Anisotropy of the Josephson Critical Current between UPt$_3$ and Nb

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Josephson critical current densities J_c between a single crystal UPt$_3$ and an s-wave superconductor have been investigated for UPt$_3$-Cu-Nb junctions and UPt$_3$-Nb junctions. The anisotropic temperature dependence is observed at about the lower critical temperature T_c^- of UPt$_3$; the increase in J_c with decreasing temperatures becomes fast below T_c^- for a current flow parallel to the crystallographic c axis and it becomes rather slow below T_c^- for a current flow parallel to the b axis. This anisotropy gives clear evidence that the Josephson effect reflects the anisotropic superconductivity of UPt$_3$. [S0031-9007(98)07805-3]

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Since the discovery of heavy-fermion superconductivity in CeCu$_2$Si$_2$ and U-based systems, many studies have been made to clarify whether the pairing state of heavy-fermion superconductors (HFS) has a lower spatial symmetry than a conventional s-wave state. Among HFS, UPt$_3$ possesses a complex field temperature $(H\text{-}T)$ phase diagram [1], suggesting that it is an unconventional superconductor. In addition, the temperature dependence of various physical properties, such as specific heat [2], and the direct observation of the energy gap by point-contact spectroscopy [3] suggest a gap function vanishing on the Fermi surface. Recent studies on the NMR Knight shift [4] and the equilibrium magnetization in the superconducting mixed state [5] have suggested that UPt$_3$ is an odd-parity superconductor. It is still an open question as to which type of representation the order parameter of UPt$_3$ belongs.

The Josephson effect between a conventional and an unconventional superconductor gives information about the spatial symmetry of the order parameter, although no Josephson current [6] or even a quasicritical current [7] has thus far been observed for point contacts between UPt$_3$ and a conventional superconductor. It will be useful to investigate another type of Josephson junction, for example, a SNS$'$ junction in which two superconductors S and S$'$ are separated by a normal metal N. Recently, we have reported that the SNS$'$ junction between HFS (CeCu$_{2.2}$Si$_2$, UPd$_2$Al$_3$) and Nb also shows the Josephson effect [8]. In this paper, the distinct Josephson effect of UPt$_3$-Cu-Nb junctions (SNS$'$ junctions) and UPt$_3$-Nb junctions (SS$'$ junctions), especially the anisotropy according to the current directions, is described and is related to the unconventional order parameter of UPt$_3$.

The single crystals of UPt$_3$ have been grown by the Czochralski pulling method in a tetra-arc furnace. The residual resistivity ratio (RRR) was above 500, which indicates that the sample quality is sufficiently good. A clear double superconducting transition was observed at $T_c^+ \sim 0.58$ K and $T_c^- \sim 0.53$ K in a specific heat measurement for a single crystal of which RRR was similar to the one used for the present investigation [9]. The upper critical temperature T_c^+ determined in an ac susceptibility measurement was 0.54 K. Two pieces of crystals denoted as S-1 and S-2 were cut from the same ingot 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. The SNS$'$ junctions were fabricated on the surface perpendicular to the b and c axes. Throughout this paper the junctions are denoted as $I \parallel b$ or $I \parallel c$ on the assumption that the preferred current direction is perpendicular to the surface. The sample surface was rf sputter etched by Ar ion, and then Cu (normal metal), SiO$_2$, and Nb (s-wave superconductor) were deposited by the rf sputtering technique, as shown in Fig. 1. The thickness d_N of Cu and the junction area S are listed in Table I. The SS$'$ junctions were fabricated in the same way without the Cu layer. The current leads were attached to one end of the Nb strip and UPt$_3$, and the voltage leads were attached to the other end of the Nb strip and UPt$_3$. The electronic mean free path ℓ_N in Cu at 4.2 K was 0.6 μm ($d_N = 10, 20$ μm) and 0.2 μm ($d_N = 0.8$ μm), which was calculated from the resistivity of the adjacent Cu strip deposited at the same time.

The dc voltage was measured using a SQUID voltmeter which is constructed with a series combination of a

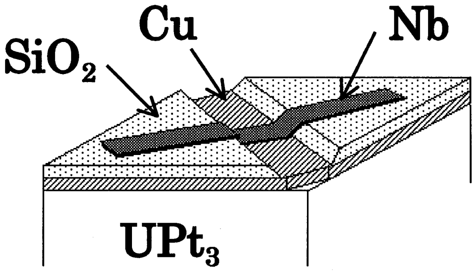


FIG. 1. Sample arrangement of UPt$_3$-Cu-Nb junctions. UPt$_3$-Nb junctions have a similar structure without the Cu layer.

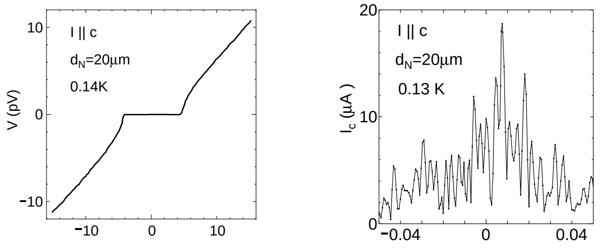
TABLE I. Properties of samples. The samples without the d_N value are the UPt$_3$-Nb junctions.

Substrate	d_N (μm)	S (mm^2)	R ($\mu\Omega$)	ρ_b (n$\Omega \cdot$ cm^2)
S-2 ($I \parallel c$)	$\cdots$	0.12	4.9	5.7
S-1 ($I \parallel c$)	10	0.21	0.87	1.8
S-2 ($I \parallel c$)	20	0.20	0.81	1.5
S-1 ($I \parallel b$)	$\cdots$	0.15	7.1	10
S-2 ($I \parallel b$)	0.8	0.19	3.5	6.7
S-1 ($I \parallel b$)	10	0.26	3.3	8.4
S-2 ($I \parallel b$)	20	0.19	1.1	2.0

standard resistor (2.49 $\mu\Omega$) and an inductance coupled to the SQUID. The voltage resolution was about 10^{-13} V. The sample was cooled down to 30 mK using a dilution refrigerator. The magnetic field in the sample region was reduced to less than 3 mOe by a μ-metal shield.

At temperatures which are not low enough to permit a measurable supercurrent, the current-voltage characteristics of all of the samples are Ohmic and the junction resistance R can be determined, as listed in Table I. The boundary resistivity defined as $\rho_b = (R - R_{Cu}) \times S$ for SNS$'$ junctions and $\rho_b = R \times S$ for SS$'$ junctions, where R_{Cu} is the calculated resistance of Cu, is also tabulated. As the temperature is lowered, measurable supercurrent appears, as shown in Fig. 2(a). A continuous rise in voltage is observed, as the current is increased from the critical value I_c. The I-V curve is single valued and not hysteretic, which is typical for SNS$'$ junctions.

To test the uniformity of the SNS$'$ junction, a magnetic field was applied parallel to the Cu film, and the critical supercurrent I_c was measured. If a field is applied to a uniform junction whose width normal to the field is smaller than the Josephson penetration depth λ_J, a Fraunhofer diffraction pattern should be observed. In the present junctions, estimated λ_J becomes smaller than the width of the junction when I_c exceeds approximately 20 μA. Figure 2(b) shows the typical magnetic field dependence of I_c. Although the falling envelope is seen with an increase in the magnetic field, I_c oscillates with no definite period. This pattern suggests that the junction is not uniform, that is, the local critical supercurrent density fluctuates spatially. When I_c increases and the junction becomes self-field limited, I_c becomes less sensitive to the magnetic field. In the case of SS$'$ junctions, the decrease in J_c is small within the magnetic field which can be applied; typically, J_c is still more than half of the maximum value when a field of 1.3 Oe is applied.



(a) (b)

FIG. 2. Typical properties of the UPt$_3$-Cu-Nb junction where$I \parallel c$ and $d_N = 20$ μm. (a) I-V characteristic showing Josephson critical current and (b) magnetic field dependence of I_c suggesting that the junction is not uniform. The solid line through the data points is a guide to the eye.

We show in Fig. 3 the temperature dependence of critical supercurrent density J_c, defined simply as $J_c = I_c/S$. The critical supercurrent rises rapidly as the temperature is reduced, and is still increasing even at the largest current value which can be flowed without heating the sample. The J_c value becomes small with an increase in Cu thickness d_N. Comparison of the samples with the same d_N or without the Cu layer indicates that J_c is smaller when the current flows parallel to the b axis.

Figure 4 shows the temperature dependence of J_c at about the lower transition temperature T_c^- of UPt$_3$. Since the transition at T_c^- is suggested to be of second order [2] and J_c of SS$'$ and SNS$'$ junctions is generally expressed as the sum of the terms which involve $|F_S||F_{S'}|$ or its integral powers, where $|F_S|$ and $|F_{S'}|$ are the condensation amplitude of S and S$'$, respectively [10], the transition at T_c^- is observed as a kink, as indicated by the arrows in Fig. 4. The difference in T_c^- from 0.53 K determined by the specific heat measurement may be attributed to the difference between ingots or to the fact that the Josephson effect reflects the superconductivity of the surface region where the junction is prepared rather than that of the bulk sample.

In the case of $I \parallel c$, small J_c is observed only for the SS$'$ junction above T_c^-, while J_c increases steeply with

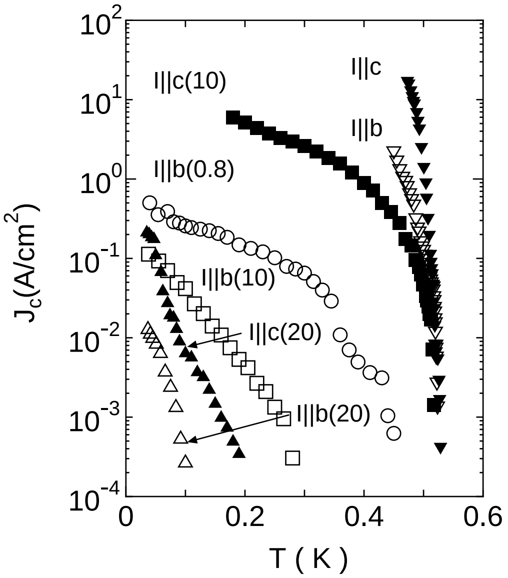


FIG. 3. Temperature dependence of J_c for five UPt$_3$-Cu-Nb junctions and two UPt$_3$-Nb junctions. In the case of UPt$_3$-Cu-Nb junctions, the thickness d_N (μm) of Cu is referred to by the figures in parentheses.

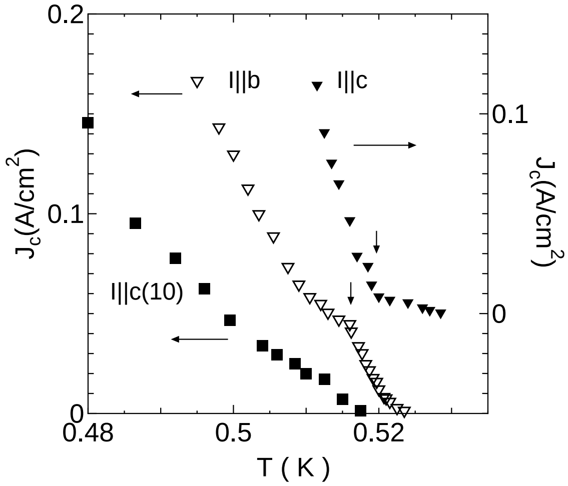


FIG. 4. Temperature dependence of J_c at about T_c^-. In the case of UPt$_3$-Cu-Nb junctions, the thickness d_N (μm) of Cu is referred to by the figures in parentheses. The arrows (↓) indicate the values of T_c^- observed in this experiment.

a decrease in temperature below T_c^-. The fact that J_c becomes measurable below T_c^- for the SNS$'$ junction may relate to the result that the gap-related spectra for $I \parallel c$ are observed only below T_c^- in point-contact spectroscopy [3]. In contrast to $I \parallel c$, the increase in J_c becomes rather slow below T_c^- for $I \parallel b$.

The small difference in T_c^- between $I \parallel c$ and $I \parallel b$ may be interpreted several ways: (1) It may indicate the small difference in T_c^- of the bulk sample between two pieces of crystals (S-1, S-2) cut from the same ingot, as listed in Table I; (2) the strain which is inevitably introduced during the fabricating process of the junction, and may depend on the crystallographic direction perpendicular to the surface where the junction is made, easily causes the change in T_c^- of the surface region even in the same single crystal, since the transition temperature of UPt$_3$ is quite sensitive to the stress or the defects. It should be noted that the difference in T_c^- between $I \parallel c$ and $I \parallel b$ is small compared with the typical transition width 10 mK which is observed in a specific heat measurement [2,9] and is partly related to the inhomogeneity of the bulk sample.

As pointed out by Yip [11], the Josephson effect of a weak link between a conventional and an unconventional superconductor is characterized by (1) a current phase relationship with a period of $2\pi/n$ and (2) the critical current just below the transition temperature T_c of the lower transition temperature superconductor is proportional to $[1 - (T/T_c)]^{n/2}$, where n is an integer determined by the symmetry. The former current phase relationship can be determined experimentally by the observation of the Shapiro steps at multiples of $V = h\nu/2ne$. Figure 5 shows the typical Shapiro steps measured by passing the ac current, in addition to the dc current, directly through the junction [12]. Since $h\nu/2e$ is 4.14 pV at a frequency of 2 kHz, the result indicates that the parameter n for both current directions is 1. Conventional Shapiro steps ($n = 1$) are also observed for UPt$_3$-Nb junctions below T_c^-. They are observed only for $I \parallel b$ above T_c^-, since it is necessary that J_c is larger than about 0.03 A/cm^2 in order to observe definite Shapiro steps in our experiment, and J_c for $I \parallel c$ above T_c^- is always smaller than this value.

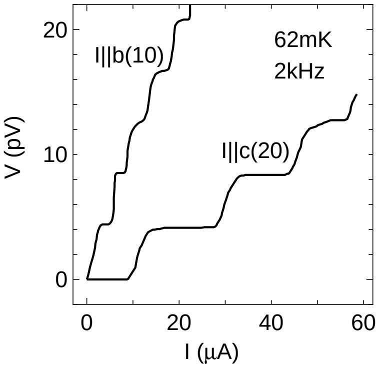


FIG. 5. Conventional Shapiro steps observed when an ac current with a frequency of 2 kHz is superposed on a dc current. Figures in parentheses refer to the thickness d_N (μm) of Cu.

The regular Shapiro steps also rule out the possibility of the proximity-induced Josephson effect (PIJE), since a spacing of the main Shapiro step is expected to be $h\nu/4e$ in the case of PIJE [13]. In addition, the result that $n = 1$ indicates that the Josephson effect occurs through the magnetically active interface if UPt$_3$ is an odd-parity (triplet) superconductor [14].

In the case that $n = 1$, J_c near T_c should be proportional to $[1 - (T/T_c)]^{1/2}$ according to the above prediction. The temperature dependence observed near T_c is contrary to expectation, as seen in Fig. 4. This discrepancy may be ascribed to the reduction of the condensation amplitude at the SN interface due to the proximity effect, which is not taken into account in Ref. [11].

Since the Josephson effect of a SNS$'$ junction between an s-wave superconductor and an unconventional superconductor has not been investigated theoretically, we shall, for the moment, apply the theory concerned with the Josephson effect between conventional superconductors. The critical supercurrent of SNS$'$ junction where S and S$'$ are conventional superconductors can be calculated by modifying that of the SNS junction derived by Clarke [15], as given by

$$J_c = A|F||F'|\frac{1}{\xi_N}e^{-d_N/\xi_N}, \tag{1}$$

where A is a constant and ξ_N is the coherence length in N given by $\xi_N = (\hbar v_F \ell_N / 6\pi k_B T)^{1/2}$; v_F and ℓ_N are the Fermi velocity and the electronic mean free path in N, respectively, and $|F|$ and $|F'|$ are the condensation amplitude at the SN and S$'$N interface, respectively. The reduction of the condensation amplitude at the SN interface due to the proximity effect is expressed as

$|F| \sim |F_S| b/\xi_S$ in which $|F_S|$ and ξ_S are the condensation amplitude deep inside S and the Ginzburg-Landau coherence length in S, respectively. The parameter b is often called the extrapolation length, given by

$$b = (\rho_N/\rho_S)\xi_N\,, \tag{2}$$

where ρ_N and ρ_S are the normal state resistivity of the normal and superconducting sides, respectively [16].

The result that $J_c(I \parallel c)$ is more than an order of magnitude larger than $J_c(I \parallel b)$ cannot be ascribed to the anisotropy of the reduction of the condensation amplitude at the UPt$_3$-Cu interface, even if we consider the proximity effect concerned with conventional superconductors. Taking the values $\rho_N = 0.085\ \mu\Omega \cdot \text{cm}$, $\rho_S = 0.24\ \mu\Omega \cdot \text{cm}$ $(I \parallel c)$ or $0.66\ \mu\Omega \cdot \text{cm}$ $(I \parallel b)$ [9], and $\xi_S = 12.5$ nm (c axis) or 11.3 nm (ab plane) [3] in Eq. (2), $b \gg \xi_S$ is satisfied at low temperatures and the reduction of the condensation amplitude due to the proximity effect is neglected at the UPt$_3$-Cu interface. On the other hand, it is obvious from Eq. (2) that the above reduction becomes remarkable and the anisotropy $J_c(I \parallel c) > J_c(I \parallel b)$ appears as the temperature approaches T_c and ξ_S increases. However, the anisotropy of J_c is estimated to be of the order of that of ρ_S and the observed large anisotropy cannot be explained.

Although the details of the Josephson effect and the proximity effect concerned with unconventional superconductors are not clear, J_c of the present junctions at higher temperatures is also expected to reflect the growth of the condensation amplitude and the reduction of it due to the proximity effect similar to conventional Josephson junctions. Since the above growth and reduction probably depend on temperature and current direction, the temperature dependence of the anisotropy ratio $J_c(I \parallel c)/J_c(I \parallel b)$ may be caused in the case of the UPt$_3$-Cu-Nb junctions with $d_N = 10\ \mu$m and the UPt$_3$-Nb junctions, as seen in Fig. 3. In addition, the proximity effect is generally sensitive to the combination of the two materials such as UPt$_3$/Cu or UPt$_3$/Nb, and the expression of J_c as a function of d_N may be different according to the current direction when unconventional superconductors are concerned. Then, it is possible that the anisotropy ratio depends on the type of junctions: SNS$'$ or SS$'$ and on d_N in the case of SNS$'$ junctions. Since very little is known about these points, the large variation of $J_c(I \parallel c)/J_c(I \parallel b)$ between the SS$'$ junctions and the SNS$'$ junctions with different d_N in Fig. 3 cannot be explained quantitatively thus far.

Since there are a number of effects which reduce the Josephson effect, such as the alloy layer or intermetallic compound at the interface, we cannot dismiss the possibility that the anisotropy $J_c(I \parallel c) > J_c(I \parallel b)$ does not relate to the unconventional superconductivity of UPt$_3$, even if the anisotropy of the temperature dependence of J_c in Fig. 4 tends to cause the anisotropy $J_c(I \parallel c) > J_c(I \parallel b)$ below T_c^-. In addition, the roughness of the interface causes the contribution of the current of which the direction is other than the preferred direction. Nevertheless, the clear contrast of the temperature dependence between $I \parallel c$ and $I \parallel b$ for the SS$'$ junctions in Fig. 4 indicates that the Josephson effect which reflects the double superconducting transition and the anisotropic superconductivity of UPt$_3$ is observed in our investigation.

The present results have not yet been explained in relation to the unconventional order parameter of UPt$_3$. For example, if the E_{1g} and E_{2u} models for the low temperature phase of UPt$_3$ are considered, the critical currents should vanish for $I \parallel c$ or $I \parallel b$ in the tunneling limit, in which the parameter $n = 1$ [17].

In conclusion, we have investigated both dc and ac Josephson effects between UPt$_3$ and an s-wave superconductor for different current directions and observed conventional Shapiro steps, the anisotropy of the J_c value and the anisotropic temperature dependence at about T_c^-. The latter anisotropy in which the increase in J_c with decreasing temperatures becomes fast below T_c^- for $I \parallel c$ and it becomes rather slow below T_c^- for $I \parallel b$ gives clear evidence that the Josephson effect relates to the anisotropic superconductivity of UPt$_3$.

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