at (b) 0.30 and (c) 0.40 K. The transition fields of the A and B phases, i.e., H_{c2}^{A} and H_{c2}^{B}, are determined as magnetic fields where the differential, $d[C(H)/T]/dH$, shows a local minimum.

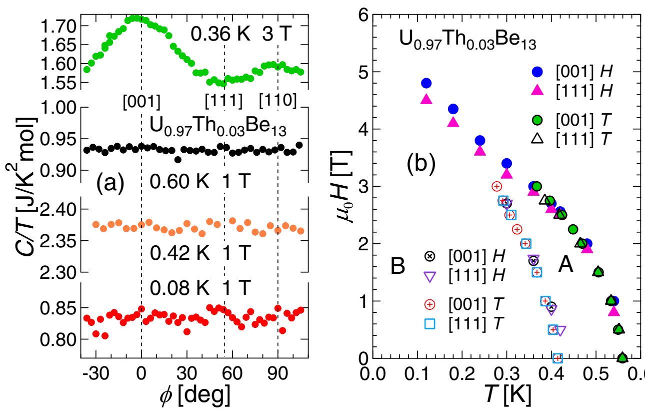

FIG. 4. (a) Angular dependence of $C(\phi)/T$, measured at $T = 0.08$ (B phase), 0.42 (A phase), and 0.60 K (normal state), in a magnetic field of 1 T. $C(\phi)/T$, measured at $T = 0.36$ K in 3 T (A phase), near H_{c2} is also plotted. (b) H-T phase diagram for the SC state of $U_{0.97}Th_{0.03}Be_{13}$ for [001] and [111], where T and H denote data obtained from temperature and field scans, respectively. Here, T_{c1} and T_{c2} were determined by considering entropy conservation at transitions in the $C(T)/T$ curves.

superconductors [37]. Moreover, there is *no* anisotropy in $C(H) \propto H$ between $H\,||\,[001]$ and [111] in low fields below $\sim$2 T. The absence of the anisotropy is further demonstrated by angle-resolved $C(\phi)/T$ in Fig. 4(a), obtained in a field of 1 T rotated in the $(1\bar{1}0)$ crystal plane at $T = 0.08$ and 0.42 K, together with the result measured in the normal state at 0.60 K. The absence of any angular dependence in $C(\phi)/T$ in a low-T low-H region again excludes the possibility of a nodal-gap structure in which a characteristic angular oscillation should be expected in $C(\phi)/T$ [33]. The present $C(H,\phi)$ data thus indicate that nodal quasiparticles are absent in $U_{0.97}Th_{0.03}Be_{13}$, similarly to the behaviors observed in pure UBe_{13} [12].

At higher fields, double-steplike anomalies are observed in $C(H)/T$ at 0.42, 0.40, and 0.36 K [Fig. 3(a)]. Here the double transitions can be clearly defined by the differential data, $d[C(H)/T]/dH$, as shown in Figs. 3(b) and 3(c). The lower-field step occurs when the boundary $T_{c2}(H)$ is crossed, while the higher-field one corresponds to the transition at $T_{c1}(H)$, i.e., the upper critical field $H_{c2}(T) \equiv H_{c2}^{A}$. Note that the position of the lower-field anomaly (H_{c2}^{B}) is fully *isotropic*, whereas the higher-field one (H_{c2}^{A}) shows an appreciable anisotropy, indicating that H_{c2} becomes anisotropic: $H_{c2}^{A}\,\|\,[001] > H_{c2}^{A}\,\|\,[111]$. The anisotropy of H_{c2}^{A} becomes larger at lower temperatures. With decreasing T, both of the transition fields shift to higher fields, getting close to each other, and are difficult to discriminate below $\sim$0.24 K [Fig. 3(a)]. These features of the transition fields are fully consistent with those observed for $T_{c1}(H)$ and $T_{c2}(H)$ shown in Fig. 1(c). Note that the isotropic behaviors in $C(H)/T$ as well as $T_{c2}(H)$ (Fig. 3) contrast starkly with the anisotropic behavior of B^{*} anomaly found in pure UBe_{13} [12], suggesting that these phenomena may result from different origins.

Figure 4(b) shows the H-T phase diagram of $U_{0.97}Th_{0.03}Be_{13}$ determined from the present $C(T,H)$ measurements, where the two SC phases are denoted as A and B phases. The overall features of the phase diagram are essentially the same as those obtained previously [27,28,38]. In Fig. 4(a), $C(\phi)/T$ data measured at $T = 0.36$ K in $\mu_0 H = 3$ T (A phase) rotated in the $(1\bar{1}0)$ are also shown; $C(\phi)/T$ shows a distinct angular oscillation with the maximum (minimum) along the [001] ([111]) direction, reflecting the anisotropy in H_{c2}^{A}.

The present experiment thus provides strong evidence that $U_{0.97}Th_{0.03}Be_{13}$ exhibits double SC transitions with two different SC order parameters. Let us discuss possible SC gap symmetries in this system. A key experimental fact is that the SC gap is fully open over the Fermi surface in both the B and C phases, as suggested by the present and previous [12] studies, respectively. This would imply either (i) the SC gap function itself to be nodeless, or (ii) the SC gap function to have nodes only in the directions in which the Fermi surface is missing. Regarding the latter, band calculations tell us that the Fermi surface is missing along the $\langle 111 \rangle$ direction, except for a tiny electron band [39,40]. Given the fact that spontaneous magnetism is observed from zero-field μSR only below T_{c2} [15], in addition, it would be natural to assume that the B phase is a time-reversal-symmetry-broken SC state. Under these constraints, two plausible scenarios can be proposed to explain the multiple SC phases in $U_{1-x}Th_xBe_{13}$. One is to employ a degenerate order parameter belonging to higher dimensional representations of the O_h symmetry (degenerate scenario). The other is to assume two order parameters belonging to different representations of the O_h group, nearly degenerate to each other (accidental scenario) [21].

Degenerate scenario. The group-theoretic classification of the gap functions under the cubic symmetry O_h has been given by several authors [21,41–43]. Among them, the two-dimensional odd-parity E_u state is a promising candidate for the order parameter which naturally explains the existing experimental data of both pure and Th-doped UBe_{13} [44]. The possibility of the odd-parity state has also been suggested from the μSR Knight shift experiments [25]. As for the odd-parity E_u state, we have two basis functions, $\boldsymbol{l}_1(k) = \sqrt{3}(\hat{\boldsymbol{x}}k_x - \hat{\boldsymbol{y}}k_y)$ and $\boldsymbol{l}_2(k) = 2\hat{\boldsymbol{z}}k_z - \hat{\boldsymbol{x}}k_x - \hat{\boldsymbol{y}}k_y$, and their combinated state, $\boldsymbol{d}(\boldsymbol{k}) = \boldsymbol{l}_1 + i\boldsymbol{l}_2 = \hat{\boldsymbol{x}}k_x + \epsilon\hat{\boldsymbol{y}}k_y + \epsilon^2\hat{\boldsymbol{z}}k_z$ with $\epsilon = e^{i(2\pi/3)}(\epsilon^3 = 1)$. The nonunitary state $\boldsymbol{d}(\boldsymbol{k}) = \boldsymbol{l}_1 + i\boldsymbol{l}_2$ has point nodes only along the $\langle 111\rangle$ direction, therefore, the nodal quasiparticle excitations can be missing considering the calculated Fermi surface [39,40]. The condition of the occurrence of each two-dimensional SC state can be examined using the Ginzburg-Landau free-energy density, $F = \alpha(T)(|\boldsymbol{l}_1|^2 + |\boldsymbol{l}_2|^2) + \beta_1(|\boldsymbol{l}_1|^2 + |\boldsymbol{l}_2|^2)^2 + \beta_2(\boldsymbol{l}_1\boldsymbol{l}_2^* + \boldsymbol{l}_1^*\boldsymbol{l}_2)^2$ with $\alpha(T) = \alpha_0(T_c - T)$, where $\beta_1 > 0$ is required for the stability. If $\beta_2 > 0$, the nonunitary state with the broken time-reversal symmetry becomes stable in lower T as a ground state (the B phase). With increasing temperature the degeneracy of the order parameters is lifted at T_{c2}, and one of them appears in the A phase ($T_{c2} < T < T_{c1}$). Logically, the other one appears in the C phase by changing dopant x. In pure UBe_{13} (the C phase), a nodeless gap function, i.e., $\boldsymbol{l}_2(k) = 2\hat{\boldsymbol{z}}k_z - \hat{\boldsymbol{x}}k_x - \hat{\boldsymbol{y}}k_y$, which is a unitary state, is likely, explaining the absence of nodal quasiparticle excitations [12] without invoking the Fermi-surface topology.

Accidental scenario. We briefly discuss the possibility of the accidental scenario, starting with the simplest and most symmetric A_{1u}, namely, $\boldsymbol{d}_{A1u}(\boldsymbol{k}) = \hat{\boldsymbol{x}}k_x + \hat{\boldsymbol{y}}k_y + \hat{\boldsymbol{z}}k_z$ with an isotropic full gap as the C phase for $x = 0$. From $x = 0.019$ to $x = 0.045$, we consider the combined state of one-dimensional representations, the above p wave A_{1u} and f wave A_{2u} with $\boldsymbol{d}_{A2u}(\boldsymbol{k}) = \hat{\boldsymbol{x}}k_x(k_y^2 - k_z^2) + \hat{\boldsymbol{y}}k_y(k_z^2 - k_x^2) + \hat{\boldsymbol{z}}k_z(k_x^2 - k_y^2)$. The combined state of A_{1u} and A_{2u}, namely, nonunitary $\boldsymbol{d}(\boldsymbol{k}) = \boldsymbol{d}_{A1u} + i\boldsymbol{d}_{A2u}$ is nodeless irrespective of the Fermi-surface topology, although $\boldsymbol{d}_{A2u}$ alone has point nodes along the $\langle 100\rangle$ and $\langle 111\rangle$ directions. Thus nodeless A_{1u} and the $A_{1u} + iA_{2u}$ states can explain the absence of nodal quasiparticles in pure and Th-doped UBe_{13}, respectively [45]. Similarly, the other order parameters belonging to different irreducible representations are possible, e.g., $A_{1u} + iE_u$; the determination of the two order parameters is not easy due to the arbitrariness of their combinations.

Finally, it is worth discussing the topology of the H-T phase diagram. In Fig. 4(b), it may appear that the lines of $T_{c1}(H)$ and $T_{c2}(H)$ merge into a single second-order transition line in a high-field region. Such case is, however, not allowed in the thermodynamic argument of the multicritical point [46,47]. Instead, a crossing of the two second-order transition lines at a tetracritical point is possible [46]. This argument imposes the existence of another second-order transition below H_{c2} for $T \lesssim 0.25$ K, but no evidence for such a transition line has been obtained so far in our measurements as well as in previous thermal-expansion studies [16]. It might be natural to consider an anticrossing of the two second-order transition lines [48]. The crossing of $T_{c1}(H)$ and $T_{c2}(H)$ in $U_{1-x}Th_xBe_{13}$ will be examined further in future studies.

To conclude, low-energy quasiparticle excitations and magnetic response of $U_{0.97}Th_{0.03}Be_{13}$ were studied by means of heat-capacity and dc magnetization measurements. The magnetization results evidence that the second transition at T_{c2} is between two different SC states. Strikingly, the present $C(T,H,\phi)$ data strongly suggest that the SC gap is fully open over the Fermi surface in $U_{0.97}Th_{0.03}Be_{13}$, excluding a number of gap functions possible in the cubic symmetry. Our new thermodynamic results entirely overturn a widely believed idea that nodal quasiparticle excitations occur in the odd-parity SC state with broken time-reversal symmetry. The absence (presence) of anisotropy for T_{c2} (T_{c1}) in fields clearly demonstrates that the gap symmetry in the B phase ($T < T_{c2}$) is distinguished from that of the A phase ($T_{c2} < T < T_{c1}$). Moreover, the isotropic behavior of the $T_{c2}(H)$ in $U_{1-x}Th_xBe_{13}$ contrasts starkly to the anisotropic field response of the B^* anomaly found in pure UBe_{13}. These findings lead to a new channel to deepen its true nature of the ground state of $U_{1-x}Th_xBe_{13}$, clarifying the origin of the unusual transition inside the SC phase.

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