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Authors

Wolf, EL
Han, Siyuan
Khim, ZG
et al.

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Proximity-Josephson effect (PJE) evidence for triplet pairing in UBe_{13} (invited)

E. L. Wolf
Polytechnic University, Brooklyn, New York 11201
Siyuan Han,[a] Z. G. Khim,[b] and K.-W. Ng[c]
Ames Laboratory, U. S. Department of Energy and Department of Physics, Iowa State University, Ames, Iowa 50011
A. J. Millis[d]
Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139
J. L. Smith and Z. Fisk
Los Alamos National Laboratory, Los Alamos, New Mexico 87545

The proximity-Josephson effect (PJE) is a powerful general method of determining the symmetry of the pair wave function in exotic superconductors. The method is simple and relatively insensitive to the surface condition of the sample. A superconducting probe (S) of known pairing symmetry (typically Nb or Ta) is brought into contact with the sample (N). Observations and arguments based on the de Gennes boundary condition at the NS interface both indicate formation of a local, proximity-induced superconducting region of depth ξ in the sample (N) under the probe. The induced pairs have the same symmetry as those of S. The expected pair-phase dependence of the coupling energy between the pairs in N and S leads to a Josephson current $I_c(T)$, which may be observed up to a junction critical temperature T^* which is typically ~ 0.8 of the T_c of S. When the measurement temperature falls below the bulk T_c of N (the exotic superconductor), a pair wave function of possibly different symmetry forms in the bulk of N and overlaps the induced pair wave function near the probe. Weak interactions between the induced and bulk pairs occur. In the case of UBe_{13} contacted with a Ta tip, the interaction weakly suppresses the induced pairs [which determine $I_c(T)$] leading to a reduction of the Josephson current at $T < T_c$. This observation of a negative S-wave proximity effect in superconducting UBe_{13}, in good agreement with a Ginzburg–Landau analysis, is strong evidence for triplet pairing in this heavy fermion compound.

I. INTRODUCTION

The heavy fermion superconductors (HFS), principally $CeCu_2Si_2$, UBe_{13}, and UPt_3, have been intensely studied for evidence of unconventional (non-BCS) electron pairing, e.g., $S = 1$, $L = 1$ (spin triplet, p wave) or $S = 0$, $L = 2$ (spin singlet, d wave). Several experiments show nonexponential power-law temperature dependence in low-temperature properties including specific heat[2] $c_p(T)$, ultrasonic attenuation[3] $\alpha(T)$, and nuclear spin lattice relaxation[4] rate $T_1^{-1}(T)$. Therefore, a single (isotropic) sharply defined gap apparently is not characteristic of the HFS. Such power-law T dependencies can indeed arise from the anisotropic gap functions expected from several of the triplet p-wave or singlet d-wave cases.[5] However, anisotropic $\Delta(\mathrm{k})$ behavior is well known in BCS superconductors, occurring even in Pb, and predicted for a singlet s-wave (conventional) superconducting state of UBe_{13} by Overhauser and Appel.[6] Gaplessness as occurs from pair breaking by magnetic impurities in BCS superconductors can also alter the T dependencies. For these reasons, a nonexponential T dependence alone is not a certain indication of triplet p-wave or singlet d-wave pairing. Experiments are called for whose results more sensitively test the symmetry properties of the pair wave functions. Such an experiment is the Josephson tunneling experiment.

II. JOSEPHSON TUNNELING

The dc Josephson current $I_c(T)$ between two S wave superconductors S_1 and S_2 is given[7] by

$$I_c(T) = I_J \sin\phi , \tag{1}$$

where ϕ is the difference $\phi = \phi_1 - \phi_2$ between the pair wave-function phases in the two electrodes. The critical current I_J in this case is proportional (T_{kq}),[2] where T_{kq} is the single-electron tunneling matrix element between S_1 and S_2. In case of irradiation by photons of frequency f, Shapiro steps will appear in $I_c(T,V)$ whose voltage spacing V_J is given by the ac Josephson relation

$$V_j = hf/2e . \tag{2}$$

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[a] Present address: Department of Physics, SUNY, Stony Brook, NY.
[b] Permanent address: Department of Physics, Seoul National University, Seoul, Korea.
[c] Present address: Department of Physics, University of Texas, Austin, TX.
[d] Present address: AT&T Bell Laboratories, Murray Hill, NJ 07974.

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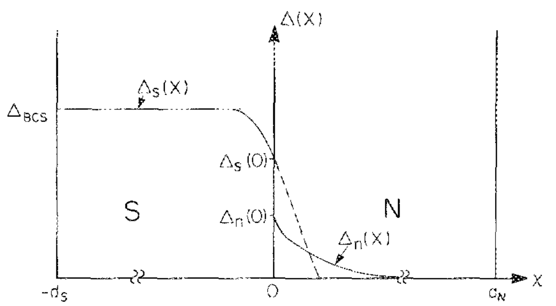

FIG. 1. Schematic form of order parameter $\Delta(x)$ near SN contact. The spatial variations extend about one coherence length from the interface at $x=0$.

In contrast, for the singlet-triplet (S-T) Josephson junction, discussed by Pals and van Haeringen,[8] the Josephson current vanishes in order $|T_{kq}|^2$, reflecting the orthogonality of pair wave functions of different symmetry. A (much weaker) Josephson current does occur in order $|T_{kq}|^4$, for which the Shapiro step spacing is halved: $V_J = hf/4e$.

The utility of this contrasting $I_c(T)$ behavior for directly identifying a triplet candidate superconductor is weakened by two complications. These are the effects of spin-orbit coupling typical of the HFS containing U or Ce atoms,[9] and the proximity-Josephson effect.[10] The former difficulty introduced by spin-orbit coupling has been emphasized by Anderson,[9] and its effects on the singlet-triplet Josephson $I_c(T)$ estimated by Fenton[11] and others. A perhaps oversimplified statement of the result is that in the HFS, with spin orbit, the spin of the pair is no longer a good quantum number. In triplet HFS with strong spin-orbit coupling, admixture of singlet pairs at a level $\sim a/\xi_0$ will occur, where a is the lattice constant and ξ_0 the coherence length. This will restore in the singlet-triplet junction a conventional $\sim|T|^2$ contribution to restore $I_c|T|$ of order $(a/\xi_0)I_J \lesssim 0.1\ I_J$. This fact makes the Pals criterion less definitive in identifying triplet superconductors.

Nevertheless, by careful observations of $I_c(T) \simeq (0.2\text{–}0.8)I_J$ in case of $CeCu_2Si_2$–Al vacuum tunnel junctions, Poppe[12] has established that $CeCu_2Si_2$ is very likely singlet paired, like Al. A second difficulty in applying the Pals criterion to singlet-triplet Josephson junctions to establish triplet pairing is the smaller size $\sim|T|^4$ of the predicted S-T Josephson current and the related difficulty in observing the halved Shapiro step spacing. Further, there is a tendency for the singlet-paired member of the S-T junction to induce local singlet pairing in the triplet material. This leads us to discussion of the proximity-Josephson effect.

III. PROXIMITY-JOSEPHSON EFFECT

I-V measurements on "SN" point contacts between a superconductor S with transition T_{cs} and a "normal metal" of lower transition T_{cn} (Fig. 1) show that the usual Josephson effects including Shapiro steps and oscillatory H dependence are typically observable[10] up to a junction critical temperature $T^* \simeq 0.8T_{cs}$. Typical data from a Ta/Mo contact above $T_{cn} = 0.92$ K are shown in Fig. 2. In retrospect, one can see that this effect should occur in SN contacts of sufficiently high electron tunneling probability. In such cases, a reasonable model of the variation of the pair potential $\Delta(x)$ near the SN interface can be deduced from the de Gennes boundary conditions[13]:

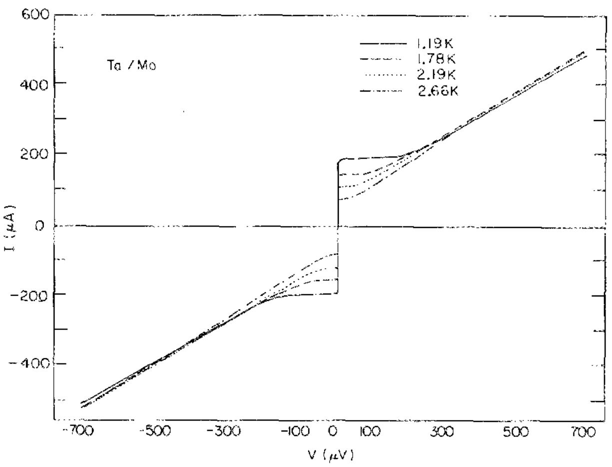

FIG. 2. *I-V* characteristics showing proximity-Josephson effect at temperatures above T_{cn}, in this case 0.92 K for bulk Mo.

$$\frac{\Delta}{N(0)V} = \text{constant}, \quad \frac{D}{V}\frac{d\Delta}{dx} = \text{constant}, \qquad (3)$$

at $x=0$, where $N(0)$ is the density of states, V the BCS effective electron–electron interaction, and D the electron diffusivity. A sketch of the result is shown in Fig. 1. The well-known result for a highly transmitting barrier is that electron pairs extend a coherence length into N. One might expect these proximity pairs to persist to temperatures approaching T_{cs}, above which pairs are no longer available from S. It is important to note that application of relations (3), following methods used, e.g., by Greenspoon and Smith,[14] allows determination of the interface values of $\Delta_N(0)$ and $\Delta_s(0)$, from which we can determine $I_c(T)$ for T near T^*.

The occurrence of the Josephson effects in this situation formally requires demonstration that the barrier-free energy, including the interaction of the pairs in S and N has an oscillatory dependence on the phase difference $\phi = \phi_1 - \phi_2$, between the pairs in S and N.

We have demonstrated that this occurs[10] for a simplification of the SN contact model of Fig. 1, in which one neglects the depression of Δ_s near $x=0$.

In a simple one-dimensional model[10] of the contact at $x=0$, we assume a proximity-induced pair wave function in N ($x \geqslant 0$) of $\psi_n = \psi_{n0} e^{-x/\xi_n} e^{i\phi_n}$, where ξ_n is a decay length. Here ϕ_n is the phase and ψ_{n0} is the modulus, whose value will be determined by minimizing the free energy of the contact. The pair wave function in S, $-\infty < x < 0$, is fixed as $\psi_s = \psi_{s0} e^{i\phi_s}$. The free energy near T^* can be estimated in Ginzburg–Landau theory, with the Josephson coupling energy taken as $\eta|\psi_s - \psi_n|^2$. Here η depends upon the coupling through the barrier at $x=0$. Thus the free energy of the induced Josephson junction is

$$F = \eta|\psi_s - \psi_n|^2 + \int_0^\infty \left(\alpha|\psi_n|^2 + \frac{\hbar^2}{2m^*}|\nabla\psi_n|^2\right)dx$$
$$= \eta\psi_{so}^2\,[\,1 + (1+\beta/\eta)y_n^2 - 2y_n\cos\phi\,]\,. \qquad (4)$$

Here $\beta = \hbar^2/2m^*\xi_n = \alpha\xi_n$, $\phi = \phi_s - \phi_n$, and the new variable is $y_n = \psi_{no}/\psi_{so}$. The value of y_n will be determined by the condition $\partial F/\partial y_n = 0$. This gives

$$y_n = (1+\beta/\eta)^{-1}\cos\phi\,, \quad |\phi| \leqslant \pi/2$$
$$= 0\,, \quad \pi/2 < |\phi| \leqslant \pi\,. \qquad (5)$$

The final result[10] for F is

$$F = \eta\psi_{so}^2\,[1 - (1+\beta/\eta)^{-1}]\cos^2\phi\,, \quad |\phi| \leqslant \pi/2$$
$$= \psi_{so}^2\,, \quad \pi/2 < |\phi| \leqslant \pi\,. \qquad (6)$$

That $y_n = 0$ is the lowest energy solution for $\pi/2 < \phi \leqslant \pi (\cos\phi \leqslant 0)$ is clear from the inherently negative coefficient of $\cos\phi$ in Eq. (4). Equation (6) contains an oscillatory term which gives rise to a Josephson effect. It is easy to see that the phase-dependent part of the free energy $F(\phi)$ in Eq. (6) has the usual period of 2π in ϕ, in spite of the $\cos 2\phi$ variation for $|\phi| \leqslant \pi/2$. Thus, the Josephson current-phase relation

$$I_c(\phi) = \frac{2e}{\hbar}\frac{\partial F}{\partial \phi}\,, \qquad (7)$$

although nonsinusoidal, retains the usual period 2π. It is therefore expected that the fundamental splitting of the Shapiro steps will be the conventional value of $V_J = hf/2e$.

An example of this behavior is shown in Fig. 3 for a Ta/UBe$_{13}$ point contact. In this figure the spacing of the steps is 52.9 μV, which is in agreement with Eq. (2) for the microwave frequency used. These data show another aspect of the proximity-induced effect in point contacts, which is a residual series resistance $R_s = dV/dI$ at $V = 0$. This is a consequence of the small dimension a, of the contact, and the fact that the bulk of the specimen remains normal. For bulk resistivity ρ, the expected value of the spreading resistance is $R_s = \rho/2a$, which is typically 0.1 Ω, depending upon a (in the range 1–10 μm) and the bulk resistivity.

The temperature dependence of $I_c(T)$ for the PJE near T^* has provided a useful test of the effect. $I_c(T)$ is relatively easy to measure, and a theoretical form of its temperature dependence near T^* has been derived[15] and found to fit data quite well. The theoretical function is based on the approximation of de Gennes[13]:

$$I_c(T) \propto \Delta_s(T)\Delta_N(T)\,, \quad T \sim T^*\,, \qquad (8)$$

which leads to

$$I_c(T) \propto (T^* - T)\left(1 + \frac{\alpha T f^2(T)}{T^* - T}\right)^{-1}, \qquad (9)$$

where

$$\alpha = \frac{3N_n^2 V_{Fn} l_n}{N_s^2 V_{Fs} l_s}\,, \quad f(T) = \left(1 + \frac{2}{\ln T/T_{cn}}\right)^{1/2}. \qquad (10)$$

Here N_n (N_s), V_{Fn} (V_{Fs}), and l_n (l_s) represent, respectively, the density of states, Fermi velocity, and mean free path in $N(S)$.

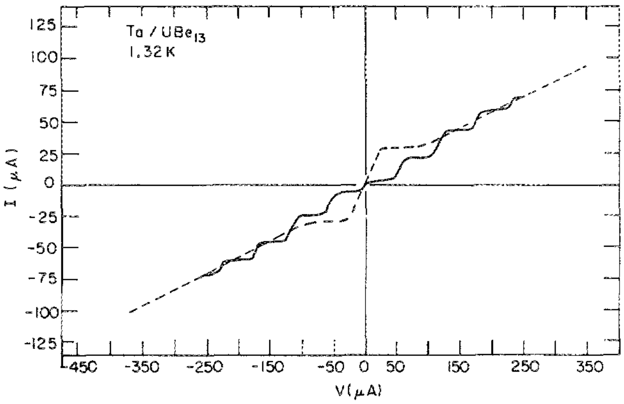

FIG. 3. I-V characteristics of Ta/UBe$_{13}$ contact in proximity regime (dashed), showing effect of microwave irradiation (solid curve). After correction for the expected series parasitic (spreading) resistence, the step spacing is $V_J = 52.9\ \mu$V, close to that predicted by Eq. (2).

The temperature dependence $I(T)$ for Ta/Mo and Ta/UBe$_{13}$ contacts is shown in Fig. 4, as fit by theory curves obtained from Eq. (9). In general, the fits obtained with Eq. (9) are satisfactory, and are superior to fits obtained previously[10] using the theories of the conventional S$_1$IS$_2$ tunnel junction and the clean weak link. We have also found that Eq. (9) provides superior fits to our data than other cases

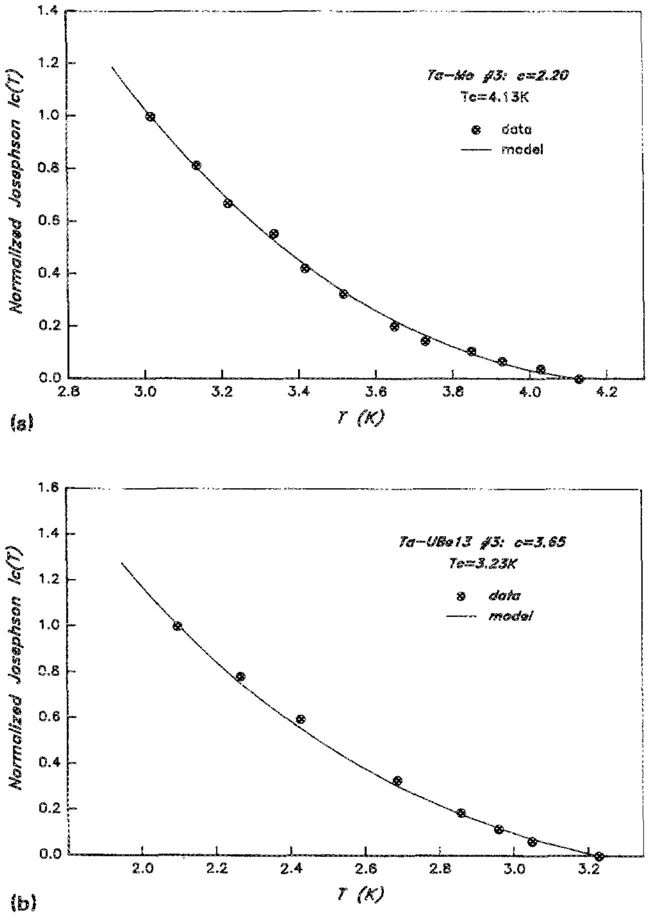

FIG. 4. (a) Temperature dependence of proximity-Josephson $I_c(T)$ near junction T_c, for Ta/Mo contact. The junction $T_c = 4.13$ K may be compared with the bulk value 4.4 K for Ta. The solid line is least-square fit of Eq. (9) to the data, with $\alpha = 2.2$. (b) Normalized $I_c(T)$ for Ta/UBe$_{13}$ contact. Theoretical line is fit by $\alpha = 3.65$. Junction T_c of 3.23 K exceeds 0.9 K, the bulk T_c of UBe$_{13}$, and is about 0.72 of that of Ta.

treated in Ref. 14. These new results are further support for our picture of the proximity-induced-Josephson effect.

It is perhaps surprising that no great differences have been observed in the susceptibility of the heavy fermion metals (we have previously studied UBe_{13},[10] $CeCu_2Si_2$,[10] $LaBe_{13}$[10]) and conventional d-band metals such as Mo or Ta, to the formation of induced singlet superconductivity in the point-contact configuration with a transition metal of higher T_c (usually either Ta or Nb). We do not have an adequate understanding of this aspect of our observations. This suggests that magnetic fluctuations, well known to suppress s-wave pairing, are not important in any of the tested materials at temperatures below the T_c of Nb, 9.2 K. The lack of a distinctive behavior in UBe_{13}, which we now believe to be triplet paired,[16] versus $CeCu_2Si_2$ (singlet paired below 0.6 K) and $LaBe_{13}$ (not known to superconduct) may simply confirm the relative weakness of the pairing interactions in all three metals (T_c below 1 K) compared to the superconductors ($T_c = 4.5$ or 9.2 K) used as the probe.

The most interesting and useful aspect of the proximity-Josephson effect has been its use to observe competition (negative proximity effect) between the induced s-wave state and the (triplet) bulk superconducting state below 0.9 K in UBe_{13}.

IV. MEASUREMENTS OF UBe_{13}

The experimental data on a Ta/UBe_{13} contact in Fig. 5 show an anomalous decrease in the critical current at 0.52 K, below the T_c of UBe_{13}, compared to its value at 0.99 K, above T_c. This negative proximity effect is inconsistent with an s-wave form of superconductivity in UBe_{13}, which would increase the low-temperature value of the induced pair potential Δ_N, and thus increase I_c. We have previously reported[16] the theoretically expected increase in I_c for an s-wave superconductor (Mo) at temperatures below its T_c, 0.92 K. A second feature of the data of Fig. 5 is inconsistent with s-wave superconductivity in UBe_{13}. Namely, the finite slope dV/dI at $V = 0$ is explained by the spreading resistance $R_s = \rho/2a$ only when the bulk UBe_{13} is in the normal state. Below its T_c, however, ρ vanishes and the slope of the V-I curve (as is shown in Fig. 5) would jump to zero at T_c if the bulk superconductivity were s wave. The failure of this to occur is additional evidence that UBe_{13} is not an s-wave superconductor. This behavior can be explained, however, on the basis of triplet order.[16]

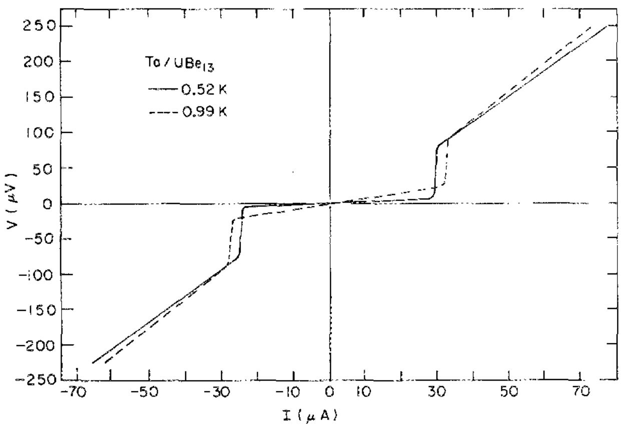

FIG. 5. Anomalous decrease in I_c of Ta/UBe_{13} contact when UBe_{13} is superconducting (solid) compared to its value above T_c (dashed). This is evidence that the superconductivity which appears in the bulk UBe_{13} suppresses the s-wave surface pairing induced by the Ta contact.

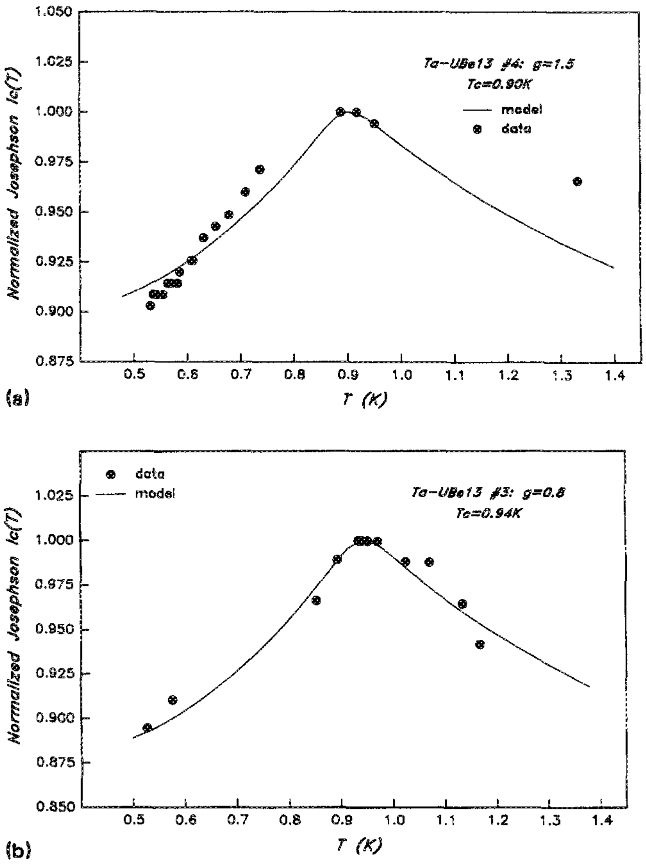

FIG. 6. (a),(b) Two sets of normalized $I_c(T)$ data of Ta/UBe_{13} contacts, showing anomalous decreases below the bulk T_c. In both cases the solid curve is a least-squares fit to the theoretical model described in the text.

The decrease in $I_c(T)$ below T_c, taken with the theoretical understanding described below, is strong evidence of triplet superconductivity in UBe_{13}.[16] Additional data showing the negative proximity effect are shown in Fig. 6. Our understanding of this effect, based on the assumption $I_c(T) \propto \Delta_N \Delta_S$ depends essentially on calculating the T dependence of Δ_N, the strength of the induced s-wave order parameter. This strength is fixed by a balance of the free energies arising from the interactions of Δ_N with Δ_S (the pairs in the Ta) and with the presumed triplet pair potential Δ_T in the bulk UBe_{13}. The result of this calculation is

$$\Delta_N = \eta \Delta_S \Bigg/ \left\{ \eta + 2\left[\ln \frac{T}{T_c^n} + \lambda \left(\frac{\Delta_T}{T_c^T} \right)^2 \right]^{1/2} \right\}, \qquad (11)$$

where $T_c^T = 0.86$ K is the T_c of UBe_{13}, T_c^n is the T_c the material would have for s-wave superconductivity, and $\lambda = \lambda_0 (\Delta_T/\Delta_\infty)^2$. In the latter, $\lambda_0 = 3.5$, and Δ_T/Δ_∞ measures the depression of Δ_T at the surface ($x = 0$) compared to the bulk value. The parameter η here and in Eq. (4) measures the coupling across the SN interface, and thus may be

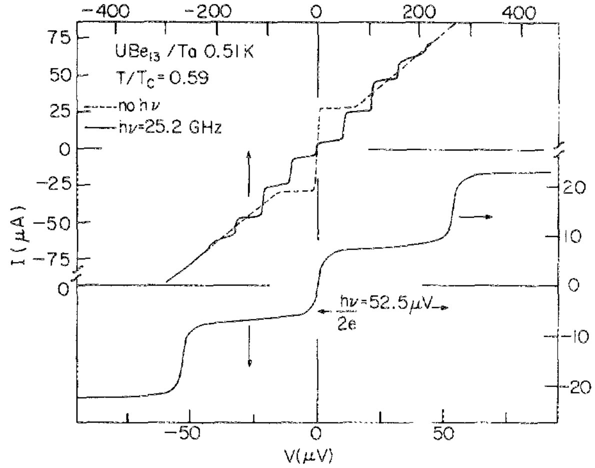

FIG. 7. Observations of microwave-induced steps in *I-V* characteristic of Ta/UBe$_{13}$ contact at the lowest temperature available, 0.51 K or $T/T_c = 0.59$. The step spacing of 52.5 μV indicates that the Josephson current flows from Ta to a singlet induced superconducting region in the UBe$_{13}$. The induced singlet surface pair potential $\Delta_n(0)$ is only gradually suppressed below T_c as indicated in Fig. 6. No sharp change in the steps occurs at the T_c of UBe$_{13}$, and there is thus no indication of direct Josephson coupling between the Ta superconductivity and the interior triplet superconductivity of the UBe$_{13}$.

expected to vary from contact to contact. All other parameters in Eq. (11) are based on material properties. Accordingly, theoretical fits (solid lines) to the data sets of Fig. 6 using Eq. (11) have been made using identical best values for the material parameters and adjusting only η. The best values, from a total of five data sets, over three runs, are $\lambda = 2.8$ and $T_c^n = 0.22$ K. These values are considered reasonable, and the overall agreement is regarded as strong support of triplet pairing in bulk UBe$_{13}$.

The possibility of unconventional singlet pairing, e.g., $S = 0$, $L = 2$ (*d* wave), is regarded as negligible,[16] in part because this would allow direct Josephson coupling between the Ta and the interior UBe$_{13}$ pairs. Careful examination of *I-V* data such as those of Fig. 7, as the temperature is varied through T_c, provides no indication of such direct coupling. The data of Fig. 7, at 0.51 K seem consistent only with $I_c(T) \propto \Delta_N \Delta_S$, where the induced surface order Δ_N is singlet.

In summary, we can say that the bulk order in UBe$_{13}$ below 0.86 K is definitely not singlet *s* wave, and, with high probability, is spin triplet *p* wave, in nature. The present experiment does not provide in itself a strong selection between the polar versus axial forms of triplet order. However, the present results, taken together with theory,[5,17] which, on symmetry grounds, rules out for cubic UBe$_{13}$ a state with a line of gap-function zeros (i.e., the polar state), imply that UBe$_{13}$ exhibits axial *p*-wave pairing. This is in accord with recent penetration depth measurements.[18]

ACKNOWLEDGMENT

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