PHYSICAL REVIEW B **92**, 241101(R) (2015)

Non-Fermi-liquid nature and exotic thermoelectric power in the heavy-fermion superconductor UBe_{13}

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(Received 23 March 2015; revised manuscript received 31 August 2015; published 2 December 2015)

We report quite exotic thermoelectric power S in UBe_{13}. At 0 T, the negative S/T continues to strongly enhance down to the superconducting transition temperature with no Fermi-liquid behavior. $|S/T|$ is dramatically suppressed and becomes rather modest with increasing field. We have also obtained precise field dependencies of (i) an anomaly in S due to an exotic Kondo effect and (ii) a field-induced anomaly in S/T associated with the anomalous upward $H_{c2}(T)$. In contrast to the field-sensitive transport property, the normal-state specific heat is magnetically robust, indicating that the largeness of the $5f$ density of states remains in high fields. This unusual behavior in UBe_{13} can be explained by a considerable change in the energy derivative of the conduction-electron lifetime $\tau_c(\epsilon)$ at the Fermi level under magnetic fields.

DOI: 10.1103/PhysRevB.92.241101 PACS number(s): 71.27.+a, 74.25.fg, 74.70.Tx, 75.30.Mb

Discoveries of heavy-fermion superconductors [1,2], organic superconductors [3], and high-T_c cuprates [4] in the 1970s–1980s gave us strong motivations to investigate nonphononic pairing mechanisms. Intrinsically, unconventional superconductivity often occurs out of a non-Fermi-liquid (NFL) state, where quantum fluctuations due to magnetic or charge instabilities are remarkable. Thus, the study of the origin of NFL behaviors is essential for elucidating the mechanism of unconventional superconductivity. To understand NFL metals, we often approach them on the basis of the primal concept of the *quasiparticle*, which has a one-to-one correspondence to a noninteracting Fermi gas. However, there exist many heavy-fermion superconductors for which the normal-state properties are far from those of Fermi-liquid (FL) metals. In particular, uranium heavy-fermion systems show quite peculiar magnetic properties and NFL metallic behaviors, reflecting strong c-f hybridizations, Fermi-surface instabilities, and exotic Kondo effects resulting from the nondipolar degrees of freedom for the $5f$ electron at the uranium site.

In the present work, we focus on the unusual heavy-fermion superconductor UBe_{13}, which crystallizes in a cubic $(O_h^6, Fm\bar{3}c)$ $NaZn_{13}$ structure. As a possible candidate for an odd-parity pairing with a nodal gap, the superconducting (SC) properties of UBe_{13} have been investigated intensively [5–9]. Quite unexpectedly, it has recently been demonstrated that the nodal quasiparticle excitations from the Fermi surfaces are *absent* [10]. Furthermore, the upper critical field $H_{c2}(T)$, which exceeds the Pauli limit and shows an *upturn* at $\sim T_{SC}/2$, cannot be explained in the framework of the BCS theory [11,12]. Also, an unusual anomaly has been observed in the SC phase from specific-heat [13,14], thermal-expansion [14], and magnetization measurements [15].

The normal-state properties of UBe_{13} are also extremely unusual. First, the large resistivity $\rho(T) \sim T$ above T_{SC} is very different from that of usual FL metals [12,16,17]. Second, the thermodynamic quantities, i.e, specific heat $C/T \propto -\log T$ [18] and magnetic susceptibility $\chi \sim -\sqrt{T}$ [19], show divergent behaviors in the normal state. To explain these NFL behaviors, the quadrupolar Kondo effect (QKE), which results in the non-Kramers Γ_3 doublet for the crystalline-electric-field (CEF) ground state of $5f^2$ (U^{4+}, $J = 4$) configuration, was proposed [20,21]. This NFL origin is of interest also in terms of a possible occurrence of an odd-frequency paring [21–23]. Another possible origin is the presence of a magnetic-field-induced antiferromagnetic (AF) quantum critical point (QCP) [18,24]. Also, it has recently been proposed that competition between formations of Kondo-Yosida (KY) and CEF Γ_1 singlets on the $5f^2$ state could explain the NFL behaviors in UBe_{13} [25,26].

A key point to understand the unusual NFL behaviors of UBe_{13} is to compare the magnetic-field dependencies of its transport properties with those of the thermodynamic quantities. The low-T C/T of UBe_{13} is insensitive to magnetic field compared to a usual spin-Kondo system [27] despite its small effective Fermi temperature of several degrees Kelvin [5]. In contrast, the resistivity is rather sensitive to magnetic field, and the negative magnetoresistance can be reproduced by the conventional spin-1/2 Kondo model [28], conflicting with the QKE. However, the spin-1/2 Kondo model cannot describe the normal-state magnetization curve [28]. Thus, there has been no clear explanation for the contradiction between the NFL behavior of thermodynamic quantities and that of transport properties in UBe_{13}.

In this Rapid Communication, in order to gain more insight into the NFL behaviors of UBe_{13}, we study its heat-carrier properties by means of the thermoelectric power (TEP). Broadly speaking, the TEP reflects the entropy flow of charge carriers [29]; thus, it is highly sensitive to the carrier concentration and the lifetime, as well as the density of states (DOS), of the electrons near the Fermi level ϵ_F. The TEP has recently attracted much interest as a useful probe to study exotic carrier properties of the strongly correlated electron systems [30–35].

The low-T TEP measurements were performed by one-heater and two-thermometers (RuO_2) techniques using a polycrystalline sample. We detected the dc voltage signal of the sample with a nanovoltmeter at low temperatures down to 0.15 K and at fields up to 16 T. We also measured the resistivity and specific heat for the same quality sample [36].

Figure 1(a) shows the temperature dependence of the resistivity at zero and fields up to 16 T. At 0 T, an almost

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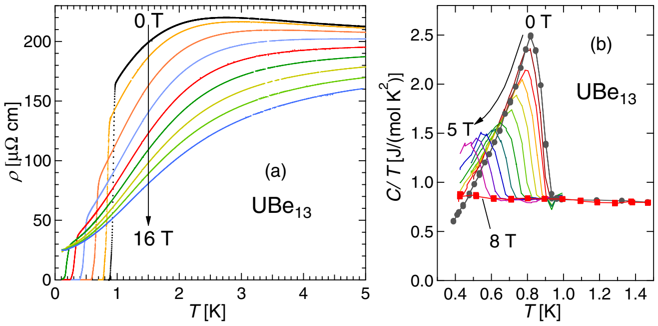

FIG. 1. (Color online) (a) Resistivity of UBe$_{13}$ at fields from 0 to 16 T (every 2 T). (b) C/T of UBe$_{13}$ at fields from 0 to 5 (every 0.5 T) and 8 T.

T-linear resistivity suddenly shows the SC transition at $T_{\mathrm{SC}}^{\rho}\sim 0.93$ K [37]. Also, $\rho(T)$ shows a maximum around 2.5 K, which shifts to higher temperatures with increasing field. Figure 1(b) shows $C(T)/T$ at fields up to 8 T. With increasing field, the large resistivity is suppressed, whereas $C/T\sim 1$ J K^{-2} mol^{-1} in the normal state is insensitive to magnetic field.

Figure 2(a) shows $S(T)$ of UBe$_{13}$ below 5 K at fields up to 16 T. At 0 T, the SC transition temperature is $T_{\mathrm{SC}}^{\mathrm{TEP}}\sim$ 0.95 K [37]. The large negative TEP is consistent with previous results measured up to 7.5 T [9]. The inset of Fig. 2(a) shows $S(T)$ at fields from 4 to 9 T in an enlarged view below 0.8 K. At 0 T, the value of S reaches approximately -18 μV/K, with a minimum anomaly at $\sim$1.4 K. We define $T_{\mathrm{min}}^{\mathrm{TEP}}$ as the temperature where $S(T)$ shows a minimum [Fig. 2(a)]. $T_{\mathrm{min}}^{\mathrm{TEP}}$ shifts to higher temperature at higher fields, corresponding to the resistivity results. This T dependence will be discussed later. Applying magnetic field substantially suppresses the large $|S|$; at 16 T, a change of sign in S occurs at $\sim$0.8 K.

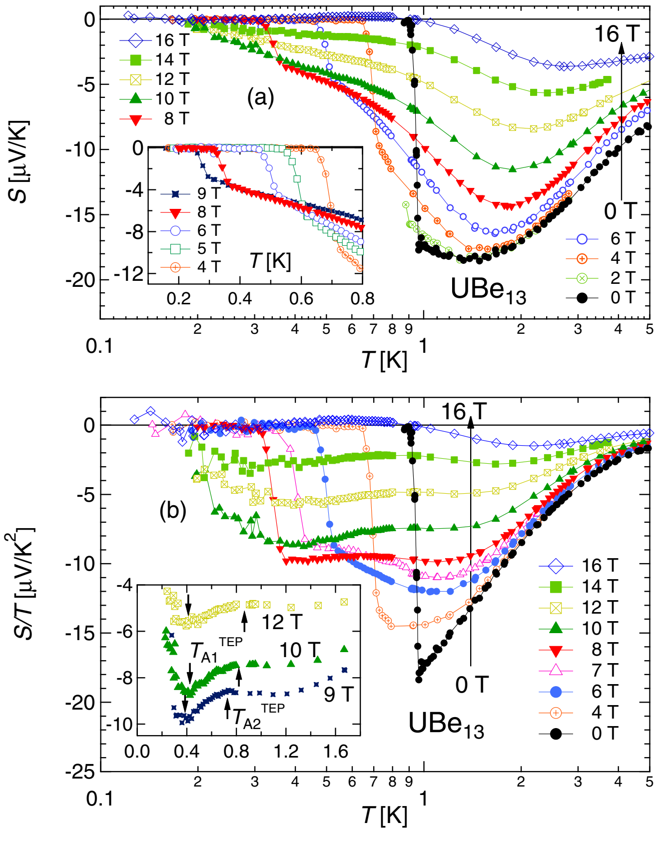

FIG. 2. (Color online) (a) The TEP S of UBe$_{13}$ at fields up to 16 T in a T-logarithmic scale. The inset shows $S(T)$ from 4 to 9 T in a T-linear scale. (b) S/T of UBe$_{13}$ at fields up to 16 T in a T-logarithmic scale. The inset shows S/T for 9, 10, and 12 T in a T-linear scale. Here, the down and up arrows indicate $T_{\mathrm{A1}}^{\mathrm{TEP}}$ and $T_{\mathrm{A2}}^{\mathrm{TEP}}$, respectively.

Using the Boltzmann equation, the TEP is described by the logarithmic derivative of the electronic conductivity $\sigma(\epsilon)$ at ϵ_{F}: $S=-\frac{\pi^2k_{\mathrm{B}}^2T}{3e}\frac{\partial\ln\sigma(\epsilon)}{\partial\epsilon}|_{\epsilon=\epsilon_{\mathrm{F}}}$. Here, k_{B} and e (>0) are the Boltzmann constant and elementary charge, respectively. This formula is also described as $S=-\frac{\pi^2k_{\mathrm{B}}^2T}{3e}[\frac{\partial\ln\tau_c(\epsilon)}{\partial\epsilon}+\frac{\partial\ln N_c(\epsilon)}{\partial\epsilon}]_{\epsilon=\epsilon_{\mathrm{F}}}$, where $\tau_c(\epsilon)$ and $N_c(\epsilon)$ are the lifetime and DOS for conduction electrons, respectively [29,38,39]. In the FL regime, the above formula leads to $S/T=$ const.

Although the normal state of UBe$_{13}$ is far from the FL regime for a single band, it is worthwhile to discuss the TEP results using the above simple model. In heavy-fermion systems, conduction electrons are strongly scattered by originally localized f electrons; then τ_c is much smaller than that in ordinary metals. At low temperature, the strong c-5f hybridization causes the sharp 5f DOS peak [$N_f(\epsilon)\propto\frac{1}{\tau_c(\epsilon)}$; Kondo resonance] near ϵ_{F} [40,41]. Since the conduction-electron DOS $N_c(\epsilon)$ does not vary rapidly, unlike the sharp 5f-electron DOS [40], the large low-T TEP in f-electron Kondo systems mainly comes from the contribution of $\frac{\partial\ln\tau_c(\epsilon)}{\partial\epsilon}|_{\epsilon_{\mathrm{F}}}=\frac{1}{\tau_c}\frac{\partial\tau_c(\epsilon)}{\partial\epsilon}|_{\epsilon_{\mathrm{F}}}\sim-\frac{1}{N_f}\frac{\partial N_f}{\partial\epsilon}|_{\epsilon_{\mathrm{F}}}$. The negative large TEP in UBe$_{13}$ implies that the sharp Kondo resonance peak may exist just below ϵ_{F}, leading to the huge $-\frac{\partial N_f}{\partial\epsilon}|_{\epsilon_{\mathrm{F}}}>0$ ($S<0$).

Figure 2(b) shows $S(T)/T$ of UBe$_{13}$ at zero and magnetic fields up to 16 T below 5 K. Interestingly, the low-T $|S/T|$ at 0 T is strongly enhanced without any FL behavior down to the SC transition. The strong enhancement of $|S/T|$ at 0 T below 2.5 K contrasts starkly with a conventional spin-1/2 Kondo system, in which the low-T ground state is the FL forming the KY singlet.

With increasing field, the large $|S/T|$ of UBe$_{13}$ is considerably suppressed on the whole, and the low-T S becomes almost zero at 16 T. Above 8 T S/T shows a new local-minimum anomaly, which becomes less distinct above 14 T; then S/T recovers FL-like behavior. S/T shows a local minimum at temperature $T_{\mathrm{A1}}^{\mathrm{TEP}}$ from $\sim$0.4 to 0.2 K, and we define $T_{\mathrm{A2}}^{\mathrm{TEP}}$ as the onset of this anomaly [$T_{\mathrm{A2}}^{\mathrm{TEP}}\sim 0.76$ K at 8 T; Fig. 2(b)]. $T_{\mathrm{A1}}^{\mathrm{TEP}}$ and $T_{\mathrm{A2}}^{\mathrm{TEP}}$ increase up to 12 T but decrease above 12 T with increasing field.

Let us now discuss why the TEP and resistivity are sensitive but the specific heat is *robust* to magnetic field in UBe$_{13}$. One may try to explain the suppression of the large resistivity [Fig. 1(a)] by the increment of n_c and/or τ_c using the formula $\rho=\frac{m^*}{n_ce^2\tau_c}$, where n_c and m^* are the concentration of conduction carriers and their effective mass, respectively. However, considering the relation of $N_f(\epsilon)\propto\frac{1}{\tau_c}$ in a heavy-fermion system, only the reduction of $\frac{1}{\tau_c}$ conflicts with the field-independent value of $C/T\propto N_f(\epsilon)$. Note that the above formula for resistivity is reasonable if $\tau_c(\epsilon)$ is almost independent of energy near ϵ_F [38]. For UBe$_{13}$, $\tau_c(\epsilon)$ is considered to be strongly energy dependent, reflecting the narrow Kondo resonance peak near ϵ_F [41]. The discrepancy between these transport quantities and specific heat versus magnetic field can be explained by strong field variations of the energy derivative of $\tau_c(\epsilon)$, although $N_f(\epsilon)$ near ϵ_F remains up to ~8 T. Due to the Zeeman effect, the sharp DOS $N_f(\epsilon)$ splits into double peaks for up and down spins [42]; if the up-spin-electron DOS shifts to above ϵ_F, the energy derivative of $\tau_c(\epsilon)$ may remarkably change at ϵ_F.

The concentration of the dominant heat carrier may also be affected by magnetic fields. As reported previously [43], the increment of the positive Hall coefficient down to ~1.5 K with cooling indicates that the hole-carrier concentration decreases at low temperature. Meanwhile, the observed negative TEP at 0 T suggests the electron-dominant heat carriers at low temperature. Then, in the presence of both electron and hole bands ($\sigma = \sigma_e + \sigma_h$), the TEP is described as $S = \frac{\sigma_e}{\sigma} S_e + \frac{\sigma_h}{\sigma} S_h$ with equal carrier numbers of electrons and holes. Since the conductivity for each band is not very different regardless of the effective mass of carriers, i.e., $\sigma_e \sim \sigma_h$ [44], the suppression of S suggests that the balance of dominant heat carriers changes considerably from $|S_e| \gg S_h$ at 0 T to $|S_e| \sim S_h$ at 16 T, meaning that the heavy-electron Fermi surface dominates the large negative S at 0 T. Such variation of the dominant heat carrier could be caused by the substantial change of the electron-hole asymmetry for the $5f$ DOS around ϵ_F due to the Zeeman effect.

Let us see the field dependence of the NFL S/T on the H-T phase diagram. Figure 3 shows the contour plot of S/T along with H_{c2} and the observed anomalies. Interestingly, the enhancement of $|S/T|$ is most pronounced at zero field. Also, the initial slope of $H_{c2}(T)$, i.e., $-dH_{c2}/dT|_{T=T_c}$ is huge, reflecting the heavy effective mass of electrons [11]. The peculiar bending behavior of $H_{c2}(T)$ around 2 T appears to be associated with the reduction of the large $|S/T|$.

Next, we focus on the field-induced anomaly above 8 T (Fig. 3). Previously, a field-induced anomaly has been reported from magnetic-torque measurements [45]. The line of the magnetic-torque anomaly is similar to $T_{A1}^{TEP}(H)$ and $T_{A2}^{TEP}(H)$. Their field dependencies remind us of some kind of magnetic-field-induced phase [45,46]. However, a clear phase transition has not been observed by specific-heat [18] and magnetization measurements [47]. $\rho(T)$ at constant field shows a crossover from a $T^{3/2}$ NFL behavior to a T^2 behavior [18]. In Ref. [18], it has been speculated that the NFL behaviors in UBe$_{13}$ may be connected to AF short-range correlations due to a field-induced QCP at $H_c(T=0) \sim 4.5$ T hidden in the SC state, where the line of anomalies in thermal expansion and specific heat vanishes as $T \rightarrow 0$ K. In this case, the A coefficient of $\rho(T) = \rho_0 + AT^2$ as well as S/T should also be enhanced around ~4.5 T, but such enhancements are not seen in resistivity up to 20 T [48,49] and $|S/T|$ (Fig. 3).

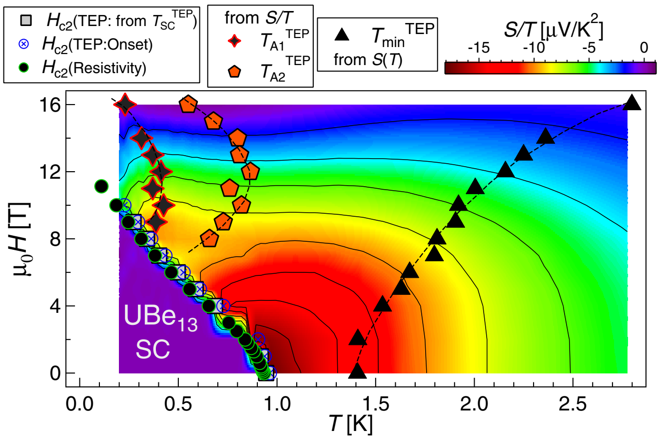

FIG. 3. (Color online) H-T phase diagram of UBe$_{13}$ obtained from resistivity and TEP measurements. T_{min}^{TEP} is the temperature where $S(T)$ shows a minimum. T_{A1}^{TEP} is the temperature where S/T shows a local-minimum anomaly above 8 T, and T_{A2}^{TEP} is its onset [Fig. 2(b)]. The dashed lines are a guide to the eye.

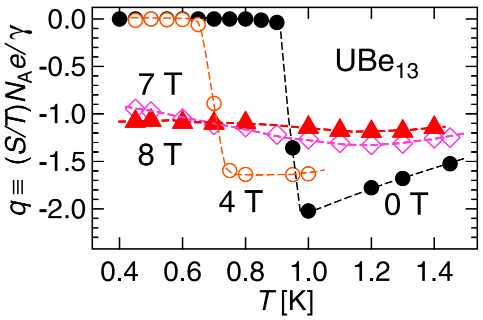

FIG. 4. (Color online) $q \equiv (S/T)N_A e/\gamma$ at 0, 4, 7, 8 T derived from the present S/T and C/T data.

To explore AF QCP, it is useful to examine the dimensionless ratio $q \equiv (S/T)N_A e/\gamma$, where N_A is Avogadro's number [39,44]. Miyake and Kohno have shown that $|q|$ decreases towards the AF QCP [44]. Figure 4 shows q of UBe$_{13}$ derived from the present S/T and C/T data. The value of $|q| \sim 2$ is larger than the values for typical heavy-fermion systems ($|q| \sim 1$) [39], but it decreases with increasing field. Due to the field-induced anomalies at $T_{A1}^{TEP}(H)$ and $T_{A2}^{TEP}(H)$, $|q|$ is slightly enhanced at 8 T. Since the large γ retains the order of 1 J K^{-2} mol^{-1} up to 12 T [18], $|q|$ does not show any criticality up to 12 T. In addition, $S(T)/T$ becomes rather modest above 12 T, implying that the system approaches the FL regime in high fields. Thus, from the TEP results, the magnetic-field region above 4 T for UBe$_{13}$ appears to correspond to the NFL-FL crossover region as observed also in the resistivity [18,49].

As discussed above, the anomalies at $T_{A1}^{TEP}(H)$ and $T_{A2}^{TEP}(H)$ cannot be explained by the field-induced AF QCP. We propose that the TEP anomalies above ~8 T may be caused by Fermi-surface reconstructions in the high-field region. Interestingly, it appears that the anomalies at $T_{A1}^{TEP}(H)$ and $T_{A2}^{TEP}(H)$ are related to the unusual upturn of $H_{c2}(T)$ above ~6–7 T (Fig. 3). Associated with the Zeeman splitting of the narrow $5f$ DOS, the Fermi-surface reconstruction in high fields and the change of the energy derivative of τ_c may affect its pair-breaking mechanism and cause the unusual upturn in $H_{c2}(T)$ around $T_{SC}/2$.

The enhancement of the NFL $|S/T|$ in UBe$_{13}$ is most remarkable at zero field. Since there is no field-induced quantum criticality in the QKE [20,21] and the competition

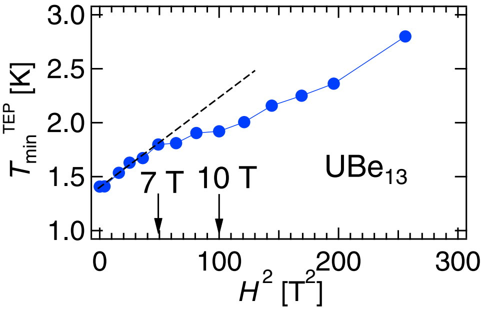

FIG. 5. (Color online) $T_{\mathrm{min}}^{\mathrm{TEP}}(H)$ obtained from the TEP S as a function of H^2.

between KY singlet and CEF singlet states [26], these candidates are more plausible for UBe$_{13}$. To obtain further information, it is intriguing to examine the field dependence of the anomaly of $S(T)$ at $T_{\mathrm{min}}^{\mathrm{TEP}}$ [Fig. 2(a)], which may characterize the energy scale of the Kondo effect in UBe$_{13}$. $T_{\mathrm{min}}^{\mathrm{TEP}}(H)$ is considered to correspond to the anomaly at around 2 K observed by resistivity (Fig. 1), specific heat [27], and the thermal-expansion coefficient [50]. Figure 5 shows $T_{\mathrm{min}}^{\mathrm{TEP}}$ vs H^2. Up to ~7 T, $T_{\mathrm{min}}^{\mathrm{TEP}}(H)$ is roughly proportional to H^2, consistent with previous studies [51], but it deviates from $\propto H^2$ above 7 T.

One possible scenario for the unusual NFL behaviors in UBe$_{13}$ could be the QKE [52], in which the $5f$ electron does not possess a magnetic moment ($\langle J_z \rangle = 0$) but a quadrupole moment, leading to the magnetically robust γ. A QKE system recovers a FL state by applying magnetic field, which splits the degeneracies of the "pseudospin" (quadrupole moment) through magnetostriction as well as the "channel" (spin) for conduction electrons [21,55]. It will be interesting to study whether such a FL recovering due to both the spin-field and channel-field splitting can reproduce the observed $T_{\mathrm{min}}^{\mathrm{TEP}}(H)$. In addition, as for its heat-carrier properties, the QKE may create the strong hole-carrier scattering due to the Kondo resonance peak below ϵ_{F}, leading to the large negative TEP (about -20 μV/K) with the electron-dominant thermal current [56]. In addition, since conduction electrons have the spin degree of freedom, the energy dependence of $\tau_c(\epsilon) \propto 1/N_f(\epsilon)$ near ϵ_{F} could change considerably in magnetic fields, even though the largeness of $N_f(\epsilon)$ remains, reflecting the nonmagnetic degree of freedom for the originally localized $5f$ electron.

Another possible origin of the NFL state in UBe$_{13}$ is the competition between the formation of a KY singlet and that of a localized CEF Γ_1 singlet of the $5f^2$ state [25,26]. At the characteristic temperature T_{F}^*, $5f$ electrons resolve the entropy by choosing a KY singlet or a CEF Γ_1 singlet as the ground state. According to theoretical studies [25,26], both $\gamma(H)$ and $T_{\mathrm{F}}^*(H)$ can be insensitive up to a certain field, H_z^*. Considering the magnetically robust γ [Fig. 1(b)], we roughly estimate $H_z^* \sim 10$ T; thus, $T_{\mathrm{F}}^*(H)$ is also expected to be robust up to ~10 T. However, the experimentally obtained $T_{\mathrm{min}}^{\mathrm{TEP}}(H)$ is rather sensitive below ~7 T (Fig. 5); thus, $T_{\mathrm{min}}^{\mathrm{TEP}}$ cannot be explained by T_{F}^* in this theory [25,26], which treats the CEF level in the tetragonal symmetry. Thus, further studies for the cubic symmetry are desired.

We finally compare the behaviors of UBe$_{13}$ in magnetic fields with those of a spin-1/2 Kondo system, which has been understood quantitatively. For a spin-1/2 Kondo system, the Zeeman splitting of Kondo resonance leads to the reduction of resistivity as in Ce$_x$La$_{1-x}$Al$_2$ ($x = 0.0064$) [42], in which the resistivity decreases to almost half in $H = 0.836T_{\mathrm{K}} = 1$ T at 0.08 K $= 0.1T_{\mathrm{K}}$ ($T_{\mathrm{K}} \sim 0.8$ K) [57]. In UBe$_{13}$, applying a field of 8 T, which is close to $H = 0.836T_{\mathrm{K}} \sim 10$ T, also reduces the resistivity to half, where $T_{\mathrm{K}} \sim T_{\mathrm{F}}^* \sim 8$ K [5]. However, the magnetically robust C/T in UBe$_{13}$ cannot be explained by the spin-1/2 Kondo system quantitatively, in contrast to Ce$_x$La$_{1-x}$Al$_2$ [58]. Theoretical studies of the Zeeman splitting of the Kondo resonance in nondipolar Kondo system as described above are strongly desired. It is also intrinsic to search for the Kondo resonance peak in UBe$_{13}$ with a narrow width of $k_{\mathrm{B}}T_{\mathrm{K}} \sim 0.7$ meV ($T_{\mathrm{K}} \sim 8$ K), which has not yet been observed [59].

In conclusion, we observed the quite exotic negative S/T, which strongly increases with no FL behavior down to the SC transition temperature. This strong enhancement of $|S/T|$ above T_{SC} is rapidly suppressed under magnetic field. We also investigated precise field dependencies of the observed anomalies, which characterize the energy scale of its unusual normal state. Interestingly, contrary to the drastic suppression of the TEP, the normal-state C/T is robust to magnetic field. Such unusual behavior can be explained by a considerable field variation of the energy derivative of $\tau_c(\epsilon)$, although the large $5f$-electron DOS itself remains under magnetic fields. This will be an important clue to understand the NFL behaviors of UBe$_{13}$ regardless of its physical origin.

We would like to acknowledge J. Flouquet, J. P. Brison, K. Izawa, Y. Ikeda, and K. Behnia for valuable discussions and comments. The present work was supported by an ERC starting grant (NewHeavyFermions), KAKENHI, REIMEI, and ICC-IMR.

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