Magnetic-field modulation of the Josephson effect between UPt_3 and a conventional superconductor

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Josephson critical current I_c between a single-crystal UPt_3 and Al has been measured for the junctions on the mirror-flat UPt_3 surface perpendicular to the b and c axes. A dominant maximum peak at zero magnetic field has been observed in the magnetic field dependence of I_c, suggesting that most of the Josephson currents that flow through various parts of the junction are in phase and the sum of them is observed when no magnetic field is applied. This result raises questions about the E_{2u} scenario for the odd-parity order parameter in UPt_3, in which the energy gap has nodes and the Josephson effect is forbidden for both the b- and the c-axis directions.

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I. INTRODUCTION

The heavy-fermion superconductor UPt_3 has been attracted much attention in recent years, since a wide variety of physical properties suggest its unconventional superconductivity. A power law behavior in the temperature dependence of specific heat,[1] NMR relaxation rate,[2] and ultrasound attenuation[3] suggests a gap function vanishing on the Fermi surface, which means that the pairing state has a lower spatial symmetry than a conventional s-wave state. A possibility of an odd-parity pairing state has been suggested by the NMR Knight shift[4] and the equilibrium magnetization measurements.[5] Furthermore, it possesses a complex field-temperature (H-T) phase diagram.[6] In order to explain these properties, many candidates are proposed for the odd-parity order parameter $\hat{\Delta}(\mathbf{k})$ of UPt_3, which is described by the **d** vector as $\hat{\Delta}(\mathbf{k})=i[\mathbf{d}(\mathbf{k})\cdot\boldsymbol{\sigma}]\sigma_y$, where σ_x, σ_y, and σ_z are Pauli matrices. Among them, the two-dimensional E_{2u} scenario[7] or the spin scenario,[8] is widely used for the understanding of the experimental results. It is still an open question as to what scenario explains the unconventional superconductivity in UPt_3 most successfully.

The Josephson effect between a conventional and an odd-parity superconductor gives direct information about the order parameter of the odd-parity superconductor. The ordinary Josephson effect caused by the usual (second-order) processes is forbidden if the tunneling Hamiltonian does not change the spin,[9] and it can occur along **n** direction due to the spin-orbit coupling in the odd-parity superconductor when $[(\mathbf{n}\times\mathbf{k})\cdot\mathbf{d}(\mathbf{k})]\neq 0$, where **n** and **k** are the surface normal vector and the momentum of quasiparticles, respectively.[10] The observation of the Josephson effect along a certain direction of an odd-parity superconductor implies that the **d** vector has a component perpendicular to that direction.

In our previous paper, we have reported the Josephson effect between UPt_3 and Nb for current flow parallel and perpendicular to the c axis.[11] This result contradicts the candidates with the **d** vector along the c axis. However, we cannot exclude the possibility that the roughness of the UPt_3 surface causes a contribution of the current, of which direction is other than the nominal one. Actually, the magnetic field dependence of I_c, which should be a Fraunhofer diffraction pattern when the Josephson current flows uniformly through the junction, is found to be an oscillating function with no definite period, suggesting that the Josephson coupling of the junction is inhomogeneous for some reason. In this paper, an attempt to improve the quality of the junction is described and the possibility of the existence of the Josephson effect in particular directions of UPt_3 is discussed.

II. EXPERIMENTAL

The single crystals of UPt_3 have been grown by the Czochralski pulling-method in a tetra-arc furnace. They had already been used in our previous investigations.[11] A clear double superconducting transition was observed in a specific heat measurement.[12] The residual resistivity ratio was above 500, which indicates the sample quality is sufficiently good. 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. 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 $I\parallel c$, based on the assumption that the preferred current direction is perpendicular to the surface.

In order to eliminate the roughness of the surface, the sample was polished with diamond polish down to a grain size of 1 μm, resulting in a flat mirrorlike surface. After the sample was set in a sputtering apparatus, the surface was rf sputter-etched by Ar ion and then Cu/Zn alloy (brass), SiO_2, and Al were deposited by rf sputtering technique, as shown in Fig. 1(a). The difference between the previous and the present junctions is the use of Cu/Zn alloy and Al instead of Cu and Nb, respectively.[11] The purpose of these changes are discussed below.

The sample was set in a small solenoid coil, which was linked to the mixing chamber of a dilution refrigerator and

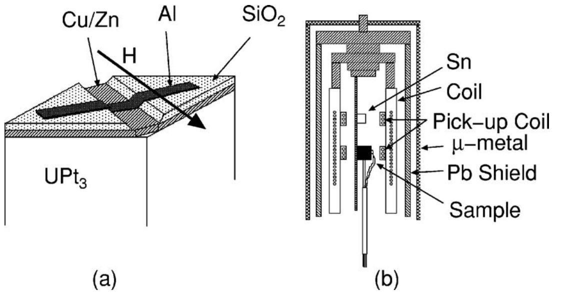

FIG. 1. (a) Sample arrangement of UPt_3-Cu/Zn-Al junctions. The arrow indicates the direction of an applied magnetic field. (b) Schematic of the experimental setup.

cooled down to 70 mK, as shown in Fig. 1(b). The current leads were attached to one end of the Al strip and UPt_3, and the voltage leads to the other end of the Al strip and UPt_3. The dc voltage was measured using a SQUID (superconducting quantum interference device) voltmeter, which is constructed with a series combination of a standard resistor (1.85 $\mu\Omega$) and an inductance coupled to the SQUID. The voltage resolution was about 10^{-12} V.

The earth magnetic field in the sample region was reduced to 10^{-4} Gauss by a three-layered μ-metal shield. This value is more than an order of magnitude smaller than that in the previous work in which a single μ-metal shield was used, and is critical to obtain reproducible data on the magnetic field dependence of I_c. In order to measure the residual magnetic field, a pair of pickup coils was wound around the sample and a standard superconductor (Sn), and the dc susceptibility was measured by means of a flux transformer method using another SQUID detector. The external magnetic field at which the susceptibility change at T_c of Sn vanishes was determined and defined as the residual-field component H_{res} along the coil axis.

III. RESULTS AND DISCUSSION

We show in Fig. 2 the typical temperature dependence of the junction resistance R and the Josephson critical current I_c for the junction with $I \parallel b$, where R is measured by flowing a direct current of 10 μA. Below the critical temperature of Al, R is the sum of the resistance of Al-Cu/Zn boundary, Cu/Zn, Cu/Zn-UPt_3 boundary, and UPt_3. As the temperature is lowered, a decrease in R due to the superconducting transition of UPt_3 is observed at 0.54 K, followed by a slow decrease, then R becomes zero due to the Josephson effect between Al and UPt_3. In the inset, representative current-voltage characteristics indicate that a continuous rise in voltage is seen above a critical value I_c and no hysteresis is observed, which is typical of SNS′ junctions. As the temperature is lowered, the Josephson critical current I_c increases rapidly.

Figure 3 shows the temperature dependence of Josephson critical current density J_c for the UPt_3-Cu/Zn-Al (UCA) and the previous UPt_3-Cu-Nb (UCN) junctions, where J_c is defined simply as $J_c = I_c / S$. Although the UCA junctions use a dirty normal metal (Cu/Zn) and a conventional superconductor with lower T_c, J_c is not so small as compared with the

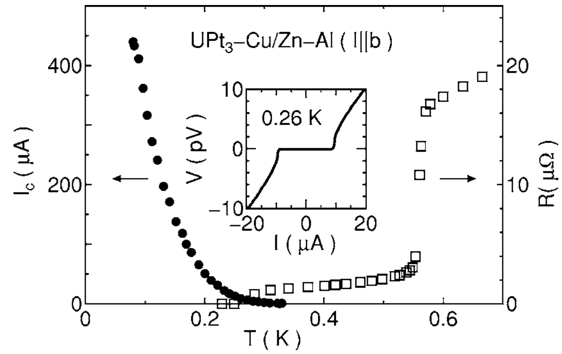

FIG. 2. Typical temperature dependence of junction resistance R and Josephson critical current I_c, where $I \parallel b$ and the current used for R measurement was 10 μA. Inset: the *I*-*V* characteristic showing the Josephson critical current.

previous one. This result at least indicates that the polishing process in the present research does not suppress the Josephson effect. The anisotropy $J_c(I \parallel c) > J_c(I \parallel b)$ observed in the UCA junctions is less pronounced than that in the previous work.[11] Even if this anisotropy is reproducable, we cannot relate it to the unconventional superconductivity of UPt_3, since a number of extrinsic effects, such as the surface conditions of UPt_3, affect the magnitude of J_c.

The quality of the junctions is demonstrated in the magnetic field dependence of I_c in Fig. 4. If a magnetic 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 three junctions, estimated λ_J is larger than the width of the junction when I_c does not exceed 200 μA. Comparing with the UCN junction, the *H*-I_c characteristics of the UCA junctions approach a Fraunhofer diffraction pattern in the sense that a global maximum of I_c at $H=0$ is dominant in comparison to the other local maximums.

The improvement of *H*-I_c curve in the present work is achieved by two methods: one is to reduce the spatial variation of the Josephson coupling and the other to eliminate the effects of flux trapping. The former is realized by obtaining a flat surface. It eliminates not only the contribution of the current of which direction is other than the nominal one, but also the spatial variation of the thickness of the N layer covering the undulating surface. The use of a "dirty" normal

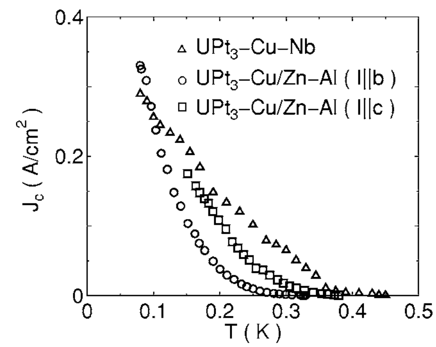

FIG. 3. Temperature dependence of Josephson critical current density J_c for three junctions.

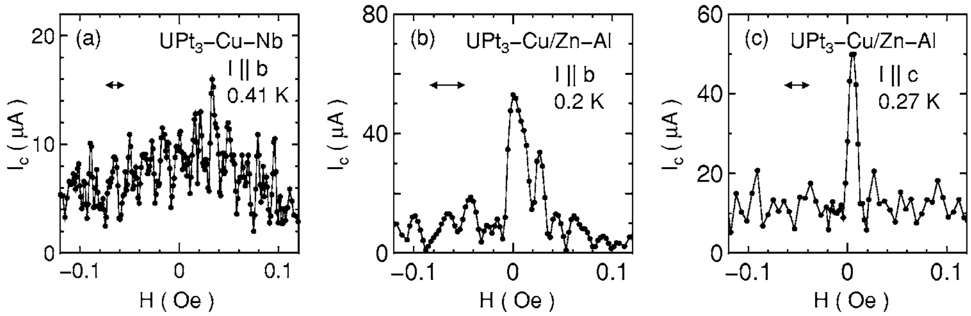

FIG. 4. Magnetic field dependence of Josephson critical current I_c for three junctions. The solid line through the data points is a guide to the eye. The arrows indicate ΔH in the text.

layer Cu/Zn makes the current flow insensitive to the SN-interface conditions such as atomic migrations or oxidation.[13]

In order to reduce trapped flux, a type I superconductor Al is used instead of Nb as a counter electrode. However, this is obviously insufficient, since most of magnetic vortices are trapped in UPt_3, as shown in Fig. 5(a). The pickup coil in Fig. 1 was used to measure the dc magnetic susceptibility of UPt_3. The sample was first cooled below T_c in zero field, then a dc magnetic field was applied and the diamagnetism (shielding effect) was observed. The sample was then warmed to temperatures above T_c to measure the change in the diamagnetic susceptibility (zero-field cooled susceptibility). In the same field, the sample was cooled again to observe the Meissner effect (field-cooled susceptibility). In contrast to the full diamagnetism, the Meissner effect, which is measured by the field-cooled method, is as low as 3%, indicating that most of the magnetic flux threading of the sample is trapped below T_c. Consequently, it is of critical importance to decrease the residual field as low as possible, as demonstrated in Fig. 5(b).

Before measuring the magnetic field dependence of I_c at certain field intervals, the continuous but qualitative variation of I_c has been always measured to avoid missing a fine structure of H-I_c curve: A current, which was a little larger than the maximum of I_c, was passed through the junction, and the voltage was recorded continuously as a function of magnetic field. Typical results are shown in Fig. 5(b); the true I_c variation corresponding to curve A is already given in Fig. 4(c). Curves B and C are the data taken when the two-layered shield was used and H_{res} was 0.9 mG; after curve B was taken, the sample was warmed up above T_c, then cooled down to the same temperature again, and curve C was recorded. When the sample was cooled down below T_c, the field component along the coil axis was always reduced below the sensitivity limit (~0.05 mG) by flowing a compensational current through the solenoid. Nevertheless, extra peaks and troughs are observed in curves B and C, and a change in structure is seen between curve B and curve C. This result suggests that magnetic shielding is crucial to obtain reliable results, because the remaining magnetic field perpendicular to the coil axis affects the data.

If we refer to a coordinate system with the z axis along the current direction and apply an external magnetic field H in the y direction, the total current I in the junction is given by

$$I = \int\int \mathrm{d}x\mathrm{d}y J_c(x,y)\sin\left(\frac{2\pi d}{\Phi_0}Hx + \psi_0\right), \qquad (1)$$

where $J_c(x,y)$ is the local Josephson critical current density, Φ_0 is the flux quantum, and ψ_0 is the phase difference between two superconductors; d is given by $d=d_N+\lambda_S+\lambda_{S'}$, where λ_S and $\lambda_{S'}$ are the penetration depths of the superconductors S and S′, respectively.[14] This equation indicates that I has a global maximum value $\int\int \mathrm{d}x\mathrm{d}y J_c(x,y)$ at zero magnetic field, even if $J_c(x,y)$ is not a constant; I becomes small when the external magnetic field is applied and the Joseph-

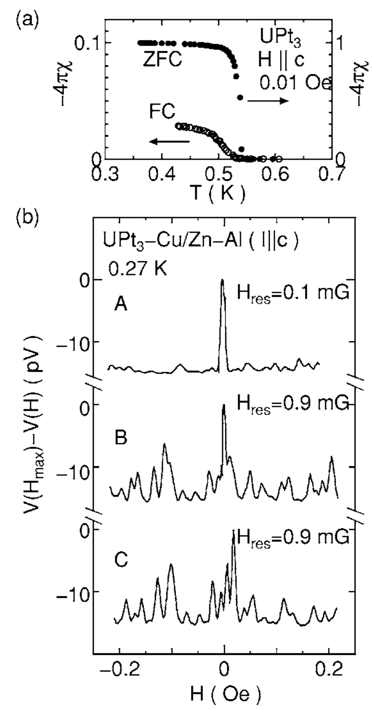

FIG. 5. (a) Temperature dependence of dc magnetic susceptibility of UPt_3 obtained by the field cooled (FC) and zero-field cooled (ZFC) methods. (b) $V(H_{max})$-$V(H)$ as a function of H, where $V(H)$ is the voltage measured by passing a constant current slightly larger than the maximum value of $I_c(H)$ observed at $H=H_{max}$, reflecting the modulation of $I_c(H)$ qualitatively. After curve B was taken, the sample was warmed up above T_c and cooled down again to measure curve C. The residual-field component H_{res} along the coil axis in Fig. 1 was always reduced below the sensitivity limit (~0.05 mOe) by flowing a compensational current through the solenoid when the sample was cooled down below T_c.

son currents flowing through various parts cancel out. However, if the spatial variation of the phase difference $\tilde{\psi}(x,y)$ exists ($\psi_0 \rightarrow \psi_0 + \tilde{\psi}(x,y)$), the peak at zero magnetic field is expected to become less dominant. One of the origins of such a variation is magnetic flux trapping that generates local magnetic fields. Another is the anisotropic order parameter of UPt_3, of which phase depends on directions, the fluctuation of local current directions caused by the roughness of UPt_3 surface leads to the spatially varying $\tilde{\psi}(x,y)$.

If the Josephson effect between UPt_3 and an *s*-wave superconductor is allowed for the *c*- and the *b*-axis directions, the present result in Fig. 4 that the magnetic field dependence of I_c is not an ideal Fraunhofer pattern. Yet a global maximum of I_c at $H=0$ is dominant in comparison to the other local maximums; it may be explained by the spatial variation of $J_c(x,y)$ in Eq. (1). Then, the selection rule $[(\mathbf{n} \times \mathbf{k}) \cdot \mathbf{d}(\mathbf{k})] \neq 0$ leads to the result that the **d** vector has components perpendicular to the *c*- and the *b*-axis directions. The spin scenario, in which two-component **d** vector is assumed, does not contradict with the present result.[15]

Even if the Josephson effect in the direction perpendicular to the surface is forbidden and only the Josephson currents that flow in other directions through some narrow paths at the interface are observed, it is possible that the present H-I_c curve is observed in a certain case, as demonstrated in Fig. 6. In order to simplify the calculation, the Josephson currents flowing due to the roughness of the surface are regarded as those flowing through the narrow windows in a flat insulating interface, as shown in Fig. 6(a). All the windows are assumed to have the same width (10^{-3} of the junction width), the same critical current i_c, and to be distributed at random. The $I_c(H)$ curves are calculated for both the case in which the phase difference at each window is the same and the case in which it varies randomly by π [ψ_0 or $\psi_0+\pi$ in Eq. (1)]; the latter occurs when the anisotropic order parameter changes the relative sign according to the direction.

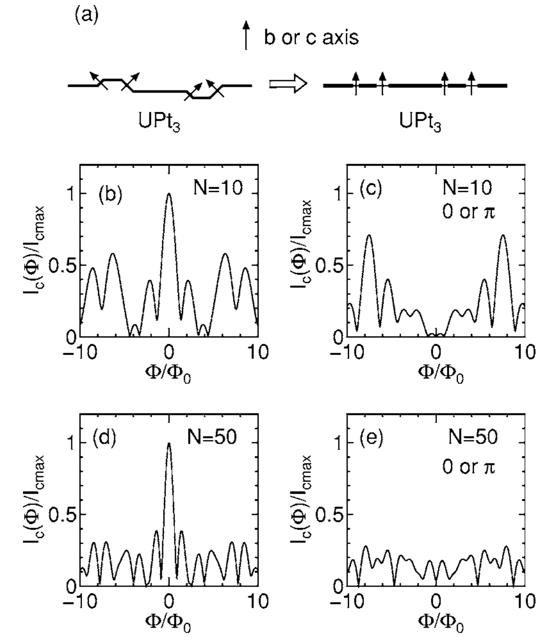

FIG. 6. (a) Schematic of the relationship between the simulation model and the case in which the Josephson effect in the direction perpendicular to the nominal surface is prohibited and yet I_c is observed by the roughness. (b)–(e) Examples of the calculated magnetic field dependence of I_c for junctions containing N windows which have the same critical current i_c and the same width (10^{-3} of the junction width), where $I_{c\,max}=Ni_c$ and $\mathbf{\Phi}_0$ and $\mathbf{\Phi}$ are the flux quantum and the magnetic flux through the junction, respectively. The distributions of the windows in (c) and (e) are the same as those in (b) and (d), respectively. The phase difference of 0 or π is added at random for each window in (c) and (e).

Figures 6(b)–6(d) show the examples of the resultant $I_c(H)$ curves. Although the patterns depend on the distribution of the windows, two characteristic features are observed: i. The central peak becomes dominant over other peaks, when the phase difference at each window is the same and the number of windows is large because the contribution of each window in Eq. (1) tend to cancel each other at finite magnetic fields. ii. Oscillating curves are always observed, when the phase difference at each window varies randomly by π; $I_c(H)$ shows either a peak or a dip at $H=0$ and does not show a global maximum. It should be noted that similar results have been reported for the faceting (110) face of $YBa_2Cu_3O_{7-x}$ superconductor, where the faceting causes a variation of the relative sign of the *d*-wave order parameter.[16]

These calculations suggest that it is difficult to explain the $I_c(H)$ curves in Fig. 4(b) and Fig. 4(c) by assuming the two-dimensional E_{2u} scenario $\mathbf{d}(\mathbf{k}) \sim (k_x+ik_y)^2 k_z \hat{z}$ for the order parameter in UPt_3, since it prohibits the Josephson effect for the *b*- and the *c*-axis directions.[17] The phase of the order parameter varies sensitively with the deviation from both directions; the order parameter acquires a phase change of $2\pi/3$ under a $\pi/3$ rotation about the *c* axis, and a change of sign under reflection in the basal plane. Even if the junctions on the surface perpendicular the *b* or *c* axis may show the Josephson effect by the currents in the directions other than both axes, those currents should not be in phase at $H=0$ as long as their directions are at random.

For a rectangular SNS′ junction in Fig. 1, the magnetic flux through the junction is expressed as $\mathbf{\Phi}=Hw(d_N+\lambda_{UPt_3}+\lambda_{Al}$ or $\lambda_{Nb})$, where w is the width of the junction perpendicular to the field direction. Considering that λ_{Al} and λ_{Nb} are much smaller than $d_N=0.8\ \mu m$ or $\lambda_{UPt_3} \sim 0.7\ \mu m$,[18] λ_{Al} and λ_{Nb} are neglected, and the oscillation period ΔH corresponding to one flux quantum $\mathbf{\Phi}_0$ threading the junction is estimated. The ΔH values are listed in Table I and indicated by arrows in Fig. 4. Although I_c does not seem to oscillate with a single period in three junctions, the result $\Delta H_{UCN} < \Delta H_{UCA(I\parallel c)} < \Delta H_{UCA(I\parallel b)}$ agrees with the periodicity in Fig. 4, at least qualitatively. The estimated ΔH in such junctions is supposed to correspond to the shortest period in the $I_c(H)$ structure. If we take the central peak as a representative one, the H interval between the global maximum and the nearest local minimum of I_c is 0.015 Oe ($I\parallel b$) and 0.012 Oe ($I\parallel c$), which is less than half of the estimated value for each junction. This discrepancy may be ascribed to flux focusing by the superconducting crystal and film; the magnetic field H_j at the junction is larger than the applied field H_{ext}. Although it is difficult to estimate H_j, it should be of the order of the field at the surface of the cube-shaped superconductor and com-

TABLE I. Properties of SNS′ junctions. The junction area S is expressed as $S=w*t$, where w and t are the width between the SiO_2 banks and the width of the Al strip, respectively. The three junctions have the same thickness d_N of N(normal metal) layer.

Substrate	S′	N	d_N	S(mm^2)	ΔH(Oe)
$I\|b$(unpolished)[a]	Nb	Cu	0.8	0.70*0.27	0.019
$I\|b$(polished)	Al	Cu/Zn	0.8	0.38*0.35	0.035
$I\|c$(polished)	Al	Cu/Zn	0.8	0.54*0.36	0.025

[a]Reference 11.

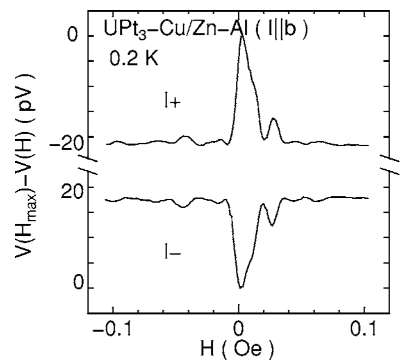

FIG. 7. $V(H_{max})$-$V(H)$ as a function of H, where $V(H)$ is the voltage measured by passing a positive or negative constant current of which absolute value is slightly larger than the maximum of $|I_c(H)|$ observed at $H=H_{max}$, showing the modulation of $I_c(H)$ qualitatively.

parable to the value $3H_{ext}/2$ of the sphere in the equatorial plane.

In Fig. 4, asymmetry between the positive and negative field directions is observed. If this behavior is intrinsic, it means that time-reversal symmetry is broken in UPt_3. Figure 7 shows the qualitative variation of positive and negative critical current as a function of magnetic field, which was recorded in the same way as in Fig. 5(b). The true I_c variation corresponding to $I+$ is already given in Fig. 4(b). Although a small shift of the field value at peaks and troughs is seen, both curves exhibit almost the same asymmetry with respect to the field direction. This result indicates that the asymmetry is not caused by self-field effects, which depend on the relation between the field and the measuring current directions. Since reversing both the current and the field direction corresponds to reversing time, the asymmetry in Fig. 7 may suggest that time-reversal symmetry is broken in UPt_3. However, we cannot exclude the possibility that the trapped vortices, even if the amount of them are reduced, cause such a behavior. It should be noted that dozens of magnetic vortices are still trapped inside the present UPt_3 crystal when the residual field (~0.1 mG) and the cross section (~0.1 cm^2) are taken into account.

IV. CONCLUSION

In conclusion, we have refined the investigation of the Josephson critical current I_c between UPt_3 and a conventional superconductor by fabricating the junction on the mirror-flat surface of a UPt_3 crystal and eliminating the residual earth magnetic field. The magnetic field dependence of I_c has shown that a global maximum at zero magnetic field is dominant in comparison with other peaks. This result does not contradict the spin scenario for the odd-parity order parameter in UPt_3, while it is difficult to explain by the two-dimensional E_{2u} scenario in which the Josephson couplings along the b and c axes are forbidden.

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