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Anomalous s-wave proximity-induced Josephson effects in UBe_{13}, $CeCu_2Si_2$, and $LaBe_{13}$: A new probe of heavy-fermion superconductivity

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Anomalous proximity-induced s-wave superconductivity has been observed via the ac Josephson effect and the magnetic field and temperature dependences of the Josephson current in Nb/$CeCu_2Si_2$, Nb/$LaBe_{13}$, and Nb/UBe_{13} junctions. The origin of the proximity-induced Josephson effect and its utility in discovering the pairing type of heavy-fermion metals are discussed.

Intense recent interest has focused on the nature of the superconducting state in the heavy-fermion metals $CeCu_2Si_2$,[1,2] and UBe_{13},[3,4] and particularly on the possibility of triplet pairing in these metals.[5,6] In the course of performing point-contact Josephson tunneling experiments[7–9] between these metals and Nb we have discovered anomalous s-wave proximity and Josephson effects which occur at temperatures T_c^* well above the inherent transition temperatures of these heavy-fermion metals. The effect is also observed in $LaBe_{13}$, a d-band metal normal above 0.45 K.[10]

The signatures of the Josephson effect between two s-wave superconductors separated by a tunnel barrier are well known and include a supercurrent $I_0 \sin\phi$, where I_0 is proportional to $\Delta_1\Delta_2/(\Delta_1+\Delta_2)$; here Δ represents the pair potential at the surface of the superconducting electrode and ϕ represents the difference in the phases of the pairs in the two electrodes. The supercurrent ($V=0$) is also split into Shapiro steps of spacing $V_J=h\nu/2e$ under irradiation with photons of frequency ν (ac Josephson effect). Various forms of weak-link junction[11,12] are discussed, e.g., by Likharev.[13]

The potential of the Josephson effect to test for triplet superconductivity was first recognized and explored both theoretically and experimentally by Pals, von Haeringen, and van Maaren.[7,8] In the simplest case of a junction with tunneling matrix element T_b, the usual I_0, of order T_b^2 is replaced by a reduced contribution of order T_b^4, with halved Shapiro step spacing, $h\nu/4e$, under irradiation, for the case of a singlet-to-triplet superconductor junction. More recent discussions have been given by Fenton[14] and by Millis.[15,16]

Following standard methods[7,9] polycrystalline samples are mounted outside a small hole in the wide face of a K-band microwave guide and are contacted by a Nb pin traversing the narrow dimension of the guide as described previously.[9]

In Nb point-contact measurements on UBe_{13} (Ref. 17) and $CeCu_2Si_2$ (Ref. 18) an anomalous apparent Josephson effect was typically observed up to effective junction critical temperatures $T_c^* \sim 7$ K, while the effect on $LaBe_{13}$ (Ref. 19) was observed at 4.2 K [Fig. 1(c)]. The Josephson I-V curves were typically similar to those reported and analyzed in Ref. 8 in that a nonzero resistance dV/dI persisted at $V=0$. In the present case $T_c^* > T_c$ this residual series resistance[20] can be naturally interpreted as the spreading resistance into the normal-state bulk: $R_s \sim \rho/a$, with ρ the bulk resistivity and a the lateral dimension of a superconducting region induced by the Nb point contact. The depth of the induced superconducting region is the proximity coherence length.

Typical observations of the ac Josephson effect (Shapiro steps) are shown in Fig. 1. Corrections for the parasitic resistance effect yield results consistent, within experimental uncertainty, with an s-wave state in the sample, localized near the Nb contact. The Josephson current feature in UBe_{13}-Nb contacts was observed in each of several different sample mountings and several different point-contact junctions were studied in each run. In all cases a Josephson current feature was easily observable between 1.2 and 4.2 K.

An oscillatory variation of the critical current with magnetic field is a fundamental property of the Josephson effect, as this demonstrates the oscillatory dependence on the phase difference ϕ between the two pair-state wave functions. In a typical $I_c(H)$ plot for a UBe_{13}-Nb contact, shown in Fig. 2, the minimum at ~ 43 Oe is consistent with a single contact of dimension[21] $a \sim 5$–10 μm, while a relatively large normalized value at the second maximum is a behavior known to occur[22] when the contact dimension is comparable to or larger than the Josephson penetration depth $\lambda_J=(\hbar/2e\mu_0 J_1 d)^{1/2}$. Here J_1 is the current density and $d=\Lambda_1+\Lambda_2+t$, where $\Lambda_{1,2}$ are London penetration

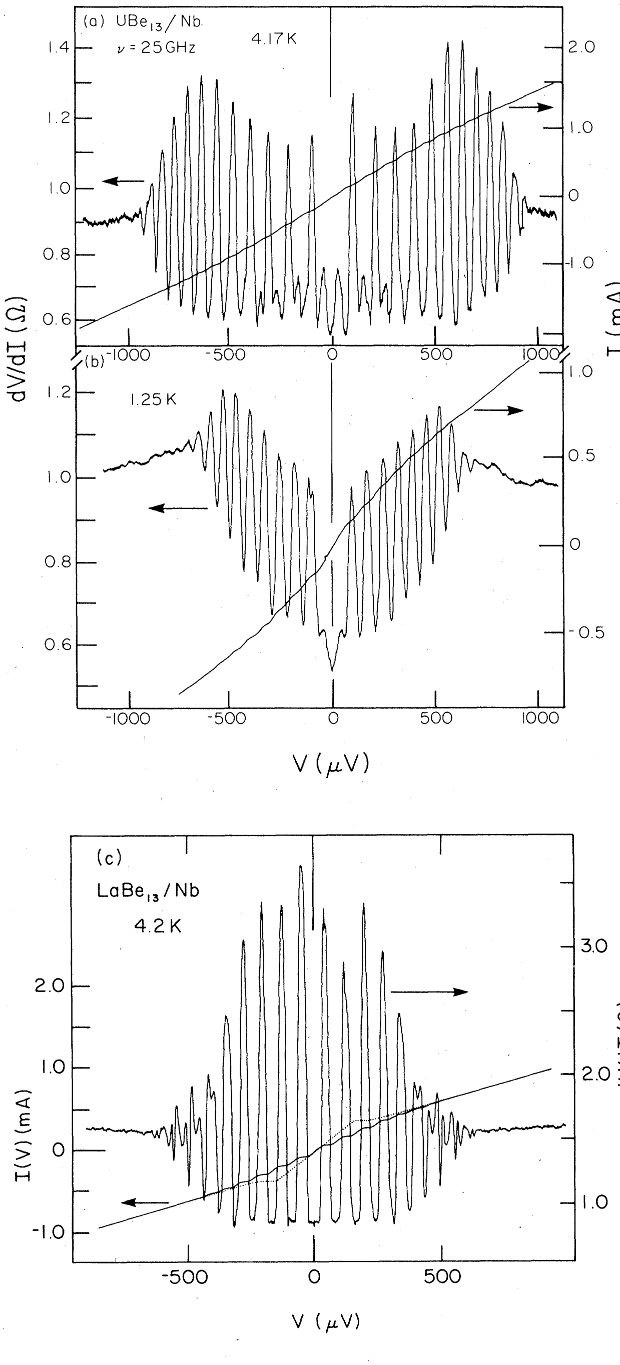

FIG. 1. (a) I-V and (dV/dI)-V spectra of UBe_{13}-Nb contact at 4.17 K, show conventional ac Josephson effect with 25.3-GHz microwaves ($\Delta V_J = h\nu/2e = 52.3$ μV). The Shapiro steps, more easily seen in the derivative trace, are spaced by 66 μeV, which is reduced to $\Delta V_J = 47 \pm 10$ μV after approximate corrections for parasitic series and parallel resistances ($R_s = 0.54$ Ω and $R_p = 1.2$ Ω) following analysis of Ref. 7. A fourth-order (T_b^4) effect, $\Delta V_J = h\nu/4e$, can evidently be ruled out. (b) ac Josephson-effect spectra of a second UBe_{13}-Nb contact at 1.25 K. The measured step spacing 74 μV yields $\Delta V_J = 54 \pm 5$ μV after corrections $R_s = 1.1$ Ω and $R_p = 1.5$ Ω. An s-wave to s-wave Josephson effect is implied. (c) I-V and dV/dI curves of Nb/$LaBe_{13}$ contact measured at 4.2 K reveals ac Josephson effect. Dotted curve is I-V at zero microwave power, solid curves are obtained under irradiation.

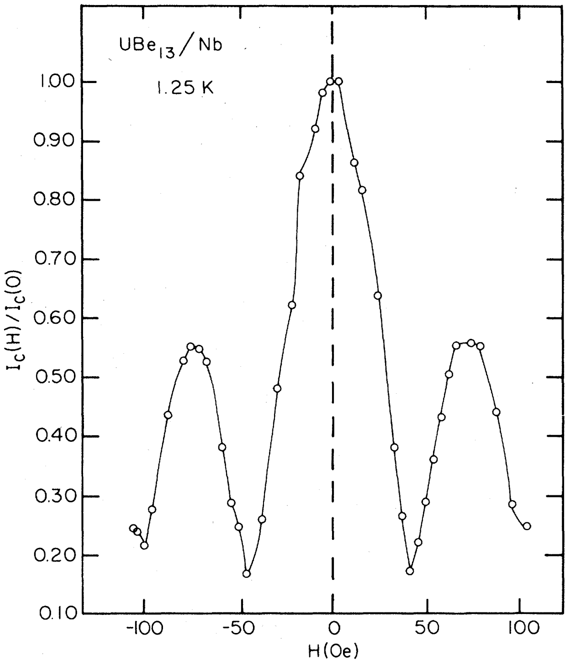

FIG. 2. Fraunhofer-type dependence of Josephson critical current I_c on parallel magnetic field, shown here for a typical UBe_{13}-Nb contact at 1.25 K. The value of $I_c(0)$ is 0.61 mA.

depths and t the barrier thickness for a tunnel junction.

For a typical $I_c \simeq 10^{-3}$ A, $J_1 \sim 10^3$ A/cm^2, $\Lambda_{Nb} = 440$ Å, and $\Lambda_{UBe_{13}} = 100$ Å, $t = 10$ Å gives $\lambda_J \cong 20$ μm, which is only a rough estimate.

The temperature dependence $I_c(T)$ typical of our Nb point-contact junctions is surveyed in Fig. 3. Typical values of $I_c(0)$ are the order of 1 mA, with spreading resistances typically 0.5 Ω, as is seen in Fig. 1. In order of magnitude, taking $R_s = \rho/a$, with $\rho \cong 200$ μΩ cm for UBe_{13}, one has $a = 4$ μm, and $J_c \cong I_c/a^2 \sim 6 \times 10^3$ A/cm^2, assuming that the spreading resistance from a contact of dimension a dominates the parasitic series resistance of the junction. This effective dimension is somewhat less than the dimension inferred above from $I_c(H)$, which could imply either a tunnel barrier of transmission on the order of ~ 0.1, or alternatively, an array of shorts which occupy ~ 0.1 of the contact area. The latter possibility seems to be favored by the general shape of the $I_c(T)$ curves shown here, which follow better a weak-link model[11-13] (the solid curves shown in comparison with the data points) than a tunneling model [the dashed curve shown in comparison to the test Nb-In junction data, curve (d), bottom].

The most anomalous aspect of our measurements using Nb ($T_c = 9.2$ K) is the persistence of the documented Josephson effects to temperatures on the order of 7.5 K, far above the inherent critical temperatures of UBe_{13} (0.85 K), $LaBe_{13}$ ($T_c < 0.45$ K), and $CeCu_2Si_2$ ($\cong 0.6$ K). These temperatures T_c^* are greatly enhanced, while the critical temperature of junctions between the same probe (Nb) and In is only slightly above the inherent T_c of the bulk electrode metal. Thus, in curve (d) of Fig. 3, $T_c^* = 3.82$ K, while the bulk T_c of In is 3.407 K. Similar small (0.3 K) enhancements of T_c^* have been previously observed using

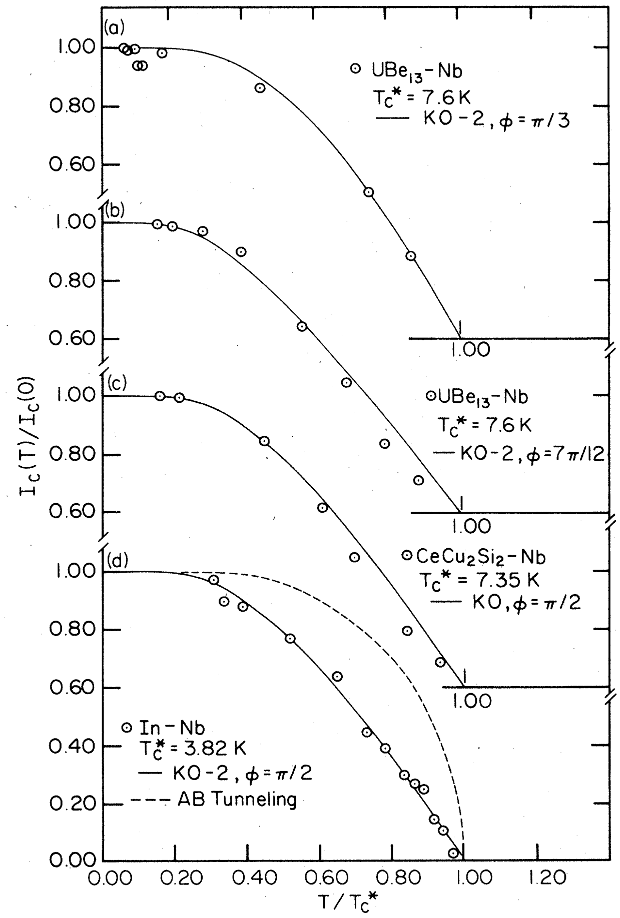

FIG. 3. Normalized critical Josephson currents vs T/T_c^* for typical Nb contacts to UBe$_{13}$ [curves (a) and (b)], CeCu$_2$Si$_2$ [(c)], and In [curve (d)]. The observed junction critical temperatures $T_c^* \sim 7.5$ K are anomalously high for heavy-fermion metals [(a)–(c)] while that seen for the test junction on In, $T_c^* = 3.82$ K, is close to the bulk T_c, 3.41 K. The data are inconsistent with the Ambegaokar-Baratoff tunneling calculation [(d), dashed curve, for $\Delta_1/\Delta_2 = 0.43$] but are described reasonably by the KO-2 clean weak-link theory (Ref. 11, solid curves, ϕ chosen arbitrarily). The observed values of $I_c(0)$(mA) are 1.24, 0.61, 0.21, and 0.65, for curves (a)–(d), respectively.

Sn point contacts;[23,24] a short report of Josephson effects in point contacts to normal metals Cu, CuZn, and Zn has been given[25] but apparently never confirmed.

Two questions are raised by these observations. The first question concerns the means by which a well-characterized Josephson effect can occur in a contact to a presumably normal metal. The second is, what properties of UBe$_{13}$, LaBe$_{13}$, and CeCu$_2$Si$_2$ allow the Josephson effect to occur at much higher temperatures T_c^*/T_c than in In and Sn?[23]

Observation of the Josephson effect requires an extended pair wave function with a definite phase residing in the electrode contacted by the Nb tip. The known geometry of the tips, observed by optical and electron microscopy, rules out the possibility that the second superconducting region is a "split" portion of the Nb tip. This, as well as the reasonable magnetic field dependences and well behaved test experiments on Nb and In electrodes, implies that the extended pair wave function being manifested by the Josephson effects must reside in the electrode opposite the Nb tip.

The basic idea of a proximity-induced Josephson effect is simple: a finite-order parameter is induced in the normal metal by the proximity effect (flow of pairs from the Nb point); the Josephson phenomena are then a consequence of coupling to this induced pair wave function. To establish the reality of this effect one must demonstrate that the free energy F of the contact is an oscillatory function of the phase difference ϕ. The following simplified analysis that plausibly demonstrates this effect is based on notes of one of us (M.R.B.), which are previously unpublished.

In a simple one-dimensional model of the Nb-metal (superconducting-normal) weak-link contact at $x=0$, assume a proximity-induced pair wave function $\psi_n = \psi_{n0} e^{-x/\xi_n} e^{i\phi_n}$ in N $(x \geqslant 0)$, where ξ_n is an appropriate 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, in a weak coupling approximation of Deutscher and Imry,[26] taken as $\eta|\psi_s - \psi_n|^2$. Here η depends upon overlap through the barrier or weak link at $x=0$. Thus, the free energy of the induced Josephson junction is

$$F = \eta|\psi_s - \psi_n|^2 + \int_0^\infty \left(\alpha|\psi_n|^2 + \frac{\hbar^2}{2m^*}|\nabla\psi_n|^2 \right) dx$$

$$= \eta\psi_{s0}^2[1 + (1+\beta/\eta)y_n^2 - 2y_n\cos\phi] \ , \qquad (1)$$

where $\beta = \hbar^2/2m^*\xi_n = \alpha\xi_n$, $\phi = \phi_s - \phi_n$, and the new variable is $y_n = \psi_{n0}/\psi_{s0}$. The correct value of y_n should be determined by the condition $\partial F/\partial y_n = 0$, and $y_n \geqslant 0$ (because y_n is essentially the modulus of the order parameter); this gives $y_n = (1+\beta/\eta)^{-1}\cos\phi$ and hence,

$$F = \begin{cases} \eta\psi_{s0}^2[1-(1+\beta/\eta)^{-1}\cos^2\phi], & |\phi| \leqslant \pi/2 \ , \\ \eta\psi_{s0}^2, \quad \pi/2 \leqslant |\phi| \leqslant \pi \ . \end{cases} \qquad (2)$$

That $y_n = 0$ is the lowest-energy solution of Eq. (1) for $\pi/2 \leqslant |\phi| \leqslant \pi$ ($\cos\phi$ negative) is clear from the inherently negative coefficient of $\cos\phi$ in 1. The oscillatory term indeed gives rise to a Josephson effect. It is important to recognize that $F(\phi)$ defined by Eq. (2) retains the usual period, 2π, in ϕ, in spite of a $\cos 2\phi$ variation for $|\phi| \leqslant \pi/2$. For this reason, the Josephson current-phase relation $J(\phi)$, given by $(2e/\hbar)\partial F/\partial\phi$, although nonsinusoidal, also retains the usual period, 2π. It is therefore expected that the fundamental splitting of the Shapiro steps will be the conventional s-wave value,[27] $V_J = \hbar\omega/2e$, as observed in all cases. At the same time, it is clear that the detailed prediction of this model, and the examination of the consequences of making its assumptions more realistic, deserve further attention. In particular, the model may be oversimplified by use of the weak coupling $\eta|\psi_s - \psi_n|^2$ form under conditions of strong NS coupling where η may also be phase dependent,[27,28] or by neglect of a term involving the superfluid velocity (phase variation) in the N region. In either case, however, the prefactor J_0 to the sinusoidal term contains $\eta^2/(\eta+\beta)$.

This formally quadratic dependence on η, which returns to the linear dependence of the SIS case, when $\beta << \eta$, reflects the fact that the superconducting state in N is induced by proximity. The material dependence of the predicted effect resides in the parameter $\beta = \hbar^2/2m^*\xi_N$ which should be small compared to η. Further work is in progress to clarify the material-parameter dependence of the proximity-induced Josephson effect.

The present importance of this effect is as a tool in probing the inherent bulk superconductivity of UBe_{13} and similar materials analogous to the earlier application by Ulrich[23] in studying paraconductivity. In reducing the temperature through the inherent T_c one should observe a change in the parasitic series resistance arising from the spreading resistance R_s to either zero (if the induced and inherent superconducting states are similar), or to a new value influenced by the boundary resistance between singlet and triplet phases, were the inherent bulk state of triplet character. [This transition is hinted at in the lowest-temperature points of curve (a) in Fig. 3.] Further study of this point is planned.

To the extent that the electronic properties advantageous to p-wave superconductivity are unfavorable to s-wave superconductivity,[5,14–16] the present observation of s-wave-induced superconductivity in UBe_{13} favors the possibility of an s-wave ground state for this metal.[6] On the other hand, the influence of the Nb pairs may be so great as to overcome the possible preference of UBe_{13} for a triplet state below 0.85 K.

Note added in proof. Since submitting this paper we have learned that concepts related to the proximity-induced Josephson effect were discussed theoretically by R. A. Ferrell [J. Low Temp. Phys. **1**, 23 (1969)] and by A. M. Kadin and A. M. Goldman [Phys. Rev. B **25**, 6701 (1982)].

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[18] The properties of the polycrystalline sample of $CeCu_2Si_2$ prepared at Geneva are listed under "sample No. 1" in Table I of Ref. 2.

[19] The single-crystal specimen of $LaBe_{13}$ was grown at Los Alamos National Laboratory.

[20] We have found that the residual resistance effect is unmeasurably small in our apparatus using a Nb test sample.

[21] This is a rough estimate because the geometry of contact is not known.

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[27] We thank P. A. Lee for correcting our earlier discussion of this point.

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