

## Observation of the Josephson effect between $\text{UBe}_{13}$ and an $s$ -wave superconductor

S. Shibata, A. Sumiyama, and Y. Oda

*Department of Material Science, Faculty of Science, Himeji Institute of Technology, Akō-gun 678-1297, Japan*

Y. Haga

*Advanced Science Research Center, Japan Atomic Energy Research Institute, Tokai 319-1106, Japan*

Y. Ōnuki

*Advanced Science Research Center, Japan Atomic Energy Research Institute, Tokai 319-1106, Japan  
and Graduate School of Science, Osaka University, Toyonaka 560-0043, Japan*

(Received 17 February 1999)

Josephson critical current densities  $J_c$  between  $\text{UBe}_{13}$  and an  $s$ -wave superconductor have been investigated for  $\text{UBe}_{13}$ -Cu-Nb and  $\text{UBe}_{13}$ -Nb junctions. The result that  $J_c$  increases with decreasing temperatures below  $T_c$  of  $\text{UBe}_{13}$ , together with conventional Shapiro steps, indicates that we have observed the Josephson effect related to the bulk superconductivity of  $\text{UBe}_{13}$ , and not the so-called proximity-induced Josephson effect, which was reported for the point contact between  $\text{UBe}_{13}$  and Ta. [S0163-1829(99)09929-4]

Since the discovery of heavy-fermion superconductivity in  $\text{CeCu}_2\text{Si}_2$  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,  $\text{UBe}_{13}$  is thought to be unconventional, because the temperature dependence of various physical properties, such as specific heat,<sup>1</sup> follows power laws at low temperatures, suggesting a gap function vanishing on the Fermi surface. In addition, a nonmonotonic decrease in  $T_c$  in the thoriated system  $\text{U}_{1-x}\text{Th}_x\text{Be}_{13}$ , together with an additional phase transition in a limited Th concentration range, has received considerable interest in recent years.<sup>2,3</sup>

The Josephson effect between  $\text{UBe}_{13}$  and a conventional superconductor has been investigated for a point contact between  $\text{UBe}_{13}$  and Ta by Han *et al.* and a significant result has been reported.<sup>4</sup> They explained the results in terms of a proximity-induced Josephson effect (PIJE), in which Ta induces an  $s$ -wave order parameter in  $\text{UBe}_{13}$  near the contact and a Josephson coupling between the induced and the Ta-superconducting state is observed. As the temperature is decreased, the critical current due to PIJE increases above  $T_c$  of  $\text{UBe}_{13}$ , reaches a maximum at  $T_c$ , and then decreases below  $T_c$ , indicating that the occurrence of the  $\text{UBe}_{13}$  order parameter suppresses the  $s$ -wave order parameter induced in  $\text{UBe}_{13}$ . This negative  $s$ -wave proximity effect has been related to the triplet order parameter of  $\text{UBe}_{13}$ . However, the interpretation in terms of PIJE has been questioned.<sup>5,6</sup> Neither PIJE nor the conventional Josephson effect has been observed in another investigation for point contacts between  $\text{UBe}_{13}$  and various superconductors.<sup>7</sup> It will be useful to fabricate another type of Josephson junction, for example, a superconductor-normal-metal-superconductor (SNS') junction in which two superconductors S and S' are separated by a normal metal N, and to investigate whether the Josephson effect exists or not. Recently, we have reported that the SNS' and SS' junctions between HFS( $\text{CeCu}_2\text{Si}_2$ ,  $\text{UPd}_2\text{Al}_3$ ,  $\text{UPt}_3$ ) and Nb also show the Josephson effect.<sup>8,9</sup> In

this paper, the distinct Josephson effect of  $\text{UBe}_{13}$ -Cu-Nb junctions (SNS' junctions) and the  $\text{UBe}_{13}$ -Nb junction (SS' junction) is described and is compared with the previous results for the point-contact junctions.

The polycrystalline sample of  $\text{UBe}_{13}$  was synthesized by arc-melting U and Be in an argon-gas atmosphere. The critical temperature  $T_c$  determined in an ac susceptibility measurement was 0.94 K, which indicates that the sample is "H-type"  $\text{UBe}_{13}$ .<sup>3</sup> A piece of the sample was cut to the cubic shape with edges of about 3 mm to use as a substrate. The surface was not polished in consideration of a report that Josephson effects have not been observed when polished surfaces of HFS are used.<sup>10</sup> Scanning electron microscope images show that the sample has an undulating surface with about 10  $\mu\text{m}$  in height and several tens of  $\mu\text{m}$  in length. The sample surface was rf sputter etched by Ar ion and then Cu (normal metal),  $\text{SiO}_2$  and Nb ( $s$ -wave superconductor) were deposited by 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' junction was fabricated in the same way without the Cu layer. The current leads were attached to one end of the Nb strip and  $\text{UBe}_{13}$  and the voltage leads to the other end of the Nb strip and  $\text{UBe}_{13}$ . The electronic mean free path  $l_N$  in Cu at 4.2 K was 0.6  $\mu\text{m}$  ( $d_N=10 \mu\text{m}$ ) and 0.2  $\mu\text{m}$  ( $d_N=0.8 \mu\text{m}$ ), which was calculated from the resistivity of the adjacent Cu strip deposited at the same time.

The dc voltage was measured using a superconducting quantum interference device (SQUID) voltmeter which is

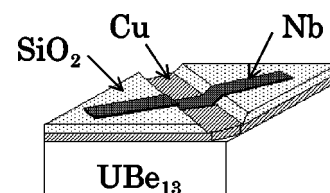


FIG. 1. Sample arrangement of  $\text{UBe}_{13}$ -Cu-Nb junctions. The  $\text{UBe}_{13}$ -Nb junction has the similar structure without the Cu layer.

TABLE I. Properties of samples. The sample without the  $d_N$  value is the  $\text{UBe}_{13}$ -Nb junction.

$d_N(\mu\text{m})$	$S(\text{mm}^2)$	$R_0(\mu\Omega)$	$\rho_b(\text{n}\Omega \text{ cm}^2)$
	0.29	120	330
0.8	0.19	24	45
10	0.14	11	15

constructed with a series combination of a 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  $\mu$ -metal shield.

We show in Fig. 2 the typical properties of the Josephson effect between  $\text{UBe}_{13}$  and Nb. A continuous rise in voltage is observed in the current-voltage characteristics, as the current is increased from the critical value  $I_c$ . The  $I$ - $V$  curve is single valued and not hysteretic, which is typical to SNS' junctions.

If a magnetic field is applied to a uniform junction whose width normal to the field is smaller than the Josephson penetration depth  $\lambda_J$ , a Fraunhofer diffraction pattern should be observed. In the present junctions, estimated  $\lambda_J$  becomes smaller than the width of the junction when  $I_c$  exceeds around  $500 \mu\text{A}$ . Although an interference pattern is seen in Fig. 2,  $I_c$  oscillates with no definite period. This pattern suggests that the junction is not uniform, that is, the local critical supercurrent density fluctuates spatially. One of the reasons for this fluctuation may be the roughness of the sample surface described above.

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 when  $\text{UBe}_{13}$  and Nb are separated by Cu. The Josephson current of the sample with  $d_N = 10 \mu\text{m}$  was too small to be observed in our experiment.

The Josephson current and the junction resistance  $R$  at about the critical temperature  $T_c$  of  $\text{UBe}_{13}$  are shown in Fig. 4. Below the critical temperature of Nb, the junction resis-

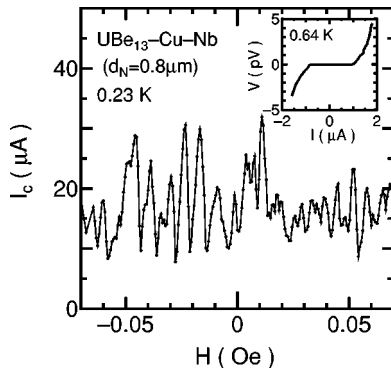


FIG. 2. Magnetic-field dependence of  $I_c$  suggesting that the junction is not uniform. The solid line through the data points is a guide to the eye. Inset:  $I$ - $V$  characteristic showing Josephson critical current.

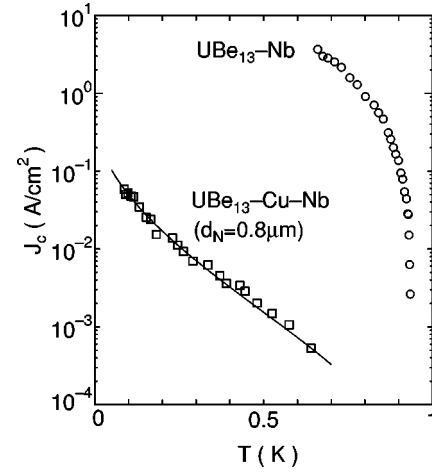


FIG. 3. Temperature dependence of  $J_c$  for  $\text{UBe}_{13}$ -Nb junction and  $\text{UBe}_{13}$ -Cu-Nb junction where the thickness  $d_N$  of Cu is  $0.8 \mu\text{m}$ . In the case of  $d_N = 10 \mu\text{m}$ ,  $J_c$  was not observed within experimental accuracy. The solid line indicates a least-squares fit to the data using the theory in the text.

tance  $R$  of the SS' junction consists of the resistance of  $\text{UBe}_{13}$  and the boundary resistance between  $\text{UBe}_{13}$  and Nb. A sharp decrease due to the vanishment of the former one is observed at  $T_c \sim 0.94$  K of  $\text{UBe}_{13}$ . We define the junction resistance just below  $T_c$  of  $\text{UBe}_{13}$  as the Josephson-junction resistance  $R_0$ , as listed in Table I. The boundary resistivity defined as  $\rho_b = (R_0 - R_{\text{Cu}}) * S$  for SNS' junctions and  $\rho_b = R_0 * S$  for SS' junction, where  $R_{\text{Cu}}$  is the calculated resistance of Cu, is also tabulated. As the temperature is lowered, measurable supercurrent appears and the Josephson critical current increases, as seen in Fig. 4. The result that the supercurrent is observed only below the superconducting transition of  $\text{UBe}_{13}$  indicates that we have observed the Josephson effect between the bulk  $\text{UBe}_{13}$  and Nb rather than the proximity-induced Josephson effect (PIJE).<sup>4</sup>

The possibility of PIJE is also ruled out from the Shapiro steps, as shown in Fig. 5. The Shapiro steps were observed by passing the ac current, in addition to the dc current, directly through the junction.<sup>11</sup> When the ac current is superposed, rf interference increases, so that only two indistinct steps can be observed. Since  $h\nu/2e$  is 207 pV at a frequency of 100 kHz, the observed steps are the conventional ones. Fractional steps have not been observed within experimental accuracy thus far. On the contrary, a spacing of the main Shapiro step is expected to be  $h\nu/4e$  in the case of PIJE.<sup>6</sup>

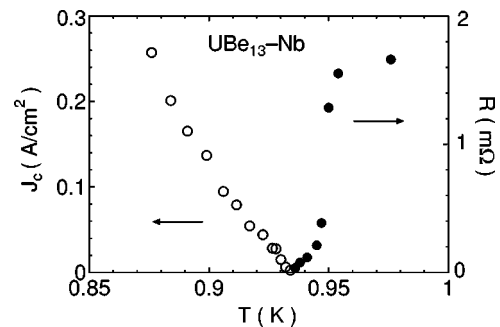


FIG. 4. Temperature dependence of  $J_c$  and junction resistance  $R$  at about the superconducting transition of  $\text{UBe}_{13}$ .

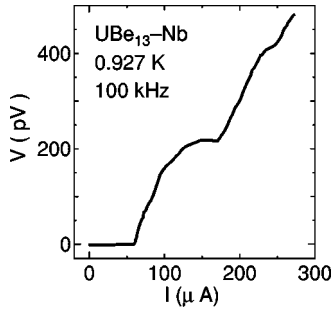


FIG. 5. Conventional Shapiro steps observed when ac current with a frequency of 100 kHz is superposed on dc current.

Since only conventional shapiro steps were observed in the point contact between  $\text{UBe}_{13}$  and Ta by Han *et al.*,<sup>4</sup> their interpretation in terms of PIJE has been questioned.<sup>6</sup>

The result that PIJE did not appear in our investigation may be attributed to the properties of an interface which depend on fabricating processes of the interface and affect the proximity effect remarkably. For example, Au shows the Meissner effect due to the proximity effect in Au-clad Nb wire, and not in two-layer systems such as Sn/Au, although the latter obviously has a smoother interface than the former.<sup>12</sup> Since PIJE is smaller than the conventional Josephson effect, an extremely clean interface which is not only smooth but also without an alloy layer or intermetallic compound may be needed.

The result that the Josephson effect with conventional Shapiro steps exists between  $\text{UBe}_{13}$  and Nb does not exclude the possibility that  $\text{UBe}_{13}$  is a triplet superconductor, since the ordinary pair tunneling of the Cooper pairs between triplet and singlet superconductors can occur due to the spin-orbit coupling.<sup>13</sup> In addition, the largest  $J_c$  value which can be measured is limited because of excessive heating, so that it is impossible to compare  $I_c R_0$  products with the ideal value between conventional superconductors derived by Ambegaokar and Baratoff.<sup>14</sup> Considering also that anisotropic Josephson effects cannot be observed in our investigation because of using polycrystalline  $\text{UBe}_{13}$ , we cannot derive distinct information about the unconventional superconductivity of  $\text{UBe}_{13}$  from the present results.

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,<sup>15</sup> as given by

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

where  $A$  is a constant and  $\xi_N$  is the coherence length in N given by  $\xi_N = (\hbar v_F l_N / 6 \pi k_B T)^{1/2}$ ;  $v_F$  and  $l_N$  being the Fermi velocity and the electronic mean free path in N, respectively;  $|F|$  and  $|F'|$  are the condensation amplitude at the SN and S'N interface, respectively. Sufficiently lower than  $T_{c'}$ ,

where the critical temperature  $T_c$  of S is lower than  $T_{c'}$  of S',  $|F'|$  can be treated as a constant and  $J_c$  should vary as  $|F| \exp(-d_N/\xi_N)/\xi_N$  or  $(|F_S|b/\xi_S) \exp(-d_N/\xi_N)/\xi_N$  in which  $|F_S|$  and  $\xi_S$  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

$$b = (\rho_N/\rho_S) \xi_N, \quad (2)$$

where  $\rho_N$  and  $\rho_S$  are the normal-state resistivity of the normal and superconducting sides, respectively.<sup>16</sup> Since  $|F_S|$  and  $\xi_S^{-1}$  are proportional to  $(1 - T/T_c)^{1/2}$ ,  $J_c$  is expected to vary with the temperature according to

$$J_c = A (1 - T/T_c) \exp(-B d_N \sqrt{T}), \quad (3)$$

where  $A$  and  $B$  are constants.

The solid line in Fig. 3 indicates a least-squares fit to the data for the SNS' junction using Eq. (3). Although the fit for this junction is good, the parameter  $B \sim 9.0 (\mu\text{m} \sqrt{\text{K}})^{-1}$  is rather larger than  $B \sim 3.1 (\mu\text{m} \sqrt{\text{K}})^{-1}$  estimated from the dirty limit expression  $B = (6 \pi k_B / \hbar v_F l_N)^{1/2}$  using  $l_N$  calculated from the resistivity of the Cu film. This may suggest that Eq. (3) is oversimplified in the case of the SNS' junction with a thin normal layer, since both  $B$  values agree well in the case of the SNS' junction with a thick Cu layer ( $d_N > 20 \mu\text{m}$ ) in which the temperature dependence of  $J_c$  is determined mostly by the exponential term.<sup>8</sup> Nevertheless, if we estimate  $J_c$  in the case of  $d_N = 10 \mu\text{m}$  by applying Eq. (3) with the  $A$  and  $B$  values obtained in the case of  $d_N = 0.8 \mu\text{m}$ ,  $J_c$  is still smaller than  $10^{-6} \text{ A/cm}^2$  even at 30 mK, providing some explanation why the Josephson effect of the SNS' junction with  $d_N = 10 \mu\text{m}$  cannot be observed within experimental accuracy.

It is still an open question whether the remarkable reduction of the Josephson effect for SNS' junctions as compared with the SS' junction or other SNS' junctions of which heavy-fermion superconductor is  $\text{CeCu}_2\text{Si}_2$ ,  $\text{UPd}_2\text{Al}_3$  or  $\text{UPt}_3$ ,<sup>8,9</sup> relates to the unconventional superconductivity of  $\text{UBe}_{13}$ , since there are a number of reasons for small  $J_c$  even between conventional superconductors. It should be noted that the large  $\rho_S \sim 100 \mu\Omega \text{ cm}$  of  $\text{UBe}_{13}$  as compared with  $\rho_N \sim 0.3 \mu\Omega \text{ cm}$  tends to reduce  $b$  in Eq. (2) and suppresses the Josephson effect.

In conclusion, we have investigated both dc and ac Josephson effects between  $\text{UBe}_{13}$  and Nb, and observed conventional Shapiro steps and the increase in  $J_c$  with decreasing temperatures below  $T_c$  of  $\text{UBe}_{13}$ . These results indicate that we have observed the Josephson effect related to the bulk superconductivity of  $\text{UBe}_{13}$ , and not the proximity-induced Josephson effect, which was reported for the point contact between  $\text{UBe}_{13}$  and Ta.

This work was supported by the Scientific Research Grant-in-Aid from the Ministry of Education, Science, and Culture. Y.O. was supported financially by a Grant-in-Aid for COE Research (10CE2004) of the Ministry of Education, Science, Sports, and Culture.

- <sup>1</sup>H. R. Ott, H. Radigier, T. M. Rice, K. Ueda, Z. Fisk, and J. L. Smith, *Phys. Rev. Lett.* **52**, 1915 (1984).
- <sup>2</sup>H. R. Ott, H. Radigier, Z. Fisk, and J. L. Smith, *Phys. Rev. B* **31**, 1651 (1985).
- <sup>3</sup>F. Kromer, R. Helfrich, M. Lang, F. Steglich, C. Langhammer, A. Bach, T. Michels, J. S. Kim, and G. R. Stewart, *Phys. Rev. Lett.* **81**, 4476 (1998).
- <sup>4</sup>S. Han, K. W. Ng, E. L. Wolf, A. Millis, J. L. Smith, and Z. Fisk, *Phys. Rev. Lett.* **57**, 238 (1986).
- <sup>5</sup>A. M. Kadin and A. M. Goldman, *Phys. Rev. Lett.* **58**, 2275 (1987).
- <sup>6</sup>E. V. Thuneberg and V. Ambegaokar, *Phys. Rev. Lett.* **60**, 365 (1988).
- <sup>7</sup>O. E. Kvitnitskaya, A. Nowack, S. Wasser, Yu. G. Naidyuk, W. Schlabitz, and Z. Fisk, *Czech. J. Phys.* **46**, 799 (1996).
- <sup>8</sup>T. Koyama, A. Sumiyama, M. Nakagawa, and Y. Oda, *J. Phys. Soc. Jpn.* **67**, 1797 (1998).
- <sup>9</sup>A. Sumiyama, S. Shibata, Y. Oda, N. Kimura, E. Yamamoto, Y. Haga, and Y. Ōnuki, *Phys. Rev. Lett.* **81**, 5213 (1998).
- <sup>10</sup>S. Wasser, A. Nowack, W. Schlabitz, A. Freimuth, O. E. Kvitnitskaya, A. A. Menovsky, and C. Bruder, *Phys. Rev. Lett.* **81**, 898 (1998).
- <sup>11</sup>J. Clarke, *Phys. Rev. B* **4**, 2963 (1971).
- <sup>12</sup>A. Sumiyama, Y. Oda, and H. Nagano, *J. Phys. Soc. Jpn.* **53**, 2449 (1984).
- <sup>13</sup>V. B. Geshkenbein and A. I. Larkin, *Pis'ma Zh. Éksp. Teor. Fiz.* **43**, 306 (1986) [*JETP Lett.* **43**, 395 (1986)].
- <sup>14</sup>V. Ambegaokar and A. Baratoff, *Phys. Rev. Lett.* **10**, 486 (1963).
- <sup>15</sup>J. Clarke, *Proc. R. Soc. London, Ser. A* **308**, 447 (1969).
- <sup>16</sup>G. Deutscher and R. W. Simon, *J. Appl. Phys.* **69**, 4137 (1991).