

Spin-Triplet p -Wave Superconductivity Revealed under High Pressure in UBe_{13} Yusei Shimizu,^{1,*} Daniel Braithwaite,¹ Dai Aoki,^{1,2} Bernard Salce,¹ and Jean-Pascal Brison¹¹Université Grenoble Alpes, INAC/PHELIQS, CEA-Grenoble, F-38000 Grenoble, France²Institute for Materials Research (IMR), Tohoku University, Oarai, Ibaraki 311-1313, Japan (Received 27 May 2018; revised manuscript received 20 June 2018; published 12 February 2019)

To unravel the nature of the superconducting symmetry of the enigmatic $5f$ heavy-fermion UBe_{13} , the pressure dependence of the upper critical field and of the normal state are studied up to 10 GPa. Remarkably, the pressure evolution of the anomalous $H_{c2}(T, P)$ over the entire pressure range up to 5.9 GPa can be successfully explained by the gradual admixture of a field-pressure-induced E_u component in an A_{1u} spin-triplet ground state. This result provides strong evidence for parallel-spin pairing in UBe_{13} , which is also supported by the recently observed fully gapped excitation spectrum at ambient pressure. Moreover, we have also found a novel non-Fermi-liquid behavior of the resistivity, $\rho(T) \sim T^n$ ($n \lesssim 1$), which disappears with the collapse of the negative magnetoresistance behavior and the existence of a superconducting ground state around $P = 6$ GPa, suggesting a close interplay between Kondo scattering and superconductivity.

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Over the past few decades, the intricate relationship between magnetism and unconventional superconductivity has been a continuously evolving hot topic in solid state physics, stimulated by frequent new discoveries. The occurrence of superconductivity near a magnetic quantum critical point, where long-range magnetic order disappears at zero temperature and non-Fermi-liquid (NFL) behavior occurs, was demonstrated in heavy-fermion systems [1]. This implied a very strong probability that the unconventional pairing is due to spin fluctuations, and it was realized that similar phase diagrams, and so possibly similar pairing mechanisms, were actually found in very different families such as the high- T_c cuprates, iron pnictides, or organic superconductors. Still after almost 40 years, many questions remain open, and surprises are frequent. In particular, a microscopic description of the pairing mechanism(s) is missing in most cases, and, for the vast majority of heavy-fermion systems, there is no firm identification of the symmetry of the superconducting (SC) order parameter. A widely believed concept is that strongly correlated quasiparticles favor nodal SC gap structures to avoid strong Coulomb repulsion [2–4]. However, recent measurements have revealed that CeCu_2Si_2 , the first heavy-fermion superconductor discovered in 1979 [2], possesses a nodeless s -wave gap [5,6].

UBe_{13} is one of the earliest discovered heavy-fermion superconductors and also one of the most mysterious. No long-range magnetic order or quantum critical point is found close to the SC phase, although unusual NFL behavior occurs. Furthermore, whereas nodal gap symmetry had originally been discussed, from the power law T dependencies of physical quantities [7–10], recently nodeless behavior has also been found in UBe_{13} [11]. However,

the most remarkable feature of UBe_{13} is the temperature dependence of its upper critical field H_{c2} (see Fig. 1). It displays a huge initial slope (-42 T/K [12]) with a strong negative curvature in a low field, suggesting paramagnetic limitation. However, surprisingly, below about $T_c/2$, H_{c2} undergoes an anomalous upturn [13–15] and reaches a high value of 13–14 T, substantially exceeding the Pauli limit: Many explanations have been proposed to explain this strange temperature dependence (see, e.g., [14–18]), but it remains essentially a mystery.

In this Letter, we describe a key study using high pressure to tune the SC properties of UBe_{13} and resolve this 30-year-old mystery. We show that the pressure evolution of the temperature dependence of H_{c2} [$H_{c2}(T, P)$], which displays an apparent marked anomaly above 5 GPa, is in complete agreement with a model of triplet superconductivity with a field-induced mixture of two order parameters [18]. This makes UBe_{13} a rare

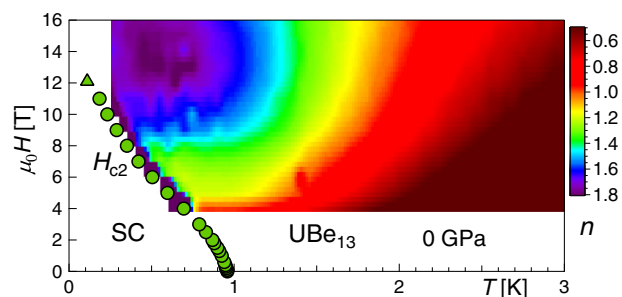


FIG. 1. H - T SC phase diagram at 0 GPa with a contour plot of the exponent (n) of the resistivity $\rho(T)$. The markers indicate H_{c2} , determined by temperature $\rho(T)$ (circles) and field $\rho(H)$ (triangle) scans.

paradigm of p -wave superconductivity, with a well-established mechanism for its response to a high magnetic field. As a bonus, this high-pressure study also demonstrates the deep interplay between NFL behavior and the appearance of superconductivity.

We have performed high-pressure experiments up to 10.3 GPa at very low temperatures down to 0.1 K. ac specific-heat, susceptibility, and resistivity measurements on polycrystalline samples were performed under high pressures using diamond-anvil cells, in a dilution refrigerator [19–22] (see Supplemental Material [23]). Resistivity data taken below 0.2 T under pressure are not shown, because they were masked by an extrinsic SC transition, presumably due to the parasitic uranium superconductivity.

Figure 1 is a good illustration of the issues raised by UBe_{13} . It shows our measurement of $H_{c2}(T)$ by resistivity at zero pressure, with its main puzzling features: a large initial slope followed by a strong negative curvature in turn followed by an upturn above 4 T. It also displays a color plot of the exponent (n) of the resistivity, i.e., $\rho(T) = \rho_0 + A'T^n$ [24] in the normal state, showing that a significant NFL behavior is seen around the SC state at low fields with $n \sim 1$ at 4 T just above T_c . A FL regime is progressively recovered with an increasing field: Previous studies [13] indicated that, at zero pressure, a FL regime seemed to be reached below 0.8 K under a magnetic field; however, with our more precise studies, even at 16 T, the exponent n approaches only $n \sim 1.8$.

Both the SC state and this NFL regime are sensitive to pressure: Figures 2(a)–2(c) show selected raw curves of $C_{ac}(T)/T$, measured at 0.7, 1.2, and 2.9 GPa: The anomalies of the SC transition are remarkably suppressed

with increasing P . Figure 2(d) shows the ac susceptibility χ_{ac} of UBe_{13} , measured under pressures from 0.5 to 5.8 GPa, displaying clearly the diamagnetic response from the SC state. Figure 2(e) shows the T - P phase diagram of UBe_{13} up to 8 GPa along with the Kondo-coherence temperature $T_{coh}(P)$, obtained from the previous magneto-resistance studies [25], and $T_{max}(P)$, the temperature at which resistivity shows a maximum (raw data not shown). $T_c(P)$ decreases almost linearly up to ~ 3 GPa and is consistent with the previous studies which extended up to ~ 2 GPa [26–28]. We have found that $T_c(P)$ decreases more slowly above 3.6 GPa, up to almost 6 GPa, and that the NFL regime is observed in the whole pressure range where superconductivity persists. Moreover, when superconductivity disappears at around 6 GPa, the FL regime appears, confirming the deep interplay between superconductivity and NFL regime in UBe_{13} .

To monitor the pressure evolution of the SC state of UBe_{13} , we have determined its upper critical field $H_{c2}(T, P)$. All the peculiar features of the temperature dependence of H_{c2} strongly change under pressure [28], a precious test bed to the different theoretical proposals. Figure 3(a) shows $H_{c2}(T)$ obtained from ac specific-heat data at 0, 0.7, 1.2, 2, and 2.9 GPa and resistivity data at 0, 2.1, 2.8, 3.6, 4.3, 5.6, and 5.9 GPa. The strong negative curvature and the upturn around $T_c/2$ are remarkably suppressed with increasing P , suggesting that the paramagnetic effect becomes less dominant. In addition, with increasing P , the initial slope $H'_{c2} \equiv [(\partial H_{c2})/(\partial T)]|_{T_c}$ is strongly suppressed, and it tends to saturate above 3 GPa, as seen in Fig. 3(b). In fact, H'_{c2}/T_c should be proportional to $1/v_F^2$ for superconductors in the clean limit, where v_F is the Fermi velocity. A remarkable and robust feature, already visible in the inset in Fig. 3(a) on the raw $H_{c2}(T)$ data, is that H'_{c2} is increasing again at 5.6 GPa, even though between 4.3 and 5.6 GPa the evolution of H_{c2} is continuous and not large. It is even more clear in Fig. 3(b), which reports the normalized value of H'_{c2}/T_c and shows that it is almost doubled between 4.3 and 5.6 GPa: Once deconvolved from the decrease of T_c , this is a very large effect. At first sight, it suggests a pressure decrease of v_F above 4.3 GPa, which could arise from band-structure effects (acting on the “bare” Fermi velocity) or from correlation effects (acting on the renormalization of the Fermi velocity). Up to now, no phase transition, which might cause a change of the band structure, has been detected in UBe_{13} up to 5.6 GPa. And, as regards electronic correlations, the trend is clearly toward a continuous pressure decrease: For example, the A coefficient of the resistivity decreases almost by a factor of 2 between 4.3 and 5.6 GPa [see Fig. 4(f) and the associated discussion]. Therefore, it was expected that $1/v_F$ and so H'_{c2}/T_c should continue to decrease at 5.6 GPa in contradiction with the present data. This is also why, e.g., the “extreme strong-coupling” model of UBe_{13} [28], where $H_{c2}(T, P)$ is

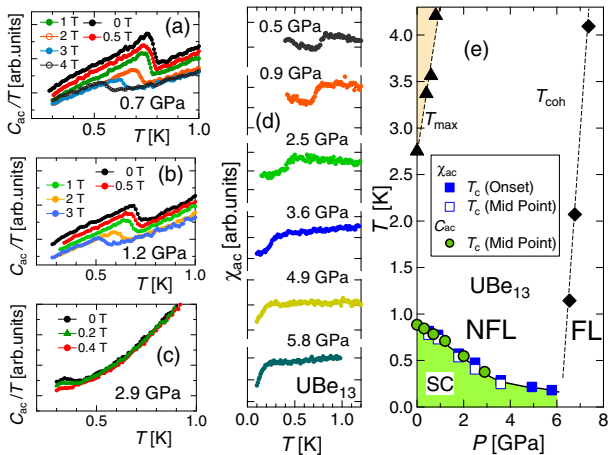


FIG. 2. C_{ac}/T (in arbitrary units) of UBe_{13} under high pressures, (a) 0.7, (b) 1.2, and (c) 2.9 GPa in zero and several fields. (d) $\chi_{ac}(T)$ in UBe_{13} , at zero field under pressures from 0.5 to 5.8 GPa. (e) T - P phase diagram of UBe_{13} . The SC transition temperatures were obtained from our C_{ac} and χ_{ac} measurements along with $T_{max}(P)$. Here, $T_{coh}(P)$ is taken from Ref. [25]. The dashed and solid lines are guides to the eye.

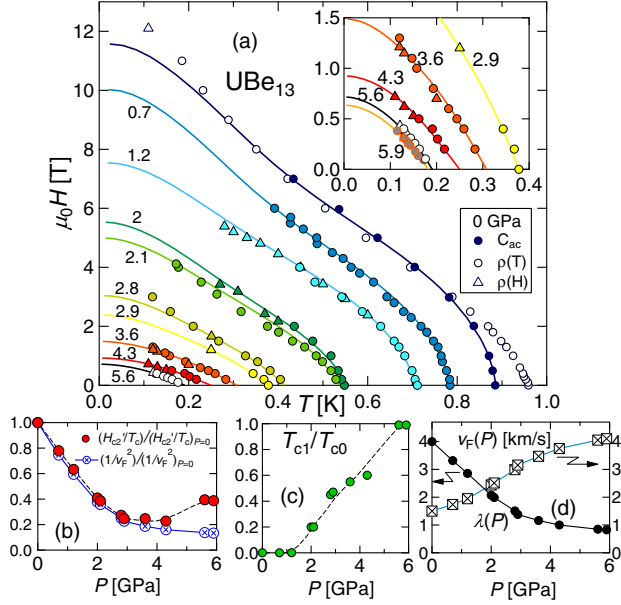


FIG. 3. (a) $H_{c2}(T)$ of UBe₁₃, obtained from ac specific-heat (0, 0.7, 1.2, 2, and 2.9 GPa) and resistivity (0, 2.1, 2.8, 3.6, 4.3, 5.6, and 5.9 GPa) measurements; circle and triangle markers indicate data determined from T and H scans, respectively; solid lines are fits to the $A_{1u} \oplus E_u$ model; the inset is a focus above 2.9 GPa. (b) $1/[v_F(P)]^2$ used in the fits above, compared with the resulting H'_{c2}/T_c , both normalized by their values at $P = 0$. (c) Pressure dependence of T_{c1}/T_{c0} , controlling the admixture of A_{1u} and E_u in the fits. (d) Strong-coupling constant $\lambda(P)$ (left) reproducing $T_c(P)$, and the resulting $v_F(P) = v_F(0)\{[1 + \lambda(0)]/[1 + \lambda(P)]\}$ (right).

presumed to arise from the sole pressure suppression of the strong-coupling constant λ , would fail at 5.6 GPa.

Interestingly, we found that a previously proposed model [18] for UBe₁₃, relying on a spin-triplet p -wave SC state, naturally reproduces the whole set of $H_{c2}(T, P)$ data up to 5.9 GPa, including the unexpected increment of H'_{c2} at 5.6 GPa. This model [18] relies on a SC ground-state order parameter, which is a fully gapped A_{1u} p -wave triplet state with a finite spin component $|S_z = 0\rangle$, equivalent to the B phase of superfluid ³He. This A_{1u} state displays a paramagnetic limitation with the initial strong negative curvature that would be partially lifted thanks to the field-induced admixture with an E_u SC order parameter, leading to the positive curvature at intermediate fields.

More precisely, H_{c2} depends on a new parameter which is the ratio of the pair potential for the A_{1u} and E_u representations, parametrized by the ratio of their respective critical temperatures (T_{c0} and T_{c1}) [31]. The d -vector order parameter is taken to be an admixture of one-dimensional A_{1u} ($\Psi_0 \propto \hat{x}k_x + \hat{y}k_y + \hat{z}k_z$) and two-dimensional E_u ($\Psi_1 \propto \hat{x}k_x + \hat{y}k_y - 2\hat{z}k_z$ and $\Psi_2 \propto \hat{y}k_y - \hat{x}k_x$) triplet states [18], and the calculations of H_{c2} are done in the weak-coupling limit. Note that, for a given Fermi velocity and critical temperature, the E_u state has a larger H'_{c2} than the A_{1u} state, so that the amount of admixture of the two

representations controls the paramagnetic limitation and also partly the orbital limitation. It is precisely this feature that will induce the anomalous increment of H'_{c2}/T_c .

The best fits of the data are presented as solid lines in Fig. 3(a), and the resulting evolution of T_{c1}/T_{c0} , controlling the admixture of A_{1u} and E_u states, is shown in Fig. 3(c). T_{c1}/T_{c0} is almost negligible below 2 GPa and then steadily increases to reach a value close to 1 at 5.6 GPa. The quality of the fits in Fig. 3(a), and the details of the parameter evolution in Fig. 3(b), show that this naturally accounts for the increase of H'_{c2}/T_c at this pressure: It results from the growth of the weight of the E_u component, which overcompensates the decrease of T_c (for H'_{c2}) and the slight increase of v_F . Indeed, the other parameter which has been varied from one pressure to the other, besides T_c , is the Fermi velocity controlling the orbital limitation. We have chosen to correlate the variation of these two parameters. It has been pointed out that the large specific-heat jump $\Delta C/\gamma T_c \sim 2.6$ [7] in UBe₁₃ suggests strong-coupling superconductivity. So we have used a separate calculation, this time in the strong-coupling regime (see [28,32]), to deduce how the strong-coupling constant λ should vary with the pressure in order to reproduce the variation of $T_c(P)$. From that, we imposed a variation of v_F according to $v_F(P) = v_F(0)\{[1 + \lambda(0)]/[1 + \lambda(P)]\}$ [Fig. 3(d)], with $v_F(0)$ and $\lambda(0) = 4$ adjusted against the zero pressure measurements and the main variation of $H'_{c2}/T_c(P)$.

Overall, the present data for the pressure evolution of H_{c2} in UBe₁₃ give strong support to an A_{1u} with a field-induced E_u admixture SC order parameter. They are compatible with a constantly increasing Fermi velocity, in coherence with a pressure decrease of the electronic-correlation strength, and the absence of a pressure-induced Fermi-surface anomaly below 6 GPa. Let us also note that recent specific-heat measurements in rotating fields [11] have revealed a fully gapped ground state for UBe₁₃ at zero pressure, further supporting the dominant fully gapped A_{1u} state used for the present fit of H_{c2} at $P = 0$.

We shall now discuss briefly the pressure effects of the NFL behavior of UBe₁₃. Figure 4 shows the resistivity in UBe₁₃ as a function of (left panel) T and (right panel) T^2 , obtained at (a) 0, (b) 2.8, (c) 3.6, and (d) 5.6 GPa. In Figs. 4(e) and 4(f), the resistivity vs T^2 and A coefficients for $R(T) = \rho_0 + AT^2$ are also plotted [29]. Except for 10.3 GPa, the resistivity is strongly field dependent. Note that no field-induced quantum critical behavior is seen in $A(H, P)$ as shown in Fig. 4(f).

At 2.8 and 3.6 GPa, interestingly, a quasi- T -linear behavior is observed in low fields ($\mu_0 H \lesssim 2$ T). This NFL behavior disappears with an increasing field, while the resistivity is strongly suppressed with the negative magnetoresistance. When the superconductivity disappears at $H_{c2}(T \rightarrow 0)$, a positive magnetoresistance, i.e., a band-like behavior, appears. Approaching the Kondo coherence at 6 GPa, the NFL regime characterized by a quasi- T -linear

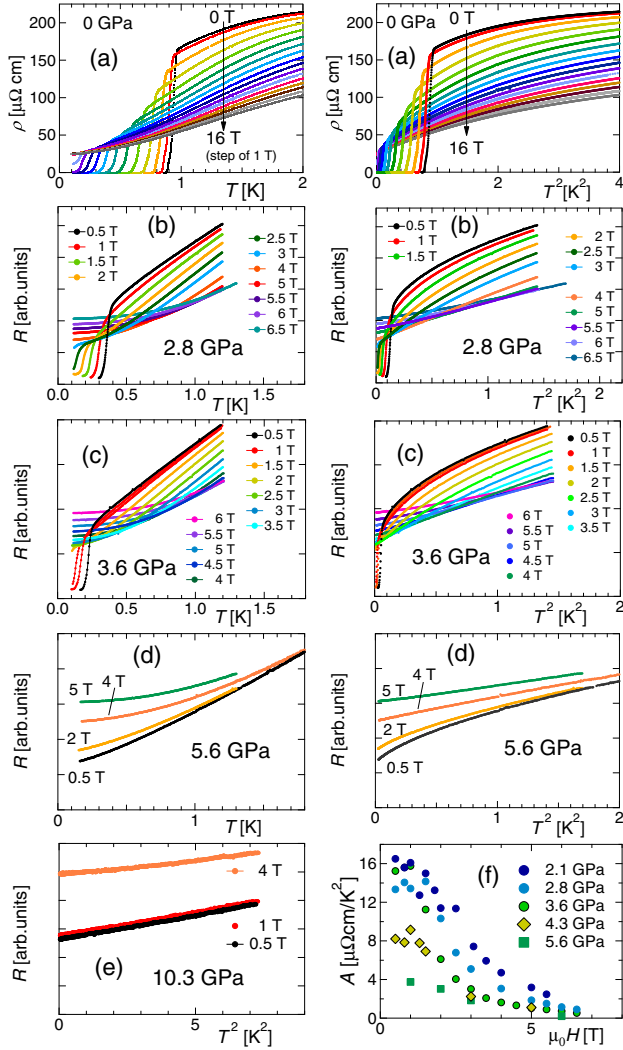


FIG. 4. Resistivity of UBe_{13} at (a) 0 GPa and under high pressures (in arbitrary units) of (b) 2.8, (c) 3.6, and (d) 5.6 GPa in zero and several fields vs (left) T and (right) T^2 . (e) Resistivity as a function of T^2 obtained at 10.3 GPa and (f) the field dependence of the A coefficient, obtained semiquantitatively for each pressure [29].

behavior becomes less pronounced: At 5.6 GPa, where the weak SC phase is still observed below ~ 0.4 T, only a positive magnetoresistance is observed (above 0.5 T). At 10.3 GPa, a FL behavior is observed in all magnetic fields. Aronson *et al.* showed that under pressure the (FL) Kondo coherence is recovered below 1 K for $6 < P < 6.5$ GPa [25]. In our low- T measurements, we could not accurately pinpoint the onset of FL behavior in a zero field because of the superconductivity. However, with an applied field of 1 T, we observe the NFL behavior down to the lowest temperature at 5.6 GPa, but a FL regime is recovered below 0.35 K at 5.9 GPa (see Supplemental Material [23]), which is compatible with the previous results [25]. These facts suggest that the NFL behavior is observed only in the pressure range where superconductivity occurs. This coincidence of the NFL regime with superconductivity looks very

similar to the behavior of UCoGe [33], and, although it still does not allow to identify a pairing mechanism in UBe_{13} , it does suggest a common origin for the fluctuations responsible for the NFL regime and those governing for the pairing mechanism. Nevertheless, the relation between the anomalous resistivity and superconductivity in UBe_{13} is far from trivial. Indeed, while the resistivity varies strongly in fields, the Sommerfeld coefficient has been shown to be quite field insensitive [13,34], implying that the SC pairing strength does not depend on the field.

Applying pressure on UBe_{13} depresses T_c and pushes the $5f$ -electron system away from the strongly correlated NFL regime [see $A(T, P)$], even if a remarkable feature of the NFL behavior in UBe_{13} is that the quasi-linear- T dependence of the resistivity accompanied by negative magnetoresistance exists over a wide P range, at least $0 \lesssim P \lesssim 4$ GPa.

It is instructive to compare with the situation in $\text{U}_{1-x}\text{Th}_x\text{Be}_{13}$ [35–37], for which quantum criticality appears with Th substitution [37]. Th substitution is expanding the lattice constant [36,38] and so can be viewed as a negative pressure. Given the recent arguments that the second anomaly at T_{c2} does not come from an antiferromagnetic transition [39] but from a SC double transition [40] with the time-reversal-symmetry broken state [41], we can exclude that superconductivity in UBe_{13} appears close to a magnetic quantum critical point: Neither positive nor negative pressure reveals magnetic ordering, in contrast to CeCoIn_5 [42].

A plausible scenario is quantum criticality between two nonmagnetic singlet states: A competition between a Kondo-singlet (itinerant) and Γ_1 -singlet crystalline-electric-field (CEF) (localized) ground states on $5f^2(\text{U}^{4+})$, invoking similar behaviors to those in two-channel-Kondo systems [43–45]. Indeed, such quantum criticality should be sensitive to a tuning parameter like pressure: A negative magnetoresistance behavior is switched to a positive magnetoresistance behavior with increasing hybridization effects [45], in agreement with our observations in low- and high- P regions (Fig. 4). In contrast, the originally proposed quadrupolar-Kondo effect [46] results in NFL behaviors due to a singularity of $5f$ electrons with a non-Kramers Γ_3 CEF ground state. It is not expected to depend on pressure without a crystal symmetry change. However, quadrupolar quantum criticality on the Γ_3 ground-state system strongly depends on hybridization effects, as observed recently in $\text{PrTi}_2\text{Al}_{20}$ [47]. Further studies for understanding the magnetoresistance in UBe_{13} [48,49] are necessary to challenge its NFL nature.

In conclusion, our low- T pressure experiments strongly support spin-triplet superconductivity in UBe_{13} with a fully gapped p -wave A_{1u} state and a field-induced admixture of an E_u component. This is a rare clear-cut example of triplet pairing in strongly correlated electron matter, in stark contrast to the recent identification of s -wave superconductivity in the first heavy-fermion CeCu_2Si_2 [5,6]. And even more rare, the

A_{1u} ground state of UBe_{13} at ambient pressure, like the B phase of superfluid ^3He , appears as “strong topological superconductivity” (in the classification of Ref. [50]). Moreover, experimentally good single crystals display very little residual values in the specific heat [11,13]. So UBe_{13} , with its nodeless gap symmetry, might be the best system to observe the influence of the predicted surface excitations (Majorana fermions), being free from bulk excitations. We have also found that the domain of existence under pressure of an anomalous NFL behavior of resistivity matches that of the SC state in UBe_{13} , pointing to a common origin for the spin-triplet pairing mechanism and this NFL regime.

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