Unusual pressure evolution of the Meissner and Josephson effects in the heavy-fermion superconductor UPt_3

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The Josephson effect between a single-crystal UPt_3 and a conventional superconductor Al has been investigated under pressure for the junction on the UPt_3 surface perpendicular to the hexagonal c[0001] axis. Simultaneously measured magnetization has revealed that the Meissner fraction approaches a minimum value of $\sim 1\%$ at the critical pressure of $P_c \sim 0.4$ GPa. The critical temperature T_c and the temperature T_J where a measurable Josephson critical current I_c first appears decrease with increasing pressure; the decreasing rate of $|dT_c/dP|$ is almost constant, whereas $|dT_J/dP|$ shows an abrupt increase above P_c, indicating that the Josephson effect is suppressed above P_c. These results, together with the decrease in penetration depth λ derived from the magnetic-field dependence of I_c, suggest that the superconducting phase is modified above P_c by the disappearance of the symmetry-breaking antiferromagnetic order.

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The heavy-fermion superconductor UPt_3, which has the electronic specific-heat coefficient $\gamma \sim 420$ mJ K^{-2} mol^{-1}, is one of the candidates for unconventional superconductors because the temperature dependence of various physical properties, such as specific heat [1], NMR relaxation rate [2], and ultrasound attenuation [3], shows a power-law behavior suggestive of the nodes of the energy gap. The NMR Knight shifts have revealed the odd-parity pairing state in UPt_3 [4]. Its remarkable features are the complex field-temperature (H-T) phase diagrams [5]: A (low-H, high-T), B (low-H, low-T), and C (high-H, low-T) phases. In order to explain these results comprehensively, various models have been proposed for the odd-parity order parameter described by the $\boldsymbol{d}$ vector: $\hat{\Delta}(\boldsymbol{k}) = i[\boldsymbol{d}(\boldsymbol{k}) \cdot \boldsymbol{\sigma}]\sigma_y$, where σ_x, σ_y, and σ_z are Pauli matrices. At present, two favorable candidates are E_{2u}: $\boldsymbol{d}(\boldsymbol{k}) \sim (k_a + ik_b)^2 k_c \hat{c}$ [6] and E_{1u}: $\boldsymbol{d}(\boldsymbol{k}) \sim (k_a \hat{b} + k_b \hat{c})(5k_c^2 - 1)$ [7,8].

The validity of these two scenarios is still controversial; small-angle neutron scattering [9], the angle dependence of the Josephson effect [10,11], and the polar Kerr effect [12] support the E_{2u} scenario, whereas thermal conductivity supports the E_{1u} scenario [7]. We have investigated the Josephson effect between UPt_3 and BCS superconductors and observed an anisotropic temperature dependence of Josephson critical current I_c at the transition from the A to the B phase [13]. The definite observation of the Josephson effect in the B phase for current flow along the hexagonal $a[11\bar{2}0]$, $b[10\bar{1}0]$, and c[0001] axes supports the E_{1u} scenario [14]. To confirm the validity of the scenario, the other phase, that is, the C phase is left to investigate.

Since the C phase appears in high magnetic fields, the Josephson effect, which is suppressed sensitively by a small magnetic field, cannot be observed. However, it has been found by longitudinal sound velocity [15] and dilatometric measurements that the C phase appears even in zero magnetic field by applying a uniaxial pressure above $P_c \sim 0.3$ GPa along the c axis, whereas only the B phase was observed above P_c by specific-heat measurements under hydrostatic pressure [16,17], where P_c is the pressure that makes the A phase disappear. These inconsistent results are not ascribed to the difference between hydrostatic and uniaxial pressures since the uniaxial stress on the basal plane has little effect on the double transition between the A and the B phases [18]. One possibility is that the BC phase boundary is so flat as a function of temperature that the specific heat shows no peak at the transition. The appearance of the C phase above P_c, on the other hand, seems to contradict the disappearance of antiferromagnetic order above P_c observed by neutron scattering [19] if this order is the symmetry-breaking field to split T_c of UPt_3. Thus the superconducting phase above P_c is still left unsettled [20,21].

In our preliminary investigation, we have attempted to observe the Josephson effect between UPt_3 and conventional superconductors (Al or Nb) under pressure for the a- and b-axis junctions [22] and found that the Josephson effect can be measured without the destruction of the junction. In this paper, we have investigated the superconducting phase above P_c by measuring the Meissner effect and the Josephson effect.

The single crystal of UPt_3, which was already used in our previous investigations [13,14,22–25], was grown by the Czochralski pulling method. Its high quality is ensured by the residual resistance ratio above 500 and double-superconducting-transition temperatures of $T_c^+ \sim 0.58$ and $T_c^- \sim 0.52$ K [26]. The crystal was cut to the cubic shape with edges of about 3-mm along the $a[11\bar{2}0]$, $b[10\bar{1}0]$, and c[0001] axes to use as a substrate. A superconductor-normal-

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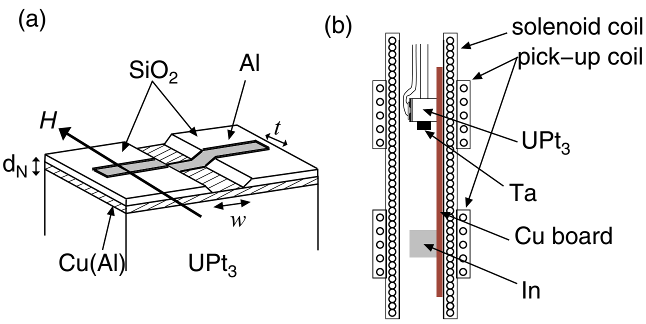

FIG. 1. Schematics of the experimental setup: (a) the Josephson junction and (b) the coils.

metal-superconductor (SNS′) junction was fabricated on the surface perpendicular to the c axis, where S, N, and S′ are UPt_3, Cu(Al) (Cu doped with 5 wt % Al), and Al, respectively, as shown in Fig. 1(a). The details of the junction fabrication were described in our previous report [14]. Hereafter, the junctions are denoted as $I \| c$ on the assumption that the preferred current direction is perpendicular to the surface. The dimensions of the present junction are listed in Table I.

Hydrostatic pressure was applied using a piston-cylinder pressure cell where daphne oil 7373 was used as a pressure medium. The small solenoid coil and an astatic pair of pickup coils were wound by superconducting wire (Nb-Ti). The sample and tantalum were set inside one of the pickup coils and indium was set inside the other as shown in Fig. 1(b). The applied pressure was determined by the change in the superconducting transition temperature of In.

The pressure cell was set to a ^{3}He-^{4}He dilution refrigerator and cooled to 60 mK. The magnetic field in the sample region was reduced by a μ-metal shield and a Cryoperm 10® shield outside the vacuum can. The magnetic flux change induced by the Meissner effect of UPt_3, Ta, and In was measured using a superconducting quantum interference device magnetometer linked to the pickup coils. The details of the determination of the residual field and the measurement of the Josephson effect are described in our previous paper [14].

In Fig. 2, we show the temperature dependence of dc magnetic susceptibility under various pressures. In the series of measurements using the present pressure cell, the lowest pressure was 0.13 GPa. The ZFC and FC susceptibilities indicate diamagnetism and the Meissner effect, respectively. The details of the measuring process are described in our paper [27]. The superconducting transition of UPt_3 was observed at lower temperatures without remarkable broadening of the transition width with increasing pressure. The sample always shows perfect diamagnetism of $4\pi\chi_{ZFC} = -1$, whereas the Meissner fraction χ_{FC}/χ_{ZFC} depends on pressure and is less than 1 by the trapping of magnetic flux in the sample.

TABLE I. Properties of the junction, where d_N, w, and t are the thickness of the Cu(Al) layer, the width between SiO_2 banks, and the width of the Al strip, respectively. The junction area S is expressed as $S = w \times t$.

Substrate	d_N (μm)	$S \times 10^{-5}$ (cm^2)	w (μm)	t (μm)
$I \| c$	0.4	1.65	42	39.4

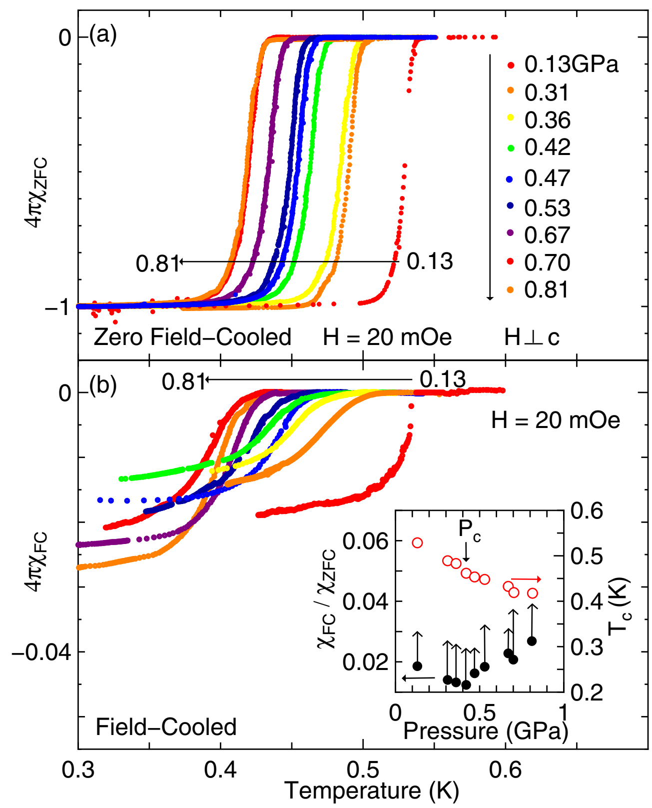

FIG. 2. Temperature dependence of dc susceptibility under various pressures for (a) zero-field-cooled (ZFC) and (b) field-cooled (FC) processes. The inset: pressure dependence of Meissner fraction ($=\chi_{FC}/\chi_{ZFC}$) at the temperature lower than T_c onset by 0.1 K (closed circle), the range of the increase extrapolated to 0 K using a differential coefficient at that temperature (arrow) and superconducting transition temperature T_c (open circle).

The pressure dependence of the superconducting transition temperature T_c, defined as the midpoint of the transition of χ_{ZFC} and the Meissner fraction at the temperature lower than the onset temperature by 0.1 K are plotted in the inset of Fig. 2(b). Although the kink of the superconducting normal-phase boundary, which is reported to appear at P_c [15,16], is unclear, the Meissner fraction shows a minimum at about 0.4 GPa, suggesting that the critical pressure P_c of the present UPt_3 is about 0.4 GPa. If the sample is broken or distorted with increasing pressure, the flux trapping sites should increase, and the Meissner fraction should decrease monotonously. We have observed, however, that the Meissner fraction is reproduced after releasing pressure. It should be noted that we have observed a steplike increase in the Meissner fraction of $CePt_3Si$ when the coexisting antiferromagnetic order disappears by applying pressure [28]. Considering that the antiferromagnetic moment vanishes above 0.37 GPa in the neutron scattering of UPt_3 [19], it may have some relationship with the flux pinning. One possibility is that antiferromagnetic domain walls act as the pinning sites below P_c. They at least explain the increase in the Meissner fraction above P_c.

We show in Fig. 3 the temperature dependence of Josephson critical current I_c under various pressures. The inset shows a typical I-V characteristic; a rapid increase in voltage owing

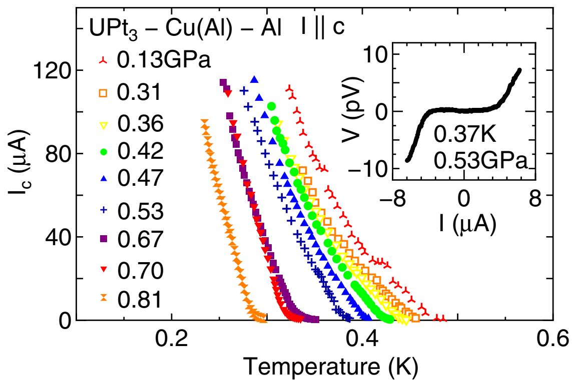

FIG. 3. (a) Temperature dependence of the Josephson critical current under various pressures for the c-axis junction. The inset: typical I-V characteristics at 0.53 GPa.

to the appearance of junction resistance is observed as the current exceeds the critical value I_c. As the pressure is increased, I_c becomes measurable at lower temperatures since the superconducting transition temperatures of both UPt_3 and Al decrease. Hereafter, we define T_J as the temperature where a measurable Josephson current first appears and use it as a measure of the Josephson coupling. Since the Josephson coupling through the normal layer of this junction is small, T_J is lower than T_c at each pressure. The pressure dependence of T_J is discussed below.

Figure 4 shows the typical magnetic-field dependence of I_c under several pressures; similar $I_c(H)$ curves have been obtained under all the applied pressures. The solid lines are fits using the Fraunhofer diffraction pattern $I_c(H) = I_{c0}|\sin[\pi(H - H_0)/\Delta H]|$, where H_0 is a parameter indicating the residual field. The I_{c0} and ΔH values obtained by fitting are shown in Fig. 4(d). The $I_c(H)$ curves were measured at temperatures where I_c in $H = 0$ is approximately 40 μA with

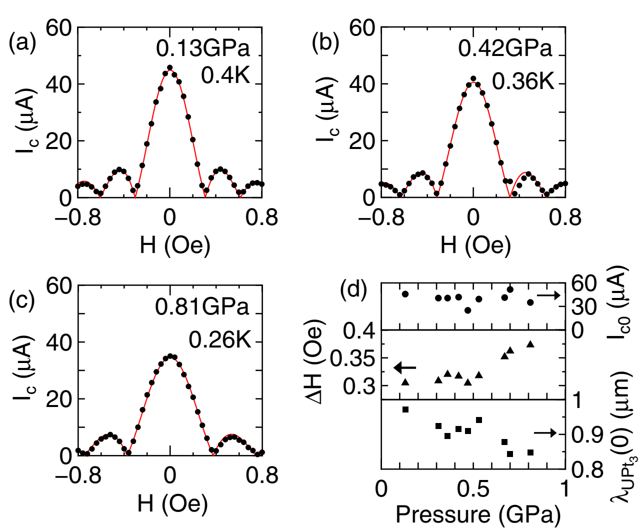

FIG. 4. (a)–(c) Magnetic-field dependence of the Josephson critical current under various pressures for the c-axis junction. The curves in the figures indicate fits using the Fraunhofer diffraction pattern. (d) Pressure dependence of maximum critical current I_{c0} and magnetic penetration depth λ at 0 K obtained by fitting.

a few exceptional cases as seen in the scatter of I_{c0} in Fig. 4(d) and the variation of ΔH depends little on I_{c0}. The $I_c(H)$ curves follow the Fraunhofer diffraction pattern, suggesting that the quality of the junction judged from the uniformity of the Josephson coupling is not deteriorated by applying pressure.

For a rectangular SNS′ Josephson junction, the magnetic flux threading through the junction is expressed as

$$\Phi = Hw(d_N + \lambda_{UPt_3} + \lambda_{Al}), \quad (1)$$

where Φ, λ_{UPt_3}, and λ_{Al} are the magnetic flux, the penetration depth of UPt_3, and Al, respectively. Considering that λ_{Al} is much smaller than $d_N = 0.4$ or $\lambda_{UPt_3} \sim 0.7$ μm [29], λ_{Al} is neglected. The oscillation period ΔH corresponds to one flux quantum Φ_0 threading through the junction.

As shown in Fig. 4(d), ΔH is approximately constant below 0.5 GPa and then begins to increase above 0.5 GPa. Considering that the compressibility κ of Cu [30] and UPt_3 [31] are 6.62×10^{-3} and 4.81×10^{-3} GPa^{-1}, respectively, the contraction of junction area should be on the order of 10^{-2} even when $P \sim 0.81$ GPa is applied; the increase in ΔH by 25% at 0.81 GPa cannot be explained. One possible explanation is that the penetration depth of UPt_3 is decreased by applying pressure above 0.5 GPa. To compare λ_{UPt_3} at different pressures, we estimate λ_{UPt_3} at 0 K using the empirical temperature dependence $\lambda(T) = \lambda(0)/\sqrt{[1-(T/T_c)^4]}$ as follows:

$$\lambda_{UPt_3}(0) = \left(\frac{\Phi_0}{\Delta H w} - d_N\right)\sqrt{1-\left(\frac{T}{T_c}\right)^4}. \quad (2)$$

The deviation from the reported value of $\sim$0.7 μm [29] is ascribed to the demagnetizing factor and the surface damage; if we assume the demagnetizing factor of 1/3 for a cubic sample, the estimated λ becomes 2/3 of the values in Fig. 4(d), whereas the surface damage tends to lengthen λ.

Since the London penetration depth is proportional to $\sqrt{m/n_s}$, where n_s and m are the superfluid density and the effective mass, respectively, a decrease in m and/or an increase in n_s may occur above P_c. Although the pressure dependence of m under hydrostatic pressure is unknown, m tends to decrease above P_c, if an abrupt change in m begins at P_c since the Sommerfeld constant γ decreases monotonously with an increase in uniaxial stress along the a axis, whereas it initially increases then levels off at P_c for the uniaxial stress along the c axis [18].

Figure 5 shows the pressure-temperature phase diagram that includes our results of the Josephson effect and magnetization measurements. The phase boundary between the normal and the superconducting phases was determined by magnetization measurements. The tricritical or tetracritical point is indicated by the arrow, where P_c corresponds to the pressure at which the Meissner fraction has a minimum. The double-transition temperatures at ambient pressure, which were observed for the UPt_3-Nb junction [23], are denoted by the open circles. The closed triangles indicate the temperature T_J where a measurable Josephson current appears. The short dashed line is the possible phase boundary between the B and the C phases drawn using the dT/dP value in the literature [15].

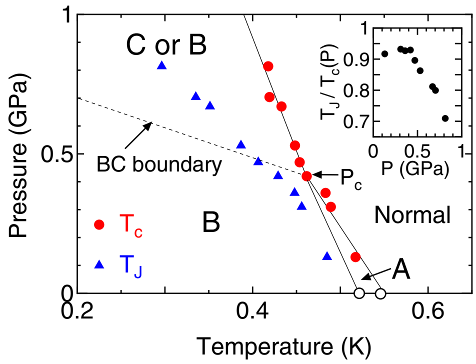

FIG. 5. Phase diagram of UPt_3 under pressure. The closed circles and closed triangles indicate T_c and T_J, respectively. The open circles indicate the double-transition temperature at ambient pressure obtained for the UPt_3-Nb junction [23]. The straight lines indicate the phase boundaries. The dashed line shows the possible BC phase boundary drawn using the reported dT/dP value [15]. The inset: pressure dependence of T_J/T_c.

As the pressure is increased, T_J decreases, and above P_c, it decreases more rapidly, which is clearly indicated by the pressure dependence of T_J/T_c in the inset, where T_J/T_c is the reduced temperature at which I_c reaches a measurable value. In this paper, we use T_J/T_c as an indicator of the magnitude of the Josephson effect since the rapid increase in I_c below T_J prevents the estimate and comparison of I_c values at 0 K. It is obvious that the Josephson effect is suppressed above P_c.

The Josephson effect between spin-singlet and spin-triplet superconductors is forbidden if the tunneling Hamiltonian does not change the spin, and it can occur along the $\boldsymbol{n}$ direction owing to the Rashba spin-orbit coupling in the spin-triplet superconductor, expressed as [32,33]

$$I = \frac{-2e}{\hbar} Im \sum_{k,l} \frac{T(\boldsymbol{k},\boldsymbol{l})T'(-\boldsymbol{k},-\boldsymbol{l})(\hat{\boldsymbol{k}} \times \hat{\boldsymbol{n}}) \cdot \boldsymbol{d}^*(\boldsymbol{k})\Psi(\boldsymbol{l})}{E_k E_l (E_k + E_l)}, \tag{3}$$

where $\boldsymbol{k}$ and E_k ($\boldsymbol{l}$ and E_l) are the wave number and the energy of the quasiparticle for the triplet (singlet) superconductor, respectively; $T(\boldsymbol{k},\boldsymbol{l})$ and $T'(-\boldsymbol{k},-\boldsymbol{l})$ are the spin diagonal matrix element and the tunneling matrix element due to spin-orbit coupling, respectively; $\boldsymbol{n}$ is a unit vector normal to the interface, and $\hat{\boldsymbol{k}} = \boldsymbol{k}/k_F$. The order parameter for the singlet (triplet) superconductor is written as $\Delta^s(\boldsymbol{k}) = i\Psi(\boldsymbol{l})\sigma_y$ $\{\Delta^t(\boldsymbol{k}) = i[\boldsymbol{d}(\boldsymbol{k}) \cdot \boldsymbol{\sigma}]\sigma_y\}$, where σ_x, σ_y, and σ_z are Pauli matrices.

In the case of the E_{2u} scenario, that is, $\boldsymbol{d}(\boldsymbol{k}) \sim (k_a + ik_b)^2 k_c \hat{c}$ in the B phase and $\boldsymbol{d}(\boldsymbol{k}) \sim 2ik_a k_b k_c \hat{c}$ in the C phase, $(\hat{\boldsymbol{k}} \times \hat{\boldsymbol{n}}) \cdot \boldsymbol{d}^*$ is always zero for $\hat{\boldsymbol{n}} \| c$, and the Josephson coupling is prohibited in the B and C phases in the c-axis direction. This prohibition has been calculated in detail for E_{2u} and E_{1g} scenarios of UPt_3 [34]. Even if a slight deviation of the surface normal from the c axis exists and causes the Josephson effect, the Josephson currents that come from the two components of the order parameter have a phase difference of $\pi/2$, which should lead to a significant deviation of the $I_c(H)$ curves from the Fraunhofer diffraction pattern.

In the case of the E_{1u} scenario, that is, $\boldsymbol{d}(\boldsymbol{k}) \sim (k_a\hat{b} + k_b\hat{c})(5k_c^2 - 1)$ in the B phase and $\boldsymbol{d}(\boldsymbol{k}) \sim k_b\hat{c}(5k_c^2 - 1)$ in the C phase, the Josephson coupling along the c axis is allowed in the B phase and prohibited in the C phase because Eq. (3) gives a finite value for $\boldsymbol{d}(\boldsymbol{k}) \sim k_a\hat{b}(5k_c^2 - 1)$ and not for $\boldsymbol{d}(\boldsymbol{k}) \sim k_b\hat{c}(5k_c^2 - 1)$.

In zero magnetic field, two types of pressure-temperature phase diagrams have been proposed; in the superconducting state, one includes the A–C phases [15,21] and the other includes the A and B phases [16,17]. In both diagrams, the A phase appears in a narrow region right below T_c and disappears above P_c; the Josephson effect in the A phase cannot be investigated by the present junction because of its weak Josephson coupling through the Cu(Al) layer. A phase observed at ambient pressure for the UPt_3-Nb junction is shown in Fig. 5.

In the former phase diagram that includes the C phase, the present result indicates that the Josephson effect is allowed in the C phase if the boundary between the B and the C phases is the dashed line; the result contradicts the E_{1u} scenario. If the E_{1u} scenario is valid; the latter phase diagram that does not include the C phase is favorable; the Josephson effect is observed in the B phase both above and below P_c. The suppression of the Josephson effect above P_c is then ascribed to some modification of the B phase induced by the disappearance of the antiferromagnetic order, which is the possible symmetry-breaking field in UPt_3 [19]. It should be noted that the origin of suppression is different from that of the decrease in $\lambda \propto \sqrt{m/n_s}$; the increase in n_s should generally increase I_c, and the decrease in m should also increase I_c since the matrix elements $T(\boldsymbol{k},\boldsymbol{l})$ and $T'(-\boldsymbol{k},-\boldsymbol{l})$ in Eq. (3) are inversely proportional to m [33].

One possible explanation is as follows: In the two-component order parameter, such as E_{1u} and E_{2u}, the order parameter in the B phase is expressed as $\Delta_B = \Delta_A + \Delta_C$, where Δ_A and Δ_C are the order parameters in the A and C phases, respectively; in the E_{1u} scenario, Δ_A allows the Josephson effect along the c axis, and Δ_C prohibits it. When the antiferromagnetic order, which is the symmetry-breaking field that lifts up T_c of Δ_A, vanishes above P_c, the ratio $|\Delta_A|/|\Delta_C|$ probably decreases, and the Josephson effect along the c axis is suppressed since it occurs only by Δ_A. In contrast, the penetration depth depends on the total order parameter $\Delta_A + \Delta_C$. A similar change has been proposed for the E_{2u} order parameter as the "elliptical" to "circular" phase transition at P_c [20].

In conclusion, we have determined the critical pressure of $P_c \sim 0.4$ GPa of UPt_3 by the minimum of the Meissner fraction. The E_{1u} scenario favors the pressure-temperature phase diagram that expects only the B phase as the superconducting phase above P_c. Still, the suppression of the Josephson effect and the decrease in λ suggest that the B phase is modified above P_c.

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