Analysis of the proximity-induced Josephson effect in superconducting-normal contacts

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Several investigators have observed microwave-induced steps and other Josephson-like features in the $I-V$ characteristics of point contacts between a conventional superconductor (S) and a normal metal (N). Recently, S. Han et al. have interpreted these results in terms of a model of a proximityinduced Josephson effect (PIJE) in an SN contact and have used this model to infer the existence of p -wave pairing in superconducting UBe₁₃. We show that their model is incorrect on fundamental grounds, and that their PIJE analysis is in direct conflict with long-standing theoretical and experimental studies of the pair-field susceptibility in SN tunnel junctions. In addition, we outline a possible alternative explanation for the experimental observations of Han et al. based on a model of a phase-slip center near the tip of the S point in an SN contact.

I. INTRODUCTION

A Josephson junction normally involves two superconductors (S and S') on either side of an insulating barrier or other weak link, an SS' contact. The dc and ac Josephson effects follow from quantum interference between the complex superconducting order parameters on the two sides. Nevertheless, apparent Josephson effects in SN point contacts have been repeatedly observed by a number of researchers over many years,¹⁻⁵ where N is a normal metal or a superconductor above its critical ternperature T_c . In all of these experiments, a superconducting point (usually Nb or Ta) has been pressed into a normal flat. The qualitative interpretation given has been that weak superconductivity S' is induced by S (via the superconducting proximity effect) in the surface of N , and that the apparent Josephson effects arise from quantum interference between S and S'. More recently, Han et al. have developed a quantitative model of a proximityinduced Josephson effect (PIJE) based on these ideas. $1, 2,$

However, this interpretation is fundamentally flawed, as we pointed in our earlier Comment.⁹ For quantum interference to be observed, the phase difference between S and S' has to be ^a free and independent parameter. In the proximity effect, the order parameter in N is merely the decaying tail of that in S, and this degree of freedom is not present. A complete theoretical and experimental analysis of weakly coupled SN contacts was carried out some years ago in the context of pair-field susceptibility measurements of SN tunnel junctions, $10-13$ and the resulting Josephson effect is second order in the coupling and distinctly different in character from the usual (firstorder) Josephson effect.

On the other hand, the observations of these quasi-Josephson effects are sufficiently reproducible to require an explanation. We suggest that a superconducting phase-slip center (PSC) develops near the end of the S point of the contact, and that the usual first-order Josephson effects are realized across this PSC.

In recent years there has been considerable interest in

the realization of exotic forms of superconducting interactions, such as triplet (p-wave) pairing. It has been proposed on theoretical grounds that the coupling between a conventional s-wave superconductor and an exotic p-wave superconductor should be anomalously weak ic p-wave superconductor should be anomalously weak
due to the different pairing symmetries.^{14,15} Several years ago Han et al ² carried out a set of point contact tunneling experiments from a Ta point into a crystal of the heavy-electron superconductor UBe_{13} , while the latter was in either the normal or superconducting states. Apparent Josephson effects were observed, and interpreted within the PIJE model as providing strong evidence for p-wave pairing in UBe_{13} ^{2,7} On the basis of our analysis we question this conclusion and try to account for the observations within our phase-slip model without invoking exotic pairing.

In the following sections, we first review SN contacts and the proximity effect. This is followed by a brief discussion of classical Josephson effects, leading into an analysis of the pair-field susceptibility. Finally, with this background, we describe the apparent Josephson effects observed in SN contacts and outline a phase-slip model that may account for the observations. Throughout, we will attempt to tie together diverse perspectives to achieve a more complete picture.

II. SN CONTACTS

If one has a clean, sharp interface between a superconductor S and a normal metal N (i.e., the operating temperature T is between T_{cS} and T_{cN}), some electrons from each side will diffuse into the other, even in the absence of net current flow. This gives rise to the well-known superconducting proximity effect,¹⁶ whereby some weak induced superconductivity exists in N near the boundary, and the gap parameter Δ in S is correspondingly reduced. The scale of this reflects the finite size of a Cooper pair, the Ginzburg-Landau coherence length pan, the <u>Unizourg</u>-Landau concretive length $\xi(T) = \xi(0)/\sqrt{|T - T_c|}$, where $\xi(0) \approx \hbar v_f / \pi \Delta(0)$ in S and $\hbar v_f / 2\pi k_B T$ in N [with the standard dirty-lim corrections if the mean free path $l < \xi(0)$].

The theory of the proximity effect is based on the simple Ginzburg-Landau equations, but in contacts between unlike materials the precise boundary conditions tend to be rather complex. A special case that has been of considerable interest recently is the possibility of proximity coupling between a standard s-wave superconductor and
one with triplet p-wave pairing,^{14,15} since it has been suggested that heavy-fermion superconductors may exhibit such triplet pairing.

One important feature that the standard theories of the proximity effect do not include is the seemingly simple case of current flowing across such an NS interface. As de Gennes pointed out, 17 "When normal currents and su percurrents coexist, the implied dissipative effects must be calculated by a dynamic equation more general than the Landau-Ginzburg equations." More recently, by properly taking into account the relevant nonequilibrium effects, the problem of current flow through an SN contact has been treated both theoretically and experimentally in two key configurations: one-dimensional flow perpendicular to the interface¹⁸ and three-dimensional $(3D)$ flow through a small point contact.¹⁹

Historically, the problem of one-dimensional SN current flow began with the observation of an excess resistance associated with the superconducting side of the SN contact.¹⁸ This can be understood simply within a generalized two-fluid model of a superconductor, by reference to the transmission-line representation of nonequilibrium superconducting dynamics (see Fig. 1).²⁰ Here the resistive channel represents the flow of normal quasiparticle current J_N , the parallel inductive line the supercurrent J_s , the shunt capacitors are associated with storage of a nonzero charge imbalance between the normal and superconducting components, and the shunt resistors correspond to conversion processes that relax this charge imbalance. On the N side of the interface, the current must flow as J_N (since that is the only channel available), whereas on the S side, the current must shift from J_s far from the interface to J_{N} at the interface. This occurs over a distance $\Lambda_Q = \sqrt{D \tau_Q}$, the diffusion length associated with the charge imbalance relaxatio time τ_{0} . This implicitly assumes that Λ_{0} is large compared to ξ_s which generally holds for conventional lowtemperature superconductors. The validity of this picture has been amply demonstrated in experiments that directly measured the voltage on small superconducting and normal microprobes positioned near an NS interface.²¹ In addition, a tunnel barrier at the SN interfac

FIG. 1. Transmission-line representation of nonequilibrium processes in one-dimensional current flow through an NS interface (from Ref. 20).

increases the contact resistance, but an additional excess resistance associated with charge imbalance relaxation is still measurable.²²

This picture of excess SN resistance is altered at low temperatures $T \ll T_c$, where a collisionless process, Andreev reflection, can produce charge imbalance relaxation on the scale of ξ . Here, an electron entering S from N with energy $E < \Delta$ gets reflected as a hole, with an electron pair making up the charge. Andreev reflection is also key to an understanding of the $I-V$ characteristics of small 3D point contacts $(ξ),¹⁹$ where an excess resistance is exhibited for voltages $V > \approx \Delta$. The result mus be modified somewhat if the interface is not clean, if there is a discontinuity in materials properties (density of states, Fermi velocity, etc.), or if there is a tunnel barrier. Then there is a a continuous transition to the expected $I-V$ curve for an SN tunnel junction.

III. JOSEPHSON EFFECTS

The classical Josephson effect involves two weakly coupled superconductors separated by a thin insulating tunnel barrier (SIS). The two pair wave functions decay exponentially into the insulator, and the potential energy of the coupled superconductors is $E_J = -E_0 \cos \phi$, where ϕ is the phase difference between the two superconducting order parameters. The potential minimum is of course for $\phi=0$, but if ϕ is constrained by current flow through the junction to have some other value, then the standard relation $I_s = I_c \sin\phi$ follows, where $I_c = 2eE_0/\hbar$. I_c is proportional to the normal-state conductance of the barrier, so that $I_c R_n = f(\Delta_1, \Delta_2)$, which approaches $\pi \Delta / 2e$ at low temperature if $\Delta_1 = \Delta_2 = \Delta$.

According to the ac Josephson effect, when a voltage V is applied across a Josephson junction, the phase ϕ evolves at a rate $\omega_i = d\phi/dt = 2 \text{ eV/A}$, which gives rise to an oscillating current $I_c \sin\omega_i t$, typically at microwave frequencies. This is illustrated schematically in Fig. 2. Applying an external oscillating current at ω_i or its subharmonics gives rise to so-called Shapiro steps in the I-^V characteristics, where there is ^a dc average of the oscillating supercurrent. The shape of the Shapiro step is a reduced replica of that of the zero-voltage supercurrent or zeroth step, just shifted in voltage by $n \hbar \omega / 2e$, where n is an integer.

FIG. 2. Representation of complex order parameters in standard SIS' Josephson effect. (a) $V=0$, $\phi = \text{const}$, $I_s \propto \sin \phi$ =const. (b) $V = V_{dc}$, ψ_2 fixed, ψ_1 rotating at $\omega_0 = 2$ eV/ \hbar , time average $I_s = 0$. (c) $V = V_{dc} + V_{ac} \sin \omega t$. If $\omega = \omega_0, \psi_1$ slows on top, time average $I_s \neq 0$.

It is well known that a variety of $S x S$ configurations can give rise to Josephson effects, where x can be a region of depressed or constricted supereonducting or even normal metal.²³ For an SNS junction, the basic picture is similar to that in a SIS, except that the exponentially decaying pair wave function occurs on a length scale of $\zeta_N \approx 1000$ Å rather than ≈ 2 Å as in an insulator. The effects already mentioned such as Andreev reflection manifest themselves in such additional features as subharmonic gap structure, but all of the main features of the Josephson effect also occur. A similar situation arises in all-superconducting microconstrictions, where dynamical processes in the phase-slip center (PSC) depress the gap in that region, leading to a junction that is rather similar to an SNS. Constrictions that are too large compared to ξ_s , however, can deviate from the ideal sinusoidal $I_s(\phi)$ relation—higher-order terms must be included. A point contact is typically very small and of unknown detailed geometry, and may involve either a clean metallic contact or tunneling through an insulating layer at the tip, depending on the surface preparation and contact pressure.

Finally, in dealing with Josephson effects between a singlet and a triplet superconductor,¹⁵ the same kinds of considerations are present as in the proximity efFect. Because the two order parameters are in competition, one expects the interaction to be weaker than that between two conventional superconductors, but the amount of this weakening may depend on a number of undetermined factors.

IV. PAIR-FIELD SUSCEPTIBILITY

In this section we point out that careful application of the Josephson interaction to an SN contact leads to a second-order Josephson effect, distinctly different from those already described, known in the literature as the "pair-field susceptibility,"^{10,13} which was fully verified experimentally many years ago.^{11,12} perimentally many years ago.^{11,12}

The key to understanding this effect is to recall that in the proximity effect, the order parameter in N represents an exponentially decaying tail from that in S, and therefore the phases are not independent. This differs from an SIS junction, where the order parameters on the two sides have independent existences. For the superconductor —insulator —normal metal (SIN) case, there is a coupling energy that goes as $|\psi_N|$ cos ϕ and a supercurrent $I_s \propto |\psi_N| \sin \phi$, where ψ_N is the induced superconducting order parameter in N. This yields $\phi=0$ for the static case. If one applies a dc voltage across the junction, then the order parameter on the S side must rotate in phase space, but the order parameter in N will tend to follow. Figure 3 illustrates how this is distinctly different than the usual Josephson efFect (Fig. 2). For large enough voltages and high enough frequencies, it will lag behind because of a finite diffusion time for the superconducting electrons, which must be determined from the proper dynamical equation. Since now $\sin \phi \neq 0$, this corresponds to a nonzero I_s , in parallel with a quasiparticle current $I_n(V)$. For frequencies much higher than the inverse response time, ψ_N will lag by 90°, the coupling will van-

FIG. 3. Representation of complex order parameters in second-order Josephson effect in SIN junction. (a) $V=0$, $\phi=0$, induced $|\psi_n| \propto \cos \phi > 0$, $I_s \propto \sin \phi = 0$. (b) $V = V_{dc}$, ψ_s rotates at ω_0 , ψ_N still tied to ψ_S but lags behind by ϕ =const, dc supercurrent $I_s > 0$, no current oscillation.

ish, and both $|\psi_N|$ and I_s go to zero.

The dynamical equation in the theory¹⁰ is provided by the simple time-dependent Ginzburg-Landau equation, where $\tau_{GL} = \pi \hbar / 8k_B(T - T_c)$ is the characteristic time. The resulting voltage-dependent dc supercurrent in the SIN contact can be expressed in the form

$$
I_s \propto \frac{\sin(2\phi)}{R_n^2} \propto \frac{\omega_0 \tau_{\rm GL}}{R_n^2 (1 + \omega_0^2 \tau_{\rm GL}^2)} \tag{1}
$$

where ω_0 =2 eV/ \hbar is the Josephson frequency and R_n the normal-state junction resistance. This voltage dependence shows a broad peak, located at a voltage $V \approx \hbar/2e\tau_{\text{GL}} \approx k_B(T-T_c)/e$ and of comparable width, is in agreement with experiments on Sn/Pb and Al/P junctions with very thin oxide barriers.^{11,12} Note als junctions with very thin oxide barriers.^{11,12} Note also that I_s is proportional both to $\sin(2\phi)$ and to $1/R_n^2$, indicating clearly the second-order nature of the effect. The reason this second-order effect is visible at a11 is because the quasiparticle current $I_n(V)$ is very small for a lowleakage tunnel junction for $V \ll \Delta$ and $T \ll T_c$, and because $I_s(V)$ is reduced by moderate magnetic fields.

This picture appears to be in violation of the Josephson effect, since we have $d\phi/dt = 0$ and a dc $I_s \neq 0$ for nonzero voltage. However, this voltage is actually present only across the normal channel of the NS contact.⁹ Compare Fig. 4 to Fig. 1 describing the 1D current flow through an SN boundary. In that problem, normal

FIG. 4. Transmission-line representation of currents and voltages in a SIN proximity-effect junction. Current starts in N (on left) as I_n , but leaves N as I_s . This conversion process requires a voltage, which also drives a parallel I_n across the junction.

current was being injected into a superconductor; here supercurrent is injected into a normal metal. In Fig. 4, if we assume that the nonequilibrium voltage in S is small and that the voltage across the superconducting channel is zero, then the voltage V across the junction in the normal channel must equal that between the two channels on the N side. As was shown in Ref. 9, this provides a completely self-consistent picture of charge imbalance relaxation in the N region.

This picture also makes it clear why I_s cannot be increased from zero without creating (in steady state) a voltage across the normal channel. If one applies a current I, then initially a current $I_s=I$ will flow across the junction, without voltage, in the superconducting channel. However, this will create a charge imbalance on the N side, which can be relaxed only in the induced superconductor near the contact. This charge imbalance, in turn, shows up as a voltage across the junction in the normal channel, so that normal quasiparticle current I_n will flow in parallel with I_s . This situation is different from that in a true SS' contact, where a supercurrent $I_{s} < I_{c}$ can be removed from the junction region without buildup of a charge imbalance there. We point out that these phenomena follow directly from the picture of a generalized two-fluid model, and depend only in their magnitude on a particular dynamical equation for the order parameter.

Since there is no zero-voltage supercurrent in this problem, one would not expect to see Shapiro steps for finite voltages if an external ac current is applied. This also follows from the fact that I_s for finite voltage has no ac component at the Josephson frequency, so that there is no local oscillator to mix with the applied signal. As one would expect, a theoretical analysis²⁴ shows that application of an ac signal will produce reduced replicas of the broad peak of Eq. (1) (effectively photon-assisted tunneling), shifted by $n \hbar \omega / 2e$, where *n* is an integer. Reference 24 also suggests that for certain sets of parameters, these features will look similar to Shapiro steps, particular in a derivative plot dI/dV , and might account for the anoma lous Josephson effect seen by Han, et al.^{1,2} However, we see this explanation as rather unlikely, since the experiments also exhibited an apparent critical current,⁶ which would be difficult to explain in terms of this second-order effect.

V. PROXIMITY-INDUCED JOSEPHSON EFFECTS IN SN CONTACTS

The preceding arguments, which are consistent with careful experiments over many years, clearly indicate that a standard first-order Josephson effect across an SN contact is fundamentally impossible. Nevertheless, Josephson effects of apparently the "forbidden" type have been observed repeatedly by a number of experimenters going back some 20 years.¹⁻⁶ In all of these experiment a superconducting point (usually Nb or Ta) has been pressed into a normal flat, and I-V curves have been observed that imply a standard first-order Josephson effect with a zero-voltage critical current and constant voltage Shapiro steps, in series with a spreading resistance from

the normal metal. These have been interpreted qualitatively as arising from a Josephson effect between S and the weak induced superconductor at the surface of N. This is exactly the situation that gives rise to the secondorder Josephson effects in the pair-field susceptibility; one cannot obtain both effects from the same physical picture.

Nevertheless, Han et al. have proposed a onedimensional model of current flow through a weakly coupled NS interface, based on the time-independent Ginzburg-Landau equations, thereby obtaining a firstorder Josephson effect between the two electrodes, which they referred to as the proximity induced Josephso effect.^{1,2} Moreover, they have carried out a set of systematic point-contact measurements that have attempted to address the issue of the symmetry of superconducting ordering in UBe_{13} and other novel superconductors, and interpreted these results within the context of the PIJE. Following this, several attempts have been made to place the PIJE on a firmer theoretical foundation.^{6,25} We feel that this approach is somewhat misdirected, since as we already discussed, it cannot self-consistently account for the experimental observations.

Still, the experiments stand on their own and cannot be ignored. We therefore propose an alternative model that can explain the facts equally well. Given that the observations indicate a first-order Josephson effect, the logical candidate is some sort of phase-slip process occurring inside the S point, near the boundary with N . This would exhibit the required critical current and Shapiro steps, in series with spreading resistance. The most obvious possible explanation for such a PSC, a broken tip, is not general enough to account for the reproducibility of the observed effects.

A more likely explanation (the phase-slip or PS model) is indicated schematically in Fig. 5. Given the point geometry, the total current density is expected to have its maximum value $J \approx I/\Omega a^2$ at the interface, where a is the size of the contact and Ω the solid angle subtended. However, within a two-fluid picture, $J = J_N + J_S$, where J_N starts to rise from zero on the superconducting side of the contact. Therefore, J_S is a maximum at a small distance inside the superconducting point, and a phase-slip center will nucleate at that point when J_S exceeds the local value of the critical current density J_c .²⁶ This PSC will give rise to Josephson effects between ψ_S and $\psi_{S-\text{tip}}$, where the order parameter at the tip is weaked slightly by proximity with N.

Geshkenbein and Sokol 24 have carried out a numerical solution of one-dimensional current flow through an SN interface, and have concluded that no PSC or other time-dependent phenomena exist near the boundary. They also suggest that the results would be similar for a three-dimensional contact. Qn the contrary, our picture requires the three-dimensional geometry, so that onedimensional calculations may be insufficient. It is noteworthy in this regard that these anomalous Josephson effects have been experimentally observed only in superconducting point contacts into normal flats, never in tunnel junctions or other one-dimensional geometries, suggesting the importance of this geometry in the effect.

This PS model may also explain observations^{1,2,6} on the temperature and magnetic-field dependence of the PIJE. With a superconducting Ta or Nb point in contact with a normal flat (Mo, In, UBe_{13} , $CeCu_2Si_2$), a PIJE was observed with a nonzero I_c starting at a temperature T_c^* somewhat below T_c of the Nb (typically 7.5 versus 9.2 K) or Ta (4.0 K versus 4.3 K). This I_c rose with decreasing temperature, and continued to rise as the temperature was lowered below the T_{cN} of the Mo or In (the results on UBe_{13} will be discussed later). Within the PS model, T_c^* < T_c probably reflects the fact that the superconductivity in the tip region is weakened somewhat by proximity with the normal fat. This also suggests that the PSC is only a couple of ξ from the interface. The increase in Is only a couple of ξ from the interface. The increase in the I_c below T_c^* reflects the increasing order parameter in the tip region. Below T_{cn} , the PSC moves to the interface and the increased I_c reflects the stronger overall superconducting coupling.

The dependence of I_c on a weak magnetic field parallel to the contact was a1so measured, and in one case' showed modulation wherein about 40 Oe corresponded to half a flux quantum. This yields an effective junction area of about 0.2 μ m². Taking an estimated PSC junction "thickness" $\approx 2\xi \approx 0.2 \mu m$, this gives a width of $\approx 1 \mu m$ for the diameter of the tip at the PSC. Given the approximate nature of this analysis, these values seem quite reasonable.

FIG. 5. Physical picture of phase-slip model for Josephson effects in SN point contacts. (a) S-point contacting N flat. (b) Spatial variation of current densities near SN interface. (c) Spatial variation of order parameter near interface, showing position of PSC.

Similar quasi-Josephson effects were observed² in point-contact tunneling from a superconducting Ta point into a single crystal of the heavy-fermion superconductor UBe_{13} , with the latter in either its normal or superconducting state. However, when T was lowered below the critical temperature of UBe $_{13}$, I_c actually dropped about 10% over the same temperature range that the contact spreading resistance was going to zero. This was interpreted as providing strong support for the contention that UBe_{13} was a triplet (p-wave) superconductor, based on theoretical calculations that have indicated a depressed proximity or Josephson effect when singlet and
triplet pairings compete.^{14,15} triplet pairings compete.^{14,15}
If UBe₁₃ were in fact a *p*-wave superconductor, then

the Josephson effects due to a PSC in a Ta/UBe₁₃ point contact might mell behave similarly to the results observed in Ref. 2. While we cannot rule this out, we suggest that a more conventional explanation must be considered before the more exotic explanation can be accepted.

In order to account for this observation within our PS model, we propose the existence of a possible thin layer on the surface of the UBe_{13} crystal with a critical temperature that is depressed below that of the bulk $T_{\text{c}UBe}$, due to structural or compositional surface effects. This would not be very surprising in this material, given its complicated crystal structure and very small superconducting coherence length (\approx 10 Å). If that is the case, then just below $T_{\text{c}UBe}$, one may have a SN'S' configuration. For sufficiently low temperatures, true phase coherence is obtained across this surface region, and the series resistance goes to zero. When this happens, however, the weak link is likely to move from just inside the Ta point to the middle of this N' region, and the value of this true I_c may be reduced below that of the PSC.

Although our PS model can in principal explain the experimental observations, it depends in detai1 on a number of undetermined parameters, making a direct experimental confirmation difficult. One possibility is to use a Nb point to contact, for example, a Ta flat coated with a thin layer of Cu or Ag. If one can simulate the results of Ref. 2 on UBe_{13} with this SNS configuration, this would further support the PS model.

VI. CONCLUSIONS

Point-contact tunneling is a very useful tool for probing electronic properties of superconductors and other materials. However, one must be careful in the interpretation of results, particularly since the geometry and nature of the contact itself are undetermined. The quasi-Josephson effect observed in SN contacts is an intriguing phenomenon still not fully understood, but from our analysis this effect is not due to a direct proximityinduced Josephson effect between the S point and the induced superconductor in N . On the contrary, an interaction of this latter type provides the basis for the well established pair-field susceptibility measurements of SN tunnel junctions, which exhibit a completely different sort of second-order Josephson effect. We propose that the quasi-Josephson effect is a consequence instead of a phase-slip center in the superconducting tip, and is only weakly dependent on the nature of the normal electrode.

Our analysis suggests that the conclusion of Ref. 2, that UBe_{13} is a triplet superconductor, is premature. Based on our phase-slip model, further experimentation and analysis is needed to settle the issue, one way or the other. The question of exotic superconductivity remains of great interest, however, particularly with regard to the new oxide superconductors. We would hope that more definitive tests may yet be possible, based in part on some of the approaches discussed here.

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