Probing interfacial pair breaking in tunnel junctions based on the first and the second harmonics of the Josephson current

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It will be shown that a pronounced interfacial pair breaking can be identified in Josephson tunnel junctions provided the first j_{1c} and the second j_{2c} harmonics of the supercurrent, as well as the depairing current in the bulk $j_{\rm dp}$, are known. Namely, within the Ginzburg-Landau theory a strong interfacial pair breaking results in the relation $j_{2c}j_{\rm dp} \gg j_{1c}^2$, while in standard junctions, with negligibly small pair breaking, the relation of opposite character takes place.

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I. INTRODUCTION

A remarkable property of superconducting weak links is that the local conditions in a small transition region control the whole process of charge transport. For the same reason, interface-induced suppression of the superconducting condensate density can have a considerable influence on the Josephson effect. A strong surface pair breaking has been theoretically established for various unconventional superconductors as well as for magnetic interlayers under certain conditions.[1–9] Therefore, probing the condensate density near the interface would provide valuable information for studying and controlling fundamental characteristics of the superconducting junctions. It is still an ongoing problem for the junctions though the order parameter profiles near superconductor-vacuum surfaces have been recently determined using a scanning tunneling microscopy method with a superconducting tip.[10]

In superconducting tunnel junctions the first harmonic $j_1 = j_{1c}\sin\chi$ usually strongly dominates the Josephson current $j = j_{1c}\sin\chi + j_{2c}\sin 2\chi + \cdots$, while the second harmonic $j_2 = j_{2c}\sin 2\chi$ represents a small correction to the first one, $|j_{2c}| \ll |j_{1c}|$, mostly due to a small junction transparency. Qualitatively different phase dependencies of the two harmonics allow one to study and distinguish between them experimentally. It is therefore of interest to find out which characteristic properties of the superconducting junctions can be identified with the data provided by the two harmonics. Thus the first harmonic, as opposed to the second one, is known to be noticeably suppressed both at 0-π transitions as well as in the junctions involving unconventional superconductors with special interface-to-crystal orientations.[11–18] However, except for these special cases, the relation $|j_{2c}| \ll |j_{1c}|$ always takes place and does not qualitatively discriminate between various superconducting tunnel junctions.

This paper suggests a test, which will be derived within the Ginzburg-Landau (GL) theory and will allow identification of a pronounced interfacial pair breaking in tunnel junctions, provided the first and the second harmonics, as well as the depairing current in the bulk $j_{\rm dp}$, are known. The relation $j_{2c}j_{\rm dp} \gg j_{1c}^2$ will be shown to take place in tunnel junctions with a strong interfacial pair breaking, while in standard tunnel junctions, with negligibly small pair breaking, it will be $j_{2c}j_{\rm dp} = 0.27 j_{1c}^2 < j_{1c}^2$. The specific temperature dependencies of the two harmonics near T_c will also be determined. The self-consistency is shown to alter existing estimates of both harmonics considerably. Initially, the theory is based on the interface free energy, containing only the terms that are quadratic or bilinear in the superconducting order parameters. Later on, the study will be extended to include the next order, quartic and biquadratic, corrections. They will be shown to result in the material-dependent coefficients, which are independent of temperature near T_c and should, generally, be kept on a par with numerical terms of the order of unity in the expressions for the order parameters.

II. BASIC EQUATIONS

Consider tunnel junctions with the spatially constant width, which is much less than the Josephson penetration length, and with a plane interlayer at $x = 0$ of zero length within the GL approach. Assume the usual form of the GL free energy, which applies, for example, to s-wave and $d_{x^2-y^2}$-wave junctions: $\mathcal{F} = \mathcal{F}_{b1} + \mathcal{F}_{b2} + \mathcal{F}_{\rm int}$. Here $\mathcal{F}_{b1(2)}$ are the bulk free energies of two superconducting leads and $\mathcal{F}_{\rm int}$ is the interface free energy. For a junction with two identical superconductors, the bulk free energies have identical coefficients

$$\mathcal{F}_{b1(2)} = \int_{V_{1(2)}} (K|\nabla\Psi_{1(2)}|^2 + a|\Psi_{1(2)}|^2 + (b/2)|\Psi_{1(2)}|^4)dV_{1(2)}. \quad (1)$$

Here $K, b > 0$, $a = \alpha\tau = -\alpha(T_c - T)/T_c$.

Asymmetry can be generally maintained by different conditions on the opposite sides of the interface, as in d-wave junctions with different crystal-to-interface orientations, and/or in junctions with asymmetric magnetic interfaces. Then the interface free energy incorporates different contributions from the two superconducting banks:

$$\mathcal{F}_{\rm int} = \int_S \big[g_{11}|\Psi_1|^2 + (1/2)h_{11}|\Psi_1|^4 + g_{22}|\Psi_2|^2 + (1/2)h_{22}|\Psi_2|^4 + h_{12}|\Psi_1|^2|\Psi_2|^2 + (g_{12} + \eta_1|\Psi_1|^2 + \eta_2|\Psi_2|^2)|\Psi_1 - \Psi_2|^2 + f_{12}\big|\Psi_1^2 - \Psi_2^2\big|^2\big]dS. \quad (2)$$

In addition to the main terms, which are quadratic or bilinear in the order-parameter moduli, the quartic and biquadratic terms of the next order of smallness near T_c are kept in Eq. (2).

In tunnel junctions with small transparencies $\mathcal{D} \ll 1$ one gets $g_{12}, \eta_1, \eta_2 \propto \mathcal{D}$, $h_{12}, f_{12} \propto \mathcal{D}^2$.

For the order parameter $f(x)e^{i\chi(x)}$ normalized to $f = 1$ in the bulk without superflow, the first integral of the GL equation in the presence of the supercurrent[19] takes the form

$$\left(\frac{df}{d\tilde{x}}\right)^2 + f^2 - \frac{1}{2}f^4 + \frac{4\tilde{j}^2}{27f^2} = 2f_\infty^2 - \frac{3}{2}f_\infty^4. \quad (3)$$

Here $\tilde{x} = x/\xi$, $\xi = \sqrt{K/|a|}$ is the temperature-dependent superconducting coherence length, $\tilde{j}$ is the spatially constant normalized current density $\tilde{j} = j/j_{\rm dp} = -(3\sqrt{3}/2)(d\chi/d\tilde{x})f^2$, $j_{\rm dp} = 8|e|K^{1/2}|a|^{3/2}/3\sqrt{3}\hbar b$ is the depairing current in the bulk, and f_∞ is the asymptotic value of f in the depth of the superconducting leads.

The boundary conditions (BC) originate from the variation of $\mathcal{F}_{\rm int}$ and from the bulk gradient terms integrated by parts. One starts with the BC in standard linear approximation in $f_1(-0) \equiv f_{10}$ or $f_2(+0) \equiv f_{20}$. Taking real and imaginary parts of the BC for the complex quantity $f(x)e^{i\chi(x)}$, one finds the following linear BC and the expression for the supercurrent

$$(df_i/d\tilde{x})_0 = (-1)^i[(\tilde{g}_{ii} + \tilde{g}_{12})f_{i0} - \tilde{g}_{12}\cos\chi\, f_{\bar{i}0}], \quad (4)$$

$$\tilde{j} = (3\sqrt{3}/2)\tilde{g}_{12} f_{10} f_{20} \sin\chi. \quad (5)$$

Here $i = 1,2$, $\bar{i} = 3 - i$. The phase difference of the order parameters across the interface is $\chi = \chi_{10} - \chi_{20}$, and $\tilde{g}_{12} = g_{12}\xi(T)/K$, $\tilde{g}_{11} = g_{11}\xi(T)/K$, and $\tilde{g}_{22} = g_{22}\xi(T)/K$ are the effective dimensionless coefficients.

For tunnel junctions $|\tilde{g}_{12}| \ll 1$. Since the order parameters near pair-breaking interfaces vary on a scale $\gtrsim \xi(T)$, one gets from Eq. (4) $\tilde{g}_{ii} f_{i0} \lesssim 1$, on account of $|\tilde{g}_{12}| f_{i0} \ll 1$. This signifies, in particular, that for $\tilde{g}_{ii} \gg 1$ a strong interfacial pair breaking $f_{i0} \lesssim \tilde{g}_{ii}^{-1} \ll 1$ occurs.

III. TEST FOR A PRONOUNCED INTERFACIAL PAIR BREAKING

Consider the supercurrent within the second-order perturbation theory in $\tilde{g}_{12}$. Then, according to Eq. (5), quantities f_{10} and f_{20} should contain the terms of the zeroth and the first orders of smallness. One takes $x = \pm 0$ in Eq. (3) and substitutes there (4) and (5). Since the depairing effects in the bulk would contribute to Eq. (3) only beginning with the second-order terms, within the given accuracy $f_\infty = 1$. Then one obtains the following equations for f_{10} and f_{20}:

$$f_{i0}^4 - 2\left(1 + \tilde{g}_{ii}^2\right)f_{i0}^2 + 1 = 4\tilde{g}_{12}\tilde{g}_{ii} f_{i0}(f_{i0} - f_{\bar{i}0}\cos\chi). \quad (6)$$

In the zeroth order in $\tilde{g}_{12}$ the solutions are

$$f_{i0}^{(0)^2} = (1/2)\left(\sqrt{2 + \tilde{g}_{ii}^2} - \tilde{g}_{ii}\right)^2, \quad i = 1,2. \quad (7)$$

Equation (7) involves two solutions of Eq. (6). At $g_{ii} > 0$ it describes a pair breaking $f_0 < 1$, and then $|df^{(0)}/d\tilde{x}|_0 < 1/\sqrt{2}$. At $g_{ii} < 0$ an enhanced superconductivity at the boundary $f_0 > 1$ occurs.[20–24] Then the quantity $|df^{(0)}/d\tilde{x}|_0 > \sqrt{2}g_{ii}^2$ can take large values, and a strong enhancement would induce a characteristic scale substantially less than $\xi(T)$ of the leads (see Appendix for details).

The first-order corrections $f_{0i} \approx f_{0i}^{(0)} + f_{0i}^{(1)}$, which follow from Eqs. (6) and (7), are

$$f_{i0}^{(1)} = -\tilde{g}_{12}\left(f_{i0}^{(0)} - f_{\bar{i}0}^{(0)}\cos\chi\right)\Big/\sqrt{2 + \tilde{g}_{ii}^2}. \quad (8)$$

Substituting the order parameters f_{0i} in Eq. (5), one finds the first and the second harmonics of the supercurrent $\tilde{j} = \tilde{j}_{c1}\sin\chi + \tilde{j}_{c2}\sin 2\chi$ in the Josephson tunnel junctions:

$$\tilde{j}_{c1} = \frac{3\sqrt{3}\tilde{g}_{12}}{4}\left(\sqrt{2 + \tilde{g}_{11}^2} - \tilde{g}_{11}\right)\left(\sqrt{2 + \tilde{g}_{22}^2} - \tilde{g}_{22}\right) \times\left[1 - \frac{\tilde{g}_{12}}{\sqrt{2 + \tilde{g}_{11}^2}} - \frac{\tilde{g}_{12}}{\sqrt{2 + \tilde{g}_{22}^2}}\right], \quad (9)$$

$$\tilde{j}_{c2} = \frac{3\sqrt{3}}{8}\tilde{g}_{12}^2 \sum_{i=1}^{2} \frac{1}{\sqrt{2 + \tilde{g}_{ii}^2}}\left(\sqrt{2 + \tilde{g}_{\bar{i}\bar{i}}^2} - \tilde{g}_{\bar{i}\bar{i}}\right)^2. \quad (10)$$

The second harmonic (10) is induced by the proximity across the interface. At $g_{ii} < 0$, the quantity $|g_{ii}|$ is here assumed not to be too large to retain $|\tilde{j}_c| \ll 1$ and $|g_{ii}| \ll \sqrt{K\alpha}$. Otherwise, Eqs. (9) and (10) are applicable at any values of $\tilde{g}_{ii}$.[25] Further, the small second and third terms in the square brackets in Eq. (9) will be neglected.

One finds from Eqs. (9) and (10) the following relationship between the amplitudes $\tilde{j}_{c2}$ and $\tilde{j}_{c1}$:

$$\tilde{j}_{c2} = \frac{\tilde{j}_{c1}^2}{6\sqrt{3}} \sum_{i=1,2} \frac{1}{\sqrt{2 + \tilde{g}_{ii}^2}}\left(\sqrt{2 + \tilde{g}_{ii}^2} + \tilde{g}_{ii}\right)^2. \quad (11)$$

Under the conditions $|\tilde{g}_{ii}| \ll 1$ $(i = 1,2)$, one can disregard the interfacial proximity effects. Then in the original units $j_{c1} \propto |\tau|$, $j_{c2} \propto \sqrt{|\tau|}$, while the relative magnitudes of the two harmonics are described by the equalities

$$j_{c2} j_{\rm dp} = 0.27 j_{c1}^2, \quad j_{c2} = 0.7\tilde{g}_{12} j_{c1}. \quad (12)$$

Consider now asymmetric junctions with a pronounced interfacial pair breaking on one side of the interface, when $|\tilde{g}_{11}| \ll 1$ and $\tilde{g}_{22}^2 \gg 1$, $\tilde{g}_{22} > 0$. In d-wave junctions this can take place for interface-to-crystal orientations, which are close to (100) and (110) orientations on the opposite banks of a smooth interface. Then Eqs. (9) and (10) are reduced to $\tilde{j}_{c1} \approx 3\sqrt{3}\tilde{g}_{12}/(2\sqrt{2}\tilde{g}_{22})$, $\tilde{j}_{c2} \approx 3\sqrt{3}\tilde{g}_{12}^2/(4\tilde{g}_{22})$ while $j_{c1} \propto |\tau|^{3/2}$, $j_{c2} \propto |\tau|$ in the original units. The relationships between the harmonics are

$$j_{c2} j_{\rm dp} = 0.385\tilde{g}_{22} j_{c1}^2, \quad j_{c2} = 0.7\tilde{g}_{12} j_{c1}. \quad (13)$$

In symmetric junctions with $\tilde{g}_{11} = \tilde{g}_{22} > 0$ and $\tilde{g}_{ii}^2 \gg 1$ one gets from Eqs. (9) and (10) $\tilde{j}_{c1} = 3\sqrt{3}\tilde{g}_{12}/4\tilde{g}_{22}^2$, $\tilde{j}_{c2} = 3\sqrt{3}\tilde{g}_{12}^2/4\tilde{g}_{22}^3$. Hence, $j_{c1} \propto \tau^2$, $j_{c2} \propto \tau^2$, and

$$j_{c2} j_{\rm dp} = 0.77\,\tilde{g}_{22}\, j_{c1}^2, \quad j_{c2} = (\tilde{g}_{12}/\tilde{g}_{22})\, j_{c1}. \quad (14)$$

In symmetric junctions the quantity $\tilde{j}_{c2}/\tilde{j}_{c1} \propto \tilde{g}_{22}^{-1}$ diminishes with increasing pair breaking.[26]

It also follows from Eqs. (9)–(11) that $j_{c2} j_{\rm dp} = 0.136\ j_{c1}^2$ for $|\tilde{g}_{11}| \ll 1$ and $\tilde{g}_{22}^2 \gg 1$, $\tilde{g}_{22} < 0$. In symmetric junctions with $\tilde{g}_{ii}^2 \gg 1$, $\tilde{g}_{ii} < 0$ one gets $j_{c2} j_{\rm dp} = 0.19\ j_{c1}^2/|\tilde{g}_{22}|^3$. Under the conditions $\tilde{g}_{11} < 0$, $\tilde{g}_{22} > 0$, $\tilde{g}_{ii}^2 \gg 1$ $(i = 1,2)$ the relation is $j_{c2} j_{\rm dp} = 0.385\, \tilde{g}_{22} j_{c1}^2$.

Comparing (12)–(14), as well as the results for $\tilde{g}_{ii} < 0$, one can conclude that the quantity $|j_{c2}| j_{\rm dp}/j_{c1}^2$ always exceeds unity, when a pronounced interfacial pair breaking $\tilde{g}_{ii}^2 \gg 1$, $\tilde{g}_{ii} > 0$ takes place on at least one side of the interface. At $0.4\tilde{g}_{22} \gg 1$, the strong inequality $|j_{c2}| j_{\rm dp} \gg j_{c1}^2$ emerges as a sure sign of the strong interfacial pair breaking. By contrast, $j_{c2} j_{\rm dp}/j_{c1}^2$ is substantially less than unity for the negligibly weak pair breaking or for the enhanced superconductivity, on both sides of the interface.

Though the specific temperature dependencies, determined above for both harmonics at different strengths of the pair breaking, could be identified near T_c, there are no striking differences between them. At the same time, the power-law temperature dependencies of the harmonics $j_{ci} = j_{ci,0}|\tau|^{\nu_i}$ actually drop out of Eqs. (12)–(14), together with the dependencies of the effective coupling constants $\tilde{g}_{il} = \tilde{g}_{il,0}|\tau|^{-1/2} = (g_{il}\xi_0/K)|\tau|^{-1/2}$ $(i,l = 1,2)$ and of the depairing current $j_{\rm dp} = j_{\rm dp,0}|\tau|^{3/2}$. Hence, (12)–(14) are applicable to the "low-temperature" amplitudes of the GL theory, and then the relation $|j_{c2,0}| j_{\rm dp,0} \gg j_{c1,0}^2$ will be valid, if at least one of $\tilde{g}_{ii,0}$ satisfies the condition $\tilde{g}_{ii,0} \gg 1$. The latter condition is more restrictive than $\tilde{g}_{ii} \gg 1$. Since the quantities $\tilde{g}_{ii}$ incorporate contributions from a relatively wide angular interval of quasiparticle momentum directions, they can be quite large in anisotropically paired superconductors near T_c, but as a rule, decrease substantially when the temperature goes down.[2] However, this is generally not the case for pair-breaking effects induced by magnetic boundaries.[8,9] If $\tilde{g}_{ii,0} \ll 1$ while $\tilde{g}_{ii} \gg 1$, a crossover from $|j_{c2}| j_{\rm dp} \gg j_{c1}^2$ close to T_c to $|j_{c2}| j_{\rm dp} \lesssim j_{c1}^2$ will show up with decreasing temperature, as described by Eq. (11).

Assume now $\tilde{g}_{ii,0} \gg 1$. The GL "low-temperature" values of the quantities usually exceed their actual values at $T = 0$ by about 2–3 times. This concerns, in particular, the depairing current: $j_{\rm dp,0}/j_{\rm dp}(T = 0) \approx 2.6$.[27–30] For the standard Josephson current,[31] one obtains $j_{c1} = j_{c1,0}|\tau| = 2\pi^3 T_c|\tau|/7\zeta(3)|e|R_N$ near T_c and $j_{c1}(T = 0) = \pi\Delta_0/2|e|R_N$. Hence $j_{c1,0}/j_{c1}(T = 0) = 2.66$. Despite the value it would have for analyzing the experimental results,[10] there still is no microscopic theory for the effects of strong interfacial pair breaking in a wide temperature range. If, qualitatively, no dramatic changes of behavior take place and $\tilde{g}_{ii,0} \gg 1$, the relation $|j_{c2}| j_{\rm dp} \gg j_{c1}^2$ could remain valid with decreasing temperature below the GL domain of applicability unless anomalous temperature dependencies, if present, come into play, e.g., due to Andreev bound states with low energies $\varepsilon_B \ll \Delta_0$. The temperature dependence of $(|j_{c2}| j_{\rm dp}/j_{c1}^2)$ in the whole temperature range is of interest for further theoretical and experimental studies.

IV. MICROSCOPIC FORMULA FOR $\tilde{g}_{12}$

Microscopic expressions for $\tilde{g}_{12}$ and for $\tilde{g}_{ii}$ $(i = 1,2)$ can be obtained by comparing the Josephson currents of the GL theory with the corresponding microscopic results near T_c. Consider here standard symmetric SIS tunnel junctions with the negligibly small pair breaking $|\tilde{g}_{ii}| \ll 1$. Then the GL expression for the first harmonic should coincide with the microscopic Ambegaokar-Baratoff formula[31] near T_c: $j_{c1} = 4|e||a|g_{12}/(\hbar b) = \pi|\Delta|^2/(4|e|T_c R_N)$. Here R_N is the junction resistance in the normal state. Since $K = \hbar^2/4m$, $|a| = \alpha|\tau|$ and, in the absence of the pair breaking, the BCS gap function near T_c is $|\Delta|^2 = 8\pi^2 T_c(T_c - T)/[7\zeta(3)]$, one obtains

$$\tilde{g}_{12} = 2\pi^3 T_c m b \xi(T)/[7\zeta(3)e^2\hbar\alpha R_N]. \qquad (15)$$

Equation (15) can be transformed further with the Gor'kov microscopic formulas for b/α[32] and with the junction resistance expressed via the averaged transparency $R_N^{-1} = e^2 k_f \overline{\mathcal{D}}/4\pi^2\hbar$. Thus for dirty junctions one obtains $\tilde{g}_{12} = 0.75\overline{\mathcal{D}}\xi(T)/\ell$, while for pure junctions $\tilde{g}_{12} = 3\pi^2\overline{\mathcal{D}}\xi(T)/[14\zeta(3)\xi_0] = 1.76\overline{\mathcal{D}}\xi(T)/\xi_0$. Here ℓ is the mean free path and $\xi_0 = \hbar v_f/\pi T_c$ is the zero-temperature coherence length. The quantitative microscopic formulas obtained here for $\tilde{g}_{12}$ agree with the earlier estimates.[26] In particular, in dirty superconductors the ratio $\xi(T)/l$ can easily reach 100 even at low temperatures. Hence, for small and moderate transparencies the quantity $\tilde{g}_{12} = 0.75\overline{\mathcal{D}}\xi(T)/l$ can vary from vanishingly small values in the tunneling limit considered in this paper to those well exceeding 100 near T_c, when a substantial anharmonic behavior of the Josephson current takes place.[26]

V. NEXT ORDER TERMS IN THE CURRENT

The initial expression (5) for the supercurrent can be generalized to include the next order terms, which originate from the phase-dependent biquadratic contributions to Eq. (2). The resulting formula is obtained after replacing $\tilde{g}_{12} \to \tilde{g}_{12} + \tilde{\eta}_1(|a|/b)f_{10}^2 + \tilde{\eta}_2(|a|/b)f_{20}^2$ in Eq. (5) and adding $\tilde{j}_{f_{12}} = \tilde{j}_{c2,f}\sin 2\chi$, where

$$\tilde{j}_{c2,f} = (3\sqrt{3}/2)(|a|/b)\tilde{f}_{12} f_{10}^2 f_{20}^2. \qquad (16)$$

Here $\tilde{f}_{12} = f_{12}\xi(T)/K$, $\tilde{\eta}_i = \eta_i\xi(T)/K$, $i = 1,2$. Substituting the zeroth-order quantities (7) in Eq. (16), one obtains

$$\tilde{j}_{c2,f} = (3\sqrt{3}/8)(|a|/b)\tilde{f}_{12}\left(\sqrt{2 + \tilde{g}_{11}^2} - \tilde{g}_{11}\right)^2 \times \left(\sqrt{2 + \tilde{g}_{22}^2} - \tilde{g}_{22}\right)^2. \qquad (17)$$

Both contributions to the second harmonic (10) and (17) are of the second order in transparency $\propto\mathcal{D}^2$, but (17) also contains an additional small parameter $|\tau| = (T_c - T)/T_c$, since $|a| = \alpha|\tau|$. This allows us to disregard (17) in studying the regular problem assumed above. However, in a number of specific cases the coupling constant g_{12} can vanish for symmetry reasons.[33–35] This concerns, in particular, the asymmetric junction between identical $d_{x^2-y^2}$-wave superconductors with exact (100) and (110) interface-to-crystal orientations on opposite banks of a smooth plane interlayer.[11–15,36–41] An additional element of the point symmetry inherent in such a specific system is the reflection in the xz plane perpendicular to the interface. Free energy should be invariant under the latter transformation, while the d_{xy}-wave order parameter on one side of the interface changes its sign and the $d_{x^2-y^2}$-wave order parameter on another side keeps its value unchanged. Then the expression containing $|\Psi_1 - \Psi_2|^2$ in Eq. (2) is no longer invariant and, therefore, the coefficients $\tilde{g}_{12}$, $\tilde{\eta}_1$, and $\tilde{\eta}_2$ should vanish in the case in question. By contrast, the term containing $|\Psi_1^2 - \Psi_2^2|^2$ in Eq. (2) remains unchanged under the sign reversal of one of the order parameters and, hence, the coefficient f_{12} can maintain its regular value.

In reality, the first harmonic $\tilde{j}_{c1}$ remains finite and, along with $\tilde{j}_{c2,f}$, still represents a substantial part of the supercurrent, mainly due to interfacial imperfections such as faceting, roughness, etc.[11–15] Since $|j_{c2,f}| \ll j_{\rm dp}$, the relation $|j_{c1}| \lesssim |j_{c2,f}|$ always results in the condition $|j_{c2,f}| j_{\rm dp} \gg j_{c1}^2$, which consequently loses its importance in the special case of strongly suppressed g_{12}.

VI. NEXT ORDER TERMS IN THE BC

Let the parameters g_{12} and g_{ii} $(i = 1,2)$ be independent of T near T_c. Since $\tilde{g}_{12},\tilde{g}_{ii} \propto \xi(T)$, then close to T_c one will get $|\tilde{g}_{12}| \gg 1$ and/or $|\tilde{g}_{ii}| \gg 1$ due to large values of $\xi(T)$. However, the coupling constants $|g_{12}|$ and $|g_{ii}|$ can themselves be very small and the temperature range with large $|\tilde{g}_{12}|$ and/or $|\tilde{g}_{ii}|$ be too narrow. While the condition $|\tilde{g}_{12}| \ll 1$, resulting in the tunneling behavior, is assumed throughout this paper, the range of variations of $\tilde{g}_{ii}$, defined by the strength of interfacial proximity effects, is quite wide. It contains, for instance, small values of $|g_{ii}|$. For this reason numerical coefficients of the order of unity, originating from Eq. (3), have been kept in Eqs. (6)–(11) on an equal footing with $\tilde{g}_{ii}^2$. However, the additional terms of the next order of smallness, which come from the BC, can be comparable with the terms referred to above and should generally be taken into account.

To clarify the point, let us represent the BC schematically as $(df_i/d\tilde{x})_0 \approx \tilde{\mathcal{A}}_{i,0} + (|a|/b)\tilde{\mathcal{A}}_{i,1}$ $(i = 1,2)$. Here $\tilde{\mathcal{A}}_{i,0} = \mathcal{A}_{i,0}\xi(T)/K$ is linear in the order parameters and coincides with the right-hand side of Eq. (4). The correction $\tilde{\mathcal{A}}_{i,1} = \mathcal{A}_{i,1}\xi(T)/K$ appears in the BC both from the quartic and biquadratic terms of the interface free energy (2) and from the weak temperature dependence of the GL coefficients in $\mathcal{A}_{i,0}$. Therefore, in addition, it involves the temperature derivatives of the coefficients. As (3) contains $(df/d\tilde{x})^2$, let us consider $(df_i/d\tilde{x})_0^2 \approx \tilde{\mathcal{A}}_{i,0}^2 + 2(|a|/b)\tilde{\mathcal{A}}_{i,0}\tilde{\mathcal{A}}_{i,1}$. Here it is the crossed product, which is the next order correction to the ith equation for the self-consistent order parameters. Since the expression $2(|a|/b)\tilde{\mathcal{A}}_{i0}\tilde{\mathcal{A}}_{i1} = 2|a|\xi^2(T)\mathcal{A}_{i0}\mathcal{A}_{i1}/(bK^2) = 2\mathcal{A}_{i0}\mathcal{A}_{i1}/(bK)$ depends on temperature solely via the order parameter amplitudes entering $\mathcal{A}_{i,1(0)}$, it results in temperature independent coefficients in the equations for the order parameters.

The corresponding terms should, in general, be taken into account for the quantitative description of the Josephson current. However, as the main contribution to Eq. (3) from $(df_i/d\tilde{x})_0^2$ is quadratic and the correction is linear in $\mathcal{A}_{i0}$, for sufficiently large $|\mathcal{A}_{i0}|$ the correction is negligibly small as compared to $\mathcal{A}_{i0}^2$. For sufficiently small $|\mathcal{A}_{i0}|$ the correction can now also be disregarded as compared to the coefficients of the order of unity in Eq. (3).

In tunnel junctions, the basic correction of the given origin is described by the crossed product $4g_{ii}h_{ii}/(Kb)$. In particular, in the zeroth approximation in the transparency the order parameters are

$$f_{i0}^{(0)^2} = \left[1 + \tilde{g}_{ii}^2 + \sqrt{\left(1 + \tilde{g}_{ii}^2\right)^2 - L_i}\,\right]^{-1}, \tag{18}$$

where $L_i = 1 - (4g_{ii}h_{ii})/(Kb)$ and $g_{ii}, h_{ii} > 0$. The quantities $\tilde{g}_{ii}^2 = g_{ii}^2/K|a|$ are implied here and below to involve g_{ii}^2 in the expanded form $g_{ii}^2 \approx g_{ii,c}^2 + 2\tau g_{ii,c}(dg_{ii}/d\tau)_c$, where weak temperature dependence of g_{ii} near T_c is taken into account in linear in τ approximation.

The linear in $\tilde{g}_{12}$ first harmonic is obtained by substituting (18) in Eq. (5). Calculating also the second harmonic, one obtains the modified relation between the second and the first harmonics:

$$\tilde{j}_{c2} = \frac{\tilde{j}_{c1}^2}{3\sqrt{3}} \sum_{i=1}^{2} \frac{1}{\sqrt{2 + \tilde{g}_{ii}^2}} \left[1 + \tilde{g}_{ii}^2 + \sqrt{\left(1 + \tilde{g}_{ii}^2\right)^2 - L_i}\,\right]. \tag{19}$$

In disregarding the term $(4g_{ii}h_{ii})/(Kb)$, Eqs. (18) and (19) are reduced to the previous ones, Eqs. (7) and (11). Since the two parameters $\tilde{g}_{ii} = g_{ii}\xi(T)/K$ and $4g_{ii}h_{ii}/(Kb)$ are independent of each other, the conditions $|\tilde{g}_{ii}| \ll 1$ do not generally exclude the special case $4|g_{ii}|h_{ii}/(Kb) \gtrsim 1$. Then quadratic in h_{ii} corrections can also be noticeable. However, for sufficiently small $|g_{ii}|$ the opposite conditions $[4|g_{ii}|h_{ii}/(Kb)]^{1/2} \ll 1$ occur and allow disregarding all the corresponding terms.

For large $\tilde{g}_{ii}$ the term $(4g_{ii}h_{ii})/(Kb)$ becomes negligibly small in Eq. (19), when the temperature-dependent condition $\xi(T)\tilde{g}_{ii}^3 \gg 4h_{ii}/b$, in keeping with $\tilde{g}_{ii} \gg 1$, is valid. Also, for one and the same g_{ii}, the right-hand side in Eq. (19) is always larger than that in Eq. (11), if $(g_{ii}h_{ii})/(Kb) > 0$. Therefore, the modified formulas do not alter the main statement of this paper.

In conclusion, a test for identification of a pronounced interfacial pair breaking in Josephson tunnel junctions has been proposed and theoretically verified in this paper, based on Eqs. (9)–(11) and (19) obtained within the self-consistent theory of the Josephson current. The main statement is that the condition $j_{c2} j_{\rm dp} \gg j_{c1}^2$ indicates a strong interfacial pair breaking at least on one side of the interface, if the first and the second harmonics satisfy the conventional relation $j_{c2} \ll |j_{c1}|$.

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APPENDIX: ORDER PARAMETER PROFILES NEAR IMPENETRABLE BOUNDARIES

The GL theory allows a detailed description of the spatial profiles of the order parameters near impenetrable boundaries. Here the boundaries, which either suppress or enhance the superconductivity in their vicinities, are considered jointly.

In the case in question the supercurrent vanishes and $f_\infty = 1$. Then Eq. (3) reduces to

$$\left(\frac{df(\tilde{x})}{d\tilde{x}}\right)^2 = \frac{1}{2}[1 - f^2(\tilde{x})]^2. \tag{A1}$$

The solution of Eq. (A1), which is relevant to the order parameter near a pair-breaking surface at $x = 0$, satisfies the condition $f < 1$ throughout the half space $x > 0$ and takes the form (see, e.g., Ref. 42)

$$f_{\rm pb}(\tilde{x}) = \tanh\left(\frac{\tilde{x}+\tilde{x}_0}{\sqrt{2}}\right). \quad \text{(A2)}$$

The parameter $\tilde{x}_0 > 0$ together with the associated order parameter value on the surface should be determined from the boundary conditions.

The expression for the order parameter in superconducting half space with the pair-producing surface directly follows from Eq. (A2), since for each $f(\tilde{x})$, which meets (A1), the function $1/f(\tilde{x})$ satisfies the same equation (A1). This results in the solution

$$f_{\rm pp}(\tilde{x}) = \coth\left(\frac{\tilde{x}+\tilde{x}_0}{\sqrt{2}}\right), \quad \text{(A3)}$$

for which the condition $f_{pp} > 1$ holds throughout the half space $x > 0$.

The order parameter f_0, taken on the surface and described by Eq. (7), can be alternatively determined by minimizing full free energy (1) and (2) with the solutions (A2) or (A3). Explicit integration in Eq. (1) with Eq. (A2) or (A3) results in the part of the bulk free energy modified by the boundary. Retaining only the quadratic in the order parameter term in the surface free energy (2), one finds the full free energy per unit square of an impenetrable surface

$$\mathcal{F} = \frac{\sqrt{K}|a|^{3/2}}{\sqrt{2}b}\left[\frac{4}{3} - 2f_0 + \frac{2}{3}f_0^3 + \sqrt{2}\tilde{g}f_0^2\right], \quad \text{(A4)}$$

for both solutions. The extremum of Eq. (A4) does result in Eq. (7) irrespective of the sign of g. The surface suppresses the superconducting order parameter at $g > 0$, while at $g < 0$ the superconductivity is enhanced near the surface.

It follows from Eqs. (A2) and (A3)

$$\frac{df_{\rm pb}(\tilde{x})}{d\tilde{x}} = \frac{1}{\sqrt{2}}\,{\rm sech}^2\left(\frac{\tilde{x}+\tilde{x}_0}{\sqrt{2}}\right), \quad \text{(A5)}$$

$$\frac{df_{\rm pp}(\tilde{x})}{d\tilde{x}} = -\frac{1}{\sqrt{2}}\,{\rm csch}^2\left(\frac{\tilde{x}+\tilde{x}_0}{\sqrt{2}}\right). \quad \text{(A6)}$$

As seen from Eq. (A5), the order parameter (A2), which is suppressed near the boundary, satisfies not only the condition $f_{\rm pb}(\tilde{x}) < 1$ but also the relation

$$\left|\frac{df_{\rm pb}(\tilde{x})}{d\tilde{x}}\right| \leqslant \frac{1}{\sqrt{2}}. \quad \text{(A7)}$$

For the order parameter (A3), which is enhanced near the boundary, one gets $f_{\rm pp}(\tilde{x}) > 1$. According to Eqs. (A3) and (A6), the smaller the parameter x_0, the larger both the order parameter $f_{\rm pp,0}$ and its derivative $|df_{\rm pp}/d\tilde{x}|_0$ taken on the boundary. A large spatial derivative $|df_{\rm pp}/d\tilde{x}|_0$ corresponds to a small characteristic scale induced in a superconductor in the vicinity of the surface.

For $g < 0$ and $|\tilde{g}| \gg 1$ one gets from Eqs. (4) and (7) $f_0 \approx \sqrt{2}|\tilde{g}|$ and $|df/d\tilde{x}|_0 \approx \sqrt{2}\tilde{g}^2$. Hence, the effective characteristic scale near the surface is $x_0 \sim \xi(T)/|\tilde{g}|$. For the use of the GL theory near the surface one assumes $x_0 \gg \xi_0$. This results in the condition $|\tilde{g}| \ll 1/\sqrt{\tau}$, i.e., $|g| \ll \sqrt{K\alpha}$.

[1] L. J. Buchholtz and G. Zwicknagl, Phys. Rev. B **23**, 5788 (1981).
[2] Y. S. Barash, A. V. Galaktionov, and A. D. Zaikin, Phys. Rev. B **52**, 665 (1995).
[3] M. Matsumoto and H. Shiba, J. Phys. Soc. Jpn. **64**, 1703 (1995); **64**, 3384 (1995); **64**, 4867 (1995); **65**, 2194 (1996) .
[4] Y. Nagato and K. Nagai, Phys. Rev. B **51**, 16254 (1995).
[5] L. J. Buchholtz, M. Palumbo, D. Rainer, and J. A. Sauls, J. Low Temp. Phys. **101**, 1079 (1995); **101**, 1099 (1995).
[6] M. Alber, B. Bäuml, R. Ernst, D. Kienle, A. Kopf, and M. Rouchal, Phys. Rev. B **53**, 5863 (1996).
[7] D. F. Agterberg, J. Phys.: Condens. Matter **9**, 7435 (1997).
[8] T. Tokuyasu, J. A. Sauls, and D. Rainer, Phys. Rev. B **38**, 8823 (1988).
[9] A. Cottet, D. Huertas-Hernando, W. Belzig, and Y. V. Nazarov, Phys. Rev. B **80**, 184511 (2009).
[10] H. Kimura, R. P. Barber, S. Ono, Y. Ando, and R. C. Dynes, Phys. Rev. B **80**, 144506 (2009).
[11] E. Il'ichev, V. Zakosarenko, R. P. J. IJsselsteijn, H. E. Hoenig, V. Schultze, H.-G. Meyer, M. Grajcar, and R. Hlubina, Phys. Rev. B **60**, 3096 (1999).
[12] T. Lindström, S. A. Charlebois, A. Y. Tzalenchuk, Z. Ivanov, M. H. S. Amin, and A. M. Zagoskin, Phys. Rev. Lett. **90**, 117002 (2003).
[13] T. Lindström, J. Johansson, T. Bauch, E. Stepantsov, F. Lombardi, and S. A. Charlebois, Phys. Rev. B **74**, 014503 (2006).
[14] H. Hilgenkamp and J. Mannhart, Rev. Mod. Phys. **74**, 485 (2002).
[15] A. A. Golubov, M. Y. Kupriyanov, and E. Il'ichev, Rev. Mod. Phys. **76**, 411 (2004).
[16] S. M. Frolov, D. J. Van Harlingen, V. A. Oboznov, V. V. Bolginov, and V. V. Ryazanov, Phys. Rev. B **70**, 144505 (2004).
[17] A. Buzdin, Phys. Rev. B **72**, 100501 (2005).
[18] E. Goldobin, D. Koelle, R. Kleiner, and A. Buzdin, Phys. Rev. B **76**, 224523 (2007).
[19] J. S. Langer and V. Ambegaokar, Phys. Rev. **164**, 498 (1967).
[20] H. J. Fink and W. C. H. Joiner, Phys. Rev. Lett. **23**, 120 (1969).
[21] I. N. Khlyustikov and A. I. Buzdin, Adv. Phys. **36**, 271 (1987).
[22] V. B. Geshkenbein, Zh. Eksp. Teor. Fiz. **94**, 368 (1988) [Sov. Phys. JETP **67**, 2166 (1988)].
[23] K. V. Samokhin, Zh. Eksp. Teor. Fiz. **105**, 1684 (1994) [JETP **78**, 909 (1994)].
[24] V. F. Kozhevnikov, M. J. V. Bael, P. K. Sahoo, K. Temst, C. V. Haesendonck, A. Vantomme, and J. O. Indekeu, New J. Phys. **9**, 75 (2007).

[25] The results of Ref. 26 concern only the pair-breaking effects in symmetric junctions ($f_0 < 1$), though the main equation (3) also applies to $f_0 > 1$. In the corresponding particular case $\tilde{g}_{11} = \tilde{g}_{22} \equiv g_\delta > 0$ Eqs. (9) and (10) of this paper are reduced to Eq. (5) of Ref. 26. It follows from Eq. (10) that the second harmonic is positive and does not change its sign under the sign reversal of g_{ii}.

[26]Y. S. Barash, Phys. Rev. B **85**, 100503 (2012).
[27]J. Bardeen, Rev. Mod. Phys. **34**, 667 (1962).
[28]M. Y. Kupriyanov and V. F. Lukichev, Fiz. Nizk. Temp. **6**, 445 (1980) [Sov. J. Low Temp. Phys. **6**, 210 (1980)].
[29]J. Romijn, T. M. Klapwijk, M. J. Renne, and J. E. Mooij, Phys. Rev. B **26**, 3648 (1982).
[30]J. M. E. Geers, M. B. S. Hesselberth, J. Aarts, and A. A. Golubov, Phys. Rev. B **64**, 094506 (2001).
[31]V. Ambegaokar and A. Baratoff, Phys. Rev. Lett. **10**, 486 (1963); **11**, 104 (1963).
[32]L. P. Gor'kov, Zh. Eksp. Teor. Fiz. **36**, 1918 (1959) [JETP **9**, 1364 (1959)]; **37**, 1407 (1959) [JETP **10**, 998 (1960)].
[33]V. B. Geshkenbein and A. I. Larkin, Pis'ma Zh. Eksp. Teor. Fiz. **43**, 306 (1986) [JETP Lett. **43**, 395 (1986)].
[34]S.-K. Yip, O. F. De Alcantara Bonfim, and P. Kumar, Phys. Rev. B **41**, 11214 (1990).
[35]M. Sigrist and K. Ueda, Rev. Mod. Phys. **63**, 239 (1991).
[36]M. Sigrist and T. M. Rice, J. Phys. Soc. Jpn. **61**, 4283 (1992).
[37]Y. Tanaka, Phys. Rev. Lett. **72**, 3871 (1994).
[38]S. Yip, Phys. Rev. B **52**, 3087 (1995).
[39]M. B. Walker and J. Luettmer-Strathmann, Phys. Rev. B **54**, 588 (1996).
[40]S. Östlund, Phys. Rev. B **58**, R14757 (1998).
[41]T. Yokoyama, Y. Sawa, Y. Tanaka, and A. A. Golubov, Phys. Rev. B **75**, 020502 (2007).
[42]P. G. de Gennes, *Superconductivity of Metals and Alloys* (Addison Wesley Publishing Co, Inc., Reading, MA, 1966) .