Field-Orientation Dependence of Low-Energy Quasiparticle Excitations in the Heavy-Electron Superconductor UBe_{13}

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Low-energy quasiparticle excitations in the superconducting (SC) state of UBe_{13} were studied by means of specific-heat (C) measurements in a rotating field. Quite unexpectedly, the magnetic-field dependence of $C(H)$ is linear in H with no angular dependence at low fields in the SC state, implying that the gap is fully open over the Fermi surfaces, in stark contrast to previous expectations. In addition, a characteristic cubic anisotropy of $C(H)$ was observed above 2 T with a maximum (minimum) for $H \| [001]$ ([111]) within the $(1\bar{1}0)$ plane, in the normal as well as in the SC states. This oscillation possibly originates from the anisotropic response of the heavy quasiparticle bands, and might be a key to understand the unusual properties of UBe_{13}.

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After the development of the BCS theory of superconductivity, it was realized that Cooper instability could arise from nonphononic mechanisms [1–4]. The exploration for the unconventional pairing mechanisms was accelerated by the discoveries of exotic superconductivity (superfluidity) in ^{3}He [5–7], heavy-electron superconductors [8–11], and high-transition-temperature copper oxides [12,13] in the 1970 s–1980 s, all of which exhibit highly unusual ground-state properties.

As for f-electron compounds, heavy quasiparticles acquire huge effective masses 100–1000 times larger than ordinary conduction electrons, and hence are moving very slowly in the crystals. In this situation, the conventional Debye-phonon-mediated attraction is unlikely to overcome the Coulomb repulsion between electrons. It has therefore been widely accepted that f-electron Cooper pairs favor anisotropic spatial distributions (nodal-gap symmetry) to avoid the strong Coulomb repulsions. Determination of the gap symmetry is of primary importance because it can be directly related to the pairing interaction. However, it is by no means an easy task for heavy-electron compounds, and there are only a few examples whose superconducting (SC) gap structures are fully elucidated. Quite surprisingly, it has been clarified very recently that the SC gap in the oldest heavy-electron superconductor $CeCu_2Si_2$ is fully open over the Fermi surfaces [14], in strong contradiction to the previous expectations.

In this Letter, we focus on a cubic heavy-electron superconductor UBe_{13}. Despite extensive studies over 30 years, the nature of superconductivity in UBe_{13} is still elusive. The ^{9}Be-NMR-Knight shift has been reported to be invariant below the SC transition temperature $T_c \approx 0.86$ K [15,16], suggesting an odd-parity pairing. However, the μ^+SR-Knight-shift experiment indicates a significant decrease of the static susceptibility below T_c [17], conflicting with the NMR results. Regarding the gap structure, whereas the specific-heat $C(T)$ [18] and the magnetic penetration depth [19] experiments suggest the presence of point nodes, the NMR spin-relaxation rate [20] and the ultrasound attenuation [21] are rather indicative of line nodes. Although there is compelling evidence for unconventional pairing, the SC gap symmetry in UBe_{13} thus remains undetermined.

Another important issue with respect to the SC state in UBe_{13} is a feature observed in thermodynamic quantities such as $C(H)$ [22–24], dc magnetization $M(H)$ [25] as well as the thermal expansion [22] at fields below ~4 T, constituting a line of anomaly ("B^* anomaly") in the H-T phase diagram. Whereas the origin of this anomaly is still unresolved, it has been discussed as a precursor [23] of the second phase transition below T_c observed in $U_{1-x}Th_xBe_{13}$ $(0.019 < x < 0.045)$ [26].

The normal state of UBe_{13} is also highly unusual. It exhibits non-Fermi-liquid (NFL) behavior down to very close to T_c as revealed by electrical resistivity, specific heat [27,28], and magnetic susceptibility [29]. The origin of NFL behaviors in UBe_{13} remains unclear, and several possibilities have been discussed so far. These include the quadrupolar Kondo effect with a Γ_3-crystalline-electric-field ground state for the $5f^2$ (U^{4+}, $J = 4$) configuration [30], an antiferromagnetic quantum-critical point induced by a magnetic field [28,31], and a competition between Kondo-Yosida and Γ_1-crystalline-electric-field singlets for $5f^2$ configuration [32]. Since the SC state apparently emerges out of the NFL state, its understanding is crucial in elucidating the pairing mechanism in UBe_{13}.

In order to gain more insight into the SC gap symmetry as well as the normal state, we performed specific-heat measurements of UBe_{13} at low temperatures down to ~75 mK in magnetic fields up to 5 T. The single crystal of UBe_{13}, used in the present study was prepared by an Al-flux method [33]. This is the same crystal as was used in the previous dc magnetization study [25]. The specific heat C was measured by a standard quasiadiabatic heat-pulse method. Field-angular dependences $C(H, \phi)$ were measured with H rotating in the $(1\bar{1}0)$ crystal plane that includes three principal directions [001], [111], and [110]. The angle ϕ is measured from the [001] axis.

Figure 1(a) shows $C(T)/T$ curves measured in various magnetic fields up to 5 T. The zero-field data are also

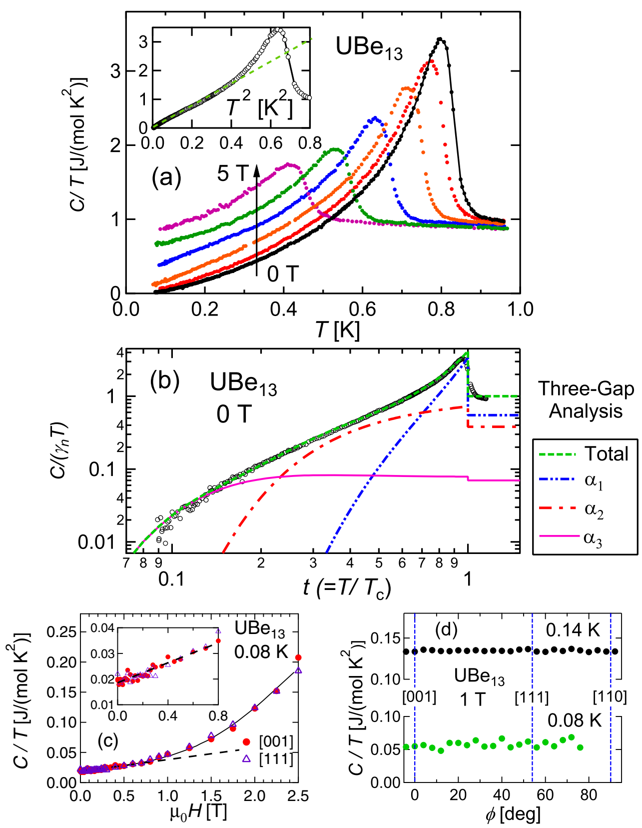

FIG. 1 (color online). (a) $C(T)/T$ of UBe_{13} at $\mu_0 H = 0$, 1, 2, 3, 4, and 5 T for $H \| [001]$. The inset shows the $C(T)/T$ vs T^2 plot for $H = 0$. (b) The result of the three-band full-gap analysis for $C(T)/T$ ($H = 0$) in log-log scale. The parameters are: $\alpha_1 = 1.9$, $\alpha_2 = 0.8$, $\alpha_3 = 0.3$, and $\gamma_1 : \gamma_2 : \gamma_3 = 55 : 38 : 7$. (c) $C(H)/T$ at $T = 0.08$ K for $H \| [001]$ (solid circles) and $H \| [111]$ (open triangles) as a function of H in the low-field region. The dashed line is a linear fit to the data below ~0.8 T for $H \| [001]$. The solid line is a guide to the eyes. (d) $C(\phi)/T$ in a field of 1 T rotated in the $(1\bar{1}0)$ plane, measured at 0.08 and 0.14 K.

plotted in Fig. 1(b) in log-log scale. There is no Schottky contribution from ^{9}Be nuclei, owing to their long nuclear spin-relaxation time of the order of 10^3 sec [20], much longer than our measuring time (10^2 sec) of the specific heat. The inset of Fig. 1(a) shows the $C(T)/T$ vs T^2 plot; $C(T)$ below ~0.6 K behaves like T^3 as previously reported [18]. Note that the residual density of states, $C(T)/T|_{T\to 0}$, is very small.

Magnetic-field dependence of the specific heat and its anisotropy in low fields reflect quasiparticle excitations within the SC gap [34–36]. In the case of line nodes, $C(H) \propto (H/H_{c2})^{1/2}$ is expected [34–36], whereas for point nodes, $C(H) \propto (H/H_{c2}) \ln(H/H_{c2})$ [37], or $C(H) \propto (H/H_{c2})^{0.64}$ [38]. In either case, the field dependence of C/T should exhibit a convex upward curvature at low fields. For a clean isotropic s-wave superconductor, on the other hand, $C(H)/T \propto H$ at low fields because low-energy quasiparticles are mainly confined in vortices whose density increases in proportion to H [39]. Figure 1(c) shows $C(H)/T$ of UBe_{13} below 2.5 T for $H \| [001]$ and $H \| [111]$ measured at 0.08 K. Surprisingly, the low-field $C(H)/T$ curve is rather *linear* in H, suggesting the absence of nodal quasiparticles. Note that there is *no* anisotropy in this H-linear behavior of $C(H)$ below ~1 T between [001] and [111] directions within an experimental accuracy [Fig. 1(c)]. The absence of the anisotropy is further confirmed by $C(\phi)/T$ obtained in a field of 1 T rotated in the $(1\bar{1}0)$ crystal plane at $T = 0.08$ and 0.14 K [Fig. 1(d)]; there is *no* significant angular variation in $C(\phi)/T$, implying that the $C(H)/T \propto H$ behavior holds for all directions. Indeed, the low-field $C(T, H)$ data do not exhibit a scaling behavior expected for a nodal superconductor [24].

We would like to emphasize that the linear slope of $C(H)/T$ in Fig. 1(c) is unusually small. In this regard, it has been argued that the line-nodal or point-nodal sublinear dependences in $C(H)/T$ described above would be smeared out at a finite temperature $T/T_c > \sqrt{H/H_{c2}}$ [40]. Even in such a case, however, the rate of increase of $C(H)/T$ should be greater than that governed by the localized-quasiparticle contribution from vortex cores approximated as $C(H)/T = (C_n/T)(H/H_{c2}^{orb}(0))$, where $\mu_0 H_{c2}^{orb}$ (= 25 T [41]) denotes the orbital-limiting field. Assuming the normal-state value C_n/T at $T \sim 0$ to be 1.1 J mol^{-1} K^{-2} by taking into account an entropy balance, we estimate the slope $(C_n/T)/(\mu_0 H_{c2}^{orb}(0))$ of the ordinary vortex core contribution to be 0.044 J mol^{-1} K^{-2} T^{-1}. The observed initial slope in Fig. 1(c) is 0.02 ± 0.006 J mol^{-1} K^{-2} T^{-1}, a factor of two smaller than this; apparently, there is a significant deficiency of quasiparticles. We will come back to this point later.

Figure 2(a) shows the field variation of $C(H)/T$ up to 5 T for $H \| [001]$ measured at $T = 0.08$, 0.14, 0.24, 0.40, and 0.95 K (closed symbols). We also plot the data for

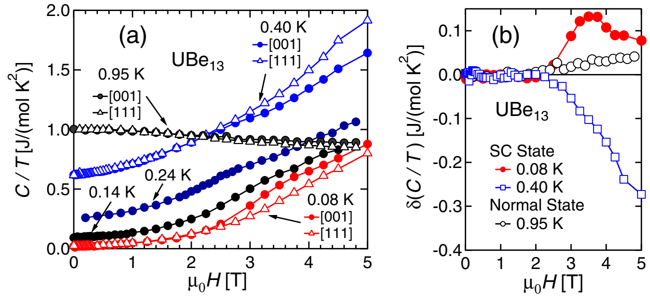

FIG. 2 (color online). (a) Magnetic-field dependence of $C(H)/T$ up to 5 T for $H\|[001]$ (solid circles) and $H\|[111]$ (open triangles) measured at $T = 0.08$, 0.14, 0.24, 0.40, and 0.95 K. (b) $\delta(C/T) \equiv (C_{[001]} - C_{[111]})/T$ as a function of H, obtained at $T = 0.08$ (solid circles), 0.40 (open squares), and 0.95 K (open circles).

$H\|[111]$ measured at $T = 0.08$, 0.40, and 0.95 K (open symbols). $C(H)/T$ curve for $H\|[001]$ in the SC state at $T = 0.08$ K exhibits a strong upturn above ~2 T. This behavior is quite reminiscent of a superconductor with a strong Pauli paramagnetic effect, as observed for $CeCu_2Si_2$ [14]. Note that a weak hump appears in $C(H)/T$ above ~3 T, whose position moves to lower fields with increasing T. This hump has been known as the "B^* anomaly" [22–25]. We observe that this anomaly in $C(H)$ is clearer for $H\|[001]$ than for $H\|[111]$. Accordingly, a substantial anisotropy develops in $C(H)$ above this field. An anisotropy has also been observed by dc magnetization curves above B^* [42,43]. In order to display the evolution of the anisotropy in $C(H)/T$, we plot in Fig. 2(b) the difference $\delta(C/T) \equiv (C_{[001]} - C_{[111]})/T$ at $T = 0.08$, 0.40, and 0.95 K, where $C_{[001]}$ ($C_{[111]}$) denotes the specific heat for $H\|[001]$ ($H\|[111]$). For $T = 0.08$ K, $\delta(C/T)$ shows a distinct positive peak around 3.5 T due to the B^* anomaly. At 0.40 K, $\delta(C/T)$ changes the sign and shows a monotonic decrease with increasing field above 2 T, reflecting the anisotropy of H_{c2} [43,44]. In the normal state at 0.95 K, $\delta(C/T)$ turns positive again and increases monotonically with increasing field above ~2 T. It is also remarkable that the normal-state C/T is substantially suppressed in a field of 5 T for both directions.

In Fig. 3, we show the field-angle dependences of C/T in the $(1\bar{1}0)$ plane measured in a magnetic field of (a) 2 T and (b) 4 T. For $\mu_0 H = 2$ T, an appreciable angular variation can be seen at $T = 0.28$ and 0.14 K, with a maximum (minimum) at [001] ([111]) and a local maximum at [110], i.e., $C_{[111]} < C_{[110]} < C_{[001]}$. Hereafter, we call this type of angular variation "type-I oscillation". At this field, the oscillation disappears for $T = 0.08$ K reflecting the absence of nodal quasiparticles. When the magnetic field is increased to 4 T, the type-I oscillation appears even at a low temperature of $T = 0.08$ K, with a huge relative amplitude of nearly 25%. Such a large anisotropy cannot be ascribed to nodal quasiparticles. With increasing T, the sign of the

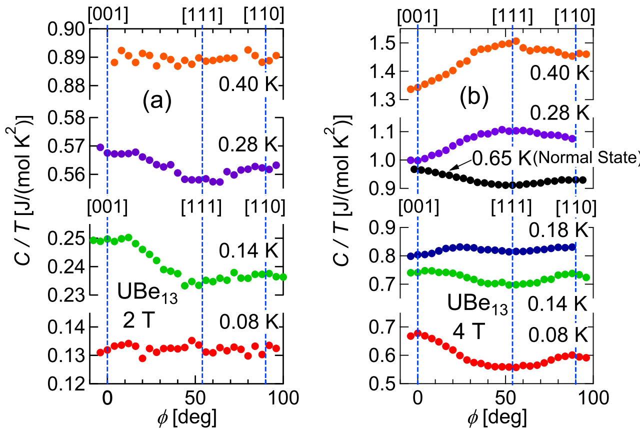

FIG. 3 (color online). Angular dependences of $C(H, \phi)/T$, measured in (a) $\mu_0 H = 2$ T and (b) 4 T at $T = 0.08$, 0.14, 0.28, and 0.40 K. Data for $T = 0.65$ K and $\mu_0 H = 4$ T in the normal state are also plotted.

oscillation changes for $T = 0.28$ and 0.40 K. We call this reversed angular variation the "type-II oscillation." The reversed oscillation at these higher temperatures is probably due to an anisotropy of H_{c2}. Note that the amplitude of both the type-I and type-II oscillations at 4 T is nearly 10-times larger than that at 2 T; the anisotropy strongly develops with increasing H as can be seen from Fig. 2(b). For $T = 0.14$ and 0.18 K, $C(H, \phi)/T$ exhibits a rather irregular angular variation because these are in the crossover region between the type-I and the type-II oscillations.

Figure 4 shows an enlarged plot for the angular dependence of $C(H, \phi)/T$ in the normal state at $T = 0.65$ K in a field of 4 T. Interestingly, the type-I oscillation is also observed in the specific heat of the normal state. This fact implies that the magnetic-field response of the heavy quasiparticles in UBe_{13} is anisotropic. Very interestingly, a similar type-I angular variation of $C(H, \phi)/T$ is observed deep in the SC state at 0.08 K, suggesting a common origin for the anisotropy. This type-I oscillation in the SC state is quite likely due to the quasiparticles in the vortex core, because of its similarity to the oscillation observed in the normal state. Having established that $C(H, \phi)/T$ in the SC state exhibits rather unusual field and angular variation above 3 T, we consider that it is not appropriate to discuss the nodal structure in this field region [24].

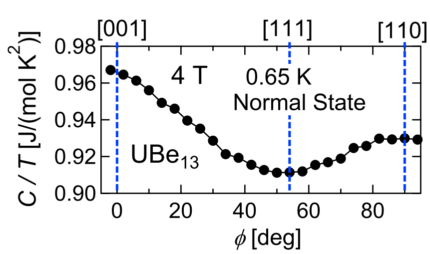

FIG. 4 (color online). Angular dependence of $C(H, \phi)/T$ in the normal state at $T = 0.65$ K with $\mu_0 H = 4$ T rotated in the $(1\bar{1}0)$ plane.

The most striking outcome of the present study is the apparent absence of nodal quasiparticles in UBe_{13}. The field and angular dependences of the specific heat in the SC state at low T and low H strongly suggest that the gap is fully open over the whole Fermi surface, although the temperature variation of the specific heat is quite similar in appearance to that of a point-node gap. A similar situation has been observed for the heavy-electron superconductor $CeCu_2Si_2$ [14], in which the T^2-like variation of $C(T)$ at zero field is successfully reproduced by a two-band full-gap model. It is, therefore, tempting to analyze the $C(T)$ of UBe_{13} by a multiband full-gap model, employing the α model for a strong-coupling superconductor [45]. In order to reproduce $C(T)$, we need to assume three bands with different amplitudes of isotropic gaps. Figure 1(b) shows the fitting result with the parameters $(\gamma_1 : \gamma_2 : \gamma_3 = 55:38:7)$, and $(\alpha_1 = 1.9, \alpha_2 = 0.8, \alpha_3 = 0.3)$, where $\alpha_i = \Delta_i/(1.76 k_B T_c)$, and Δ_i and γ_i denotes the SC gap and the electronic specific-heat coefficient of the ith band, respectively [46]. The simple model reproduces $C(T)$ of UBe_{13} remarkably well. Indeed, the calculated Fermi surfaces, consisting of two hole sheets and one electron sheet, support the occurrence of multiband superconductivity in UBe_{13} [47,48].

There are two plausible scenarios for the absence of nodal quasiparticle excitations in UBe_{13}: (i) the SC gap function itself is nodeless, or (ii) the SC gap function has nodes only in the directions in which the Fermi surfaces are missing. The case (i) includes fully symmetric A_{1g} representation. However, for a cubic point group there are several possibilities for both even and odd parity unconventional pairing states in which the gap functions are nodeless [49–51]. Regarding the case (ii), we would like to point out that, according to the band calculations, the Fermi surface is missing along the $\langle 111 \rangle$ directions, except for a tiny electron band [47,48]. Hence, the quasiparticle excitation can be full-gap-like even if *point nodes exist* for the $\langle 111 \rangle$ directions. Indeed, there are several odd-parity gap functions having only *point nodes* along the $\langle 111 \rangle$ directions [49–52]. In other words, any other symmetry having line nodes or point nodes along the $\langle 100 \rangle$ directions can be ruled out. In either case, therefore, the absence of nodal quasiparticles does not necessarily rule out the possibility of unconventional pairing in UBe_{13}.

It is remarkable that $C(H,\phi)/T$ in the normal state exhibits a characteristic angular oscillation (Fig. 4). In a cubic system, the Landau expansion of the free energy contains cubic invariants composed of magnetic-field components. Accordingly, the field and angular dependences of the electronic specific heat can be expressed in the form $C(T,H) \simeq C_0(T) + \alpha_1(T)H^2 + \alpha_2(T)H^4 + \beta(T)(H_x^4 + H_y^4 + H_z^4)$, where the last term represents the quartic invariant [53]. Note that the quadratic term is fully isotropic in cubic symmetry. Therefore, the anisotropy emerges at a relatively high field, and $\beta > 0$ ($\beta < 0$) causes the type-I (type-II) oscillation. In particular, $(C_{[001]} - C_{[110]}/C_{[110]} - C_{[111]}) \simeq 3$ should always hold, and is indeed almost satisfied for both the normal (Fig. 4) and the SC states with the type-I oscillation. We also obtain $\delta(C/T) = (C_{[001]} - C_{[111]})/T \simeq \frac{2}{3}\beta H^4$, which is roughly consistent with the $\delta(C/T)$ at 0.95 K [Fig. 2(b)].

It is most plausible that the anisotropic response of $C(H)/T$ in the normal state comes from a Zeeman effect in heavy-electron bands. The effective Fermi energy of UBe_{13}, E_F/k_B, is reported to be $\sim$8 K [18]. Therefore, a magnetic field of several tesla may result in a substantial Zeeman splitting of the heaviest band, leading to a reduction of the density of states at E_F. The data in Figs. 2(a) and 4 imply that this effect is anisotropic. Unlike the case of Ce-based (f^1) compounds in which a magnetic Kondo effect plays a key role, there is no widely accepted model for the formation of a heavy-electron state of U-based (f^2) compounds, in particular of UBe_{13} in which C/T exhibits non-Fermi-liquid behavior down to near T_c. In this regard, an intriguing candidate is a two-channel Kondo lattice model [55–57], in which odd frequency superconductivity has been predicted. In any case, the observed anisotropic field response (Fig. 4) would provide a clue to unveil the unusual normal state in UBe_{13}.

Using $E_F/k_B \sim 8$ K and $\Delta/k_B T_c \sim 3$ for the heaviest band, we estimate the lowest excitation level for the localized quasiparticle state in the vortex core to be $\Delta^2/k_B E_F \sim 1$ K. Thus, the quantum limit, in which a thermal broadening is narrower than the discrete bound state level [58,59], may be realized at the base temperature (0.08 K) of the present experiment. In this situation, the core quasiparticles are nearly empty at least for the heaviest band, and would not contribute to a H-linear increase of $C(H)/T$. This effect can explain the unusual smallness of the initial slope of $C(H)/T$ in Fig. 1(c), with the γ value of the heaviest band being roughly 1/2 of the total γ as given by the multiband analysis of $C(T)$ in Fig. 1(b). We consider that the core quasiparticles are gradually recovered above $\sim$3 T due to an overlapping of the discrete levels between neighboring vortices, and contribute to the observed anisotropy in $C(H,\phi)/T$ at 0.08 K.

Finally, we briefly discuss the possible origin of the B^* anomaly. In Ref. [23], B^* has been discussed in the context of a precursor of a second phase transition in $U_{1-x}Th_xBe_{13}$. Here we propose an alternative scenario based on a Pauli paramagnetic effect. The fact that $H_{c2}(0)$ of UBe_{13} is only $\sim$1/3 of $H_{c2}^{orb}(0)$ [41] indicates that a significant paramagnetic suppression exists. In a multiband superconductor, the paramagnetic effect can be strongly band dependent, and a minor gap may be suppressed at a field H^* much below $H_{c2}(0)$. In this situation, a small peak would appear in $C(H)$ at H^* [60], as observed in $CeCu_2Si_2$ [14]. This H^* might explain the B^* anomaly. It is also worthwhile to point out that the equilibrium magnetization of UBe_{13}

exhibits a minimum slightly below this field [25], possibly reflecting the Pauli paramagnetic effect [60].

In conclusion, we studied the SC symmetry and the low-energy quasiparticle excitations in UBe_{13} by means of specific-heat measurements on a high-quality single crystal. The isotropic H-linear dependence of $C(H)$ in low fields and low temperatures indicates the absence of nodal quasiparticle excitations in the SC gap. Whereas the present results do not exclude the possibility of unconventional pairing, the temperature dependence of the specific heat can be well explained by a multiband full-gap model. The present results, along with the recent finding of the full-gap state in $CeCu_2Si_2$ [14], urge reconsideration of pairing mechanisms in heavy-electron superconductors, for which it has been widely believed that the presence of gap nodes is necessary to avoid the strong electron-electron Coulomb repulsion. A characteristic cubic anisotropy of $C(H)$ is observed at high fields with H rotated in the $(1\bar{1}0)$ plane, not only in the SC state but also in the normal state. This might be a clue to understand the origin of NFL behavior as well as the enigmatic SC state in UBe_{13}.

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