

# AC Josephson effect in the long voltage-biased SINIS junction

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Theory of non-stationary coherent effects is developed for superconductor-normal-superconductor (SNS) structures with relatively strong normal scattering on S/N interfaces (interface resistance is large compared to intrinsic resistance of N metal). Analytical expressions are found for the time-dependent anomalous Green functions induced in the N region under the fixed-voltage-bias. The amplitude of the current oscillations is determined in non-equilibrium conditions. Non-stationary correction to the distribution function is calculated in high-temperature limit and found to be slowly decreasing with the temperature, leading to the dominance of the second-harmonic term in the Josephson current,  $I_s(t) \propto \sin(4eVt)$  at high temperatures and low voltage.

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## 1. Coherence effects and general equations.

The superconducting hybrid (normal metal – superconductor) structures are very rich systems that have been studied for the past few decades both theoretically and experimentally (cf. [1] for a relatively recent brief review). The proximity effect has been shown to induce superconductive correlations into normal part of the system, where they decay over the length  $\xi_\epsilon = \sqrt{\hbar D/\epsilon}$  that can be large at low energies. One of the consequences of the proximity effect is the ability of the normal metal to carry out phase-sensitive current if length  $L$  of the junction is smaller than  $\xi_{\epsilon_0}$  at characteristic energies  $\epsilon_0$ . Equilibrium Josephson effect in diffusive SNS systems is well understood by now and can be well explained in terms of stationary Andreev levels or more directly through Green's functions approach. When the constant voltage is applied to the junction, nonstationary Josephson effect arises. If the voltage is high,  $V \gg E_{Th}$ , than effects of coherence between superconducting reservoirs can be neglected and current is stationary with the rich subgap structure [2]. When the voltage is not very high, time-dependent contribution to the current can become important, even at rather high temperatures [3–6].

In this paper we consider nonstationary Josephson effect in a long symmetric voltage-biased SNS junction with large interface resistance, so that  $r = R_B/R_N \gg 1$  (here  $R_B$  is the single barrier resistance and  $R_N$  the resistance of the normal wire). This problem was first considered in [7], where Josephson current up to the first order in  $r^{-1}$  was calculated. It was shown, that Josephson current decays exponentially when temperature is

high comparative to the inverse diffusion time. However, it was shown later [4], that due to nonequilibrium effects time-dependent current does decay as slow as  $T^{-1}$  at high temperatures. Here we microscopically calculate the current, with all nonequilibrium effects taken into account. In doing so, we have to include the terms which are formally of higher orders in  $r^{-1}$ ; however the resulting current is not necessary smaller than lowest-order term, but can dominate it, as explained below.

To describe this system we use Keldysh method, developed for superconductivity by Larkin and Ovchinnikov [8]. In this method the Green function is  $4 \times 4$  matrix in Keldysh and particle/hole  $2 \times 2$  spaces. This matrix  $\check{G}$  contains all the information about spectrum of the system and on the distribution of electrons over the energy levels. The resulting Usadel equation is written in terms of the disorder-averaged semiclassical Green function  $\check{G} = \check{G}(t_1, t_2, \mathbf{r})$  (for the detailed review, see [9]). This equation is presented below, it can be used to describe any nonstationary phenomena with low energy scales involved (compared to Fermi energy). It involves time convolution operation:  $(f \circ g)(t_1, t_2) = \int_{-\infty}^{\infty} f(t_1, t)g(t, t_2)dt$ . It's convenient to introduce  $t = (t_1 + t_2)/2$ ,  $\tau = t_1 - t_2$ . Then Usadel equation in the absence of the vector potential (which is supposed to be zero throughout the paper) and electron pairing reads as follows (we work in units  $e = k_B = \hbar = 1$ ):

$$-D\partial_x(\check{G} \circ \partial_x \check{G}) + \partial_\tau[\check{\sigma}^3, \check{G}] + \frac{1}{2}\partial_T\{\check{\sigma}^3, \check{G}\} + i\varphi_- \check{G} = \check{I}, \quad (1)$$

with  $\check{\sigma}^3 = \check{1}\hat{\tau}^3$  and  $\check{I} = -i(\check{\Sigma}_{in} \circ \check{G} - \check{G} \circ \check{\Sigma}_{in})$  where  $\check{\Sigma}_{in}$  is self-energy matrix, that accounts for inelastic

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and dephasing processes. Time-dependent electric potential  $\varphi(t)$  enters this equation through  $\varphi_-(t_1, t_2) = \varphi(t_2) - \varphi(t_1)$ . It has to be determined self-consistently via electroneutrality condition  $\varphi(t) = \frac{\pi}{4} \text{tr} \hat{G}^K(t, t)$  [9].

Keldysh Green function  $\check{G}$  obeys normalization condition  $\check{G} \circ \check{G} = \check{1} \delta(t_1 - t_2)$  which allows for the ansatz  $\hat{G}^K = \hat{G}^R \circ \hat{h} - \hat{h} \circ \hat{G}^A$  where  $\hat{h}$  stays for the matrix distribution function. The latter can be chosen to be diagonal:  $\hat{h} = h_0 \hat{\tau}^0 + h_3 \hat{\tau}^3$ . It is convenient to implement Fourier transform over  $\tau$ , i.e. to pass to Wigner representation. This way, we write  $\check{G}(\tau, t) = \int \check{G}(\epsilon, t) e^{-i\epsilon\tau} d\epsilon$ . We note that the convolution operator simplifies in the mixed representation due to the following identity: for  $f(\epsilon, t) = e^{-i\omega_1 t} f(\epsilon)$ ,  $g(\epsilon, t) = e^{-i\omega_2 t} g(\epsilon)$ , one gets

$$(f \circ g)(\epsilon, t) = e^{-i(\omega_1 + \omega_2)t} f\left(\epsilon + \frac{\omega_2}{2}\right) g\left(\epsilon - \frac{\omega_1}{2}\right).$$

Our goal is to find  $\hat{G}^R$ ,  $\hat{G}^A$ ,  $\hat{G}^K$  for the SINIS structure and to calculate the current. Current density is expressed through Keldysh component of the matrix current  $\check{j} = \check{G} \circ \nabla \check{G}$  as follows:

$$I(t) = \frac{\pi \sigma_N}{4} \text{tr} \hat{\tau}^3 \check{j}^K(t, t). \quad (2)$$

Matrix equation (1) has to be supplemented with the Kupriyanov-Lukichev [10] boundary condition at both SIN interfaces. At the right interface it takes the form:

$$2R_{SN} \sigma_N \check{j} = [\check{G}_o, \check{G}_r], \quad (3)$$

with  $R_{SN}$  being the interface resistance of the barrier per unit area in the normal state. We use indices  $l, r$  for the left and right reservoirs.

Now we have to work out retarded, advanced and Keldysh components of equations (1), (3). In the explicit form, they are presented in the Appendix. Retarded and advanced components determine the generalized spectral properties of the system and Keldysh components describe distribution of electrons over this generalized spectrum. In what follows, we simplify these equations using the smallness of the parameter  $r^{-1}$ , as described in the next section.

We consider the constant-voltage-biased setup and neglect the spatial dependencies of all the relevant quantities in the directions, perpendicular to the wire. We measure the length in units of  $L$ , assuming that bulk superconductors are situated at points  $x = -1/2$  and  $x = 1/2$  at the voltages  $-V/2$  and  $V/2$  correspondingly. We suppose that bulk superconductors are good reservoirs at temperature  $T_S$ ; under this assumption their Green functions  $\check{G}_{l,r}$  can be obtained from the standard BCS form  $\check{G}_{BCS}$  by the gauge transformation

$\check{G}_{l,r}(t_1, t_2) = \check{S}_{l,r}(t_1) \check{G}_{BCS}(t_1, t_2) \check{S}_{l,r}^+(t_2)$ , with  $\hat{G}_{BCS}^{R(A)} = (g_S \hat{\tau}^3 + f_S \hat{\tau}^1)^{R(A)}$ ,  $\hat{G}_{BCS}^K = \tanh(\frac{\epsilon}{2T_S}) (\hat{G}_{BCS}^R - \hat{G}_{BCS}^A)$  and  $\check{S}_{l,r}(t) = \exp(\pm \frac{iVt}{2} \hat{\tau}^3) \check{1}$ . Straightforward calculation gives:

$$\hat{G}_{l,r}^R(t, \epsilon) = \begin{pmatrix} g_S^R(\epsilon \mp V/2) & e^{\mp iVt} f_S^R(\epsilon) \\ \epsilon^{\pm iVt} f_S^R(\epsilon) & -g_S^R(\epsilon \pm V/2) \end{pmatrix} \quad (4)$$

and

$$\hat{h}_{l,r}(\epsilon) = \begin{pmatrix} \tanh\left(\frac{\epsilon \mp V/2}{2T_S}\right) & 0 \\ 0 & \tanh\left(\frac{\epsilon \pm V/2}{2T_S}\right) \end{pmatrix}. \quad (5)$$

The explicit form of the energy-dependent functions  $g_S^{R(A)}$ ,  $f_S^{R(A)}$  is:

$$g_S^{R(A)}(\epsilon) = \frac{\epsilon}{\Delta} (\pm \eta_S - i\xi_S), \quad f_S^{R(A)}(\epsilon) = \xi_S \pm i\eta_S, \quad (6)$$

where

$$\eta_S = \frac{\Delta \text{sign} \epsilon}{\sqrt{\epsilon^2 - \Delta^2}} \theta(|\epsilon| - \Delta), \quad \xi_S = \frac{\Delta}{\sqrt{\Delta^2 - \epsilon^2}} \theta(\Delta - |\epsilon|). \quad (7)$$

**2. Spectral functions.** Here we calculate Green functions  $\hat{G}^{(R,A)}$  which describe proximity effect in normal region. We write matrix Green function  $\hat{G}^{R(A)}(x, \epsilon, t)$  in the form

$$\hat{G}^{R(A)} = \begin{pmatrix} \pm(1 - g_1^{R(A)}) & f_1^{R(A)} \\ f_2^{R(A)} & \mp(1 - g_2^{R(A)}) \end{pmatrix}. \quad (8)$$

Since the proximity effect is weak due to the presence of barriers, we can expand the equations for  $G^{(R,A)}$  in powers of small parameter  $r^{-1}$ . To the first order one finds:

$$g_{1,2}^R \sim r^{-2}, \quad f_{1,2}^{R,A} \sim r^{-1}. \quad (9)$$

Besides, we assume here that  $\varphi_- \sim r^{-2}$ , which is self-consistent assumption. Smallness of  $\varphi$  is due to the fact that almost all voltage drops at the barriers at  $r \gg 1$ . We consider Eqs.(35), (36) of Appendix and keep terms, proportional to  $r^{-1}$  in the equations, and terms of the order of unity in the boundary conditions for anomalous Green function  $f_1^R(x, \epsilon, t)$ , to obtain:

$$\begin{aligned} \partial_x^2 f_1^R + \kappa^2 f_1^R &= 0 \\ \partial_x f_1^R|_{x=\pm \frac{1}{2}} &= \pm r^{-1} e^{\pm iVt} f_S^R \end{aligned} \quad (10)$$

Here

$$\kappa_\epsilon = \sqrt{\frac{2i\epsilon}{E_{Th}} - \gamma} \quad (11)$$

and  $\gamma = (\tau_{in} E_{Th})^{-1}$  is the dimensionless inelastic scattering rate. Linear approximation leading to Eqs.(10) is applicable at all energies  $\epsilon$ , if  $(\gamma r)^{-1} \sim \tau_{in} E_g \ll 1$  (here  $E_g$  is the proximity-induced minigap [11] at  $\gamma = 0$ ). Other Green functions are expressed via relations

$$\begin{aligned} f_{1,2}^A(x, \epsilon, t) &= f_{1,2}^R(x, -\epsilon, t), \\ f_2^{R(A)}(x, \epsilon, t) &= f_1^{R(A)}(x, \epsilon, -t). \end{aligned} \quad (12)$$

The above equations are easy to solve, with the following result:

$$f_1^R = v^R \left[ e^{iVt} \cos \kappa_\epsilon \left( x + \frac{1}{2} \right) + e^{-iVt} \cos \kappa_\epsilon \left( x - \frac{1}{2} \right) \right]. \quad (13)$$

For the future convenience, we introduce definitions

$$\begin{aligned} u^R &= -\frac{f_S^R(\epsilon)}{r} u(\epsilon), \quad v^R = -\frac{f_S^R(\epsilon)}{r} v(\epsilon), \\ u^A &= -\frac{f_S^A(\epsilon)}{r} u(-\epsilon), \quad v^A = -\frac{f_S^A(\epsilon)}{r} v(-\epsilon), \end{aligned} \quad (14)$$

with

$$u(\epsilon) = \frac{\cos \kappa_\epsilon}{\kappa_\epsilon \sin \kappa_\epsilon}, \quad v(\epsilon) = \frac{1}{\kappa_\epsilon \sin \kappa_\epsilon},$$

so that

$$f_{1,2}^{R(A)} \left( x = \frac{1}{2} \right) = u^{R(A)} e^{\pm iVt} + v^{R(A)} e^{\mp iVt}. \quad (15)$$

Retarded and advanced normal Green functions can be found from the normalization condition:

$$g_1^R = \frac{1}{2} f_1^R \circ f_2^R, \quad g_2^R = \frac{1}{2} f_2^R \circ f_1^R. \quad (16)$$

Electric potential is

$$\varphi(t) = \frac{1}{2} \int (h_3 + g_\varphi^R \circ h_0 - h_0 \circ g_\varphi^A) d\epsilon, \quad (17)$$

with  $g_\varphi^R = \frac{1}{8} [f_2^R \circ, f_1^R]$ . Linearization over  $f_{1,2}$  breaks down at energies too close to the gap  $\Delta$ :  $|\epsilon - \Delta| \leq \leq \frac{E_{Th}}{r} \max(E_{Th}/\Delta, 1)$ , but the resulting square-root singularity  $|\epsilon - \Delta|^{-1/2}$  does not influence our results.

**3. Kinetic equation and its solution.** In this section we determine the electron distribution function. Its form is determined by interplay of several processes. The electrons diffuse from one superconducting lead to another during characteristic time  $\tau_D = E_{Th}^{-1}$ , and are subjected to normal and Andreev scattering at the

boundaries. Electron-electron scattering tends to thermalize them to equilibrium with some effective temperature  $T_e$ . Electron-phonon scattering tends to bring  $T_e$  closer to the substrate temperature  $T_S$ , thus taking the energy out of electron system. Another (and more effective at low temperatures) channel of electron cooling is the tunneling of hot electrons to superconducting reservoirs [12]. In the SINIS junction, the role of inelastic scattering is relatively large, since Andreev reflections are suppressed due to weakness of the proximity effect, so that electron spends a long time, bouncing back and forth between the boundaries. At  $\chi = \gamma r^2 \gg 1$  the electron distribution thermalizes and reads

$$\hat{h}(x, \epsilon, t) = h_0^{(0)}(\epsilon) \hat{\tau}^0 + O(\chi^{-1}), \quad (18)$$

where  $h_0^{(0)}(\epsilon) = \tanh(\frac{\epsilon}{2T_e})$  is equilibrium distribution with some effective temperature  $T_e$ ; in general,  $T_e \neq T_S$ . Effective temperature  $T_e$  has to be determined from the heat balance equation (cf. e.g. [13]). After that is done, one can calculate non-equilibrium correction to the distribution function (the second term in (18)). Note, that it can be important in terms of calculating the Josephson current, even if it small. The point is that thermal distribution leads (see below) to the amplitude  $|I_s|$  of the  $ac$  Josephson current  $I(t)$ , which is exponentially small in  $L/\xi_T$ . On the other hand, non-equilibrium corrections decay with temperature and length much slower. These non-equilibrium corrections are rather interesting, since they arise due to coherent Andreev reflections.

In order to find this nonequilibrium correction, we simplify general kinetic eqs. (37), (39) of Appendix by separating terms of different orders over small parameter  $r^{-1}$ , and adopting the simplified form of collision integral, i.e. “ $\tau$ -approximation”. In doing so, one has to consider the boundary condition up to the  $r^{-1}$  terms and kinetic up to the  $r^{-2}$  terms. Besides, these are small energies  $\epsilon \ll \Delta$ , where deviations from the equilibrium are important, so we put  $\eta_S(\epsilon) = 0$ ,  $\xi_S(\epsilon) = 1$ . We look for the distribution function in the following form:

$$h_0(x, \epsilon, t) = h_0^{(0)}(\epsilon) + r^{-2} h_0^{(2)}(\epsilon, x, t), \quad (19)$$

$$h_3(x, \epsilon, t) = r^{-2} h_3^{(2)}(\epsilon, x, t).$$

Next, due to the spatial symmetry,  $h_3^{(2)}$  and  $h_0^{(2)}$  are odd and even functions of  $x$ , correspondingly. Taking this into account, we write boundary conditions only at the  $x = 1/2$  boundary, to obtain with the necessary accuracy:

$$4\partial_x h_{0,3}^{(2)}|_{x=\frac{1}{2}} = J_1 \mp J_2, \quad (20)$$

with  $J_{1,2}$  terms which depend on  $h_0^{(0)}$  only, and are known therefore:

$$J_{1,2} = r (f_{1,2}^R \circ e^{\mp iVt} X + e^{\pm iVt} X \circ f_{2,1}^A), \quad (21)$$

with  $X(\epsilon) = \frac{1}{2} (h_{0+}^{(0)} - h_{0-}^{(0)})$ .

We used here special notation for the ‘‘energy-shift’’ subscript: for any function of energy  $f(\epsilon)$  we define

$$f_{\pm}(\epsilon) = f\left(\epsilon \pm \frac{V}{2}\right) \quad f_{++} = f(\epsilon + V) \quad f_{--} = f(\epsilon - V). \quad (22)$$

Since the boundary conditions are known, we proceed with the kinetic equation. Weakness of the proximity effect allows to neglect modifications of the diffusion coefficient, and also the terms, mixing  $h_0$  and  $h_3$  in the matrix equation (37) of Appendix. Adopting  $\tau$ -approximation for the collision integral, we obtain

$$E_{Th} \partial_x^2 h_0^{(2)} - (\partial_t h_0^{(2)} + \tau_{in}^{-1} h_0^{(2)}) = 0,$$

$$E_{Th} \partial_x^2 h_3^{(2)} - (\partial_t h_3^{(2)} + \tau_{in}^{-1} h_3^{(2)} + ir^2 \varphi_- h_0^{(0)}) = 0. \quad (23)$$

Kinetic equations (23) are to be written for each harmonic (in terms of the total time  $t$ ) of the distribution function, with the electric potential term that couples  $h_0^{(2)}$  and  $h_3^{(2)}$  due to the self-consistency condition (see (17)). In the limit of large temperatures  $T_e \gg E_{Th}$ , when the nonequilibrium contribution to the current becomes important, this term is exponentially small in  $L/\xi_T$  and we neglect it. Besides, if  $V \ll T_e$ , one has  $X = V/4T_e$ .

We look for the nonequilibrium correction in the following form:

$$h_0^{(2)}(x, \epsilon, t) = \sum_{n=0, \pm 1} A_{n, \epsilon} \cos(\kappa_n V x) e^{-2iVnt}, \quad (24)$$

$$h_3^{(2)}(x, \epsilon, t) = \sum_{n=0, \pm 1} B_{n, \epsilon} \sin(\kappa_n V x) e^{-2iVnt}.$$

Boundary conditions (20) allow to calculate  $A$ ,  $B$ :

$$A_{n, \epsilon} = -\frac{r V}{16 T_e} \frac{\tilde{A}_{n, \epsilon}}{\kappa_n V \sin\left(\frac{1}{2} \kappa_n V\right)}, \quad (25)$$

$$B_{n, \epsilon} = \frac{r V}{16 T_e} \frac{\tilde{B}_{n, \epsilon}}{\kappa_n V \cos\left(\frac{1}{2} \kappa_n V\right)},$$

with

$$\tilde{A}(n, \epsilon) = (\alpha_+ - \alpha_-) \delta_{n,0} + (v_+^R - v_-^A) \delta_{n,1} - (v_-^R - v_+^A) \delta_{n,-1},$$

$$\tilde{B}(n, \epsilon) = (\alpha_+ + \alpha_-) \delta_{n,0} + (v_+^R + v_-^A) \delta_{n,1} + (v_-^R + v_+^A) \delta_{n,-1},$$

where  $\alpha = u^R + u^A$ .

**4. Time-dependent current.** With the distribution functions (18) being determined, we turn to the calculation of time-dependent electrical current. It is convenient to calculate it in the vicinity of the right boundary, with  $\hat{j}^K$  in (2). There are lot of terms in (2), but we keep only those of them, that are proportional to the first order contribution to the anomalous function  $\hat{G}^{R(A)}$  and contribute to the oscillating part of the current. This allows us to get the leading terms at both low and high effective temperatures comparative to  $E_{Th}$ . More precise treatment will give some corrections, that are of the higher orders of  $r^{-1}$  at low temperatures and exponentially suppressed at high temperatures. In the limit of large superconducting gap  $\Delta \gg \max(E_{Th}, V)$  the result reads

$$I(t) = \frac{1}{8R} \int [I_{\epsilon}^+(t) - I_{\epsilon}^-(t)] d\epsilon \quad (26)$$

with

$$I_{\epsilon}^{\pm}(t) = (K_{1,2} \circ e^{\mp iVt} + e^{\mp iVt} \circ K_{1,2}) \quad (27)$$

with  $R$  being the resistance of the SINIS junction in the normal state:  $R = 2R_B$ . We have introduced here  $K_{1,2} = f_{1,2}^R \circ h_2 - h_1 \circ f_{1,2}^A$ , where anomalous and distribution functions are supposed to be calculated at  $x = 1/2$ . To proceed, we note that  $I(t)$  it can be considered as a sum of two different contributions, according to (18).

**Equilibrium current.** It reads:  $I_{eq} = \Re[e^{-2iVt} I_1]$  with

$$I_1 = -\frac{1}{2Rr} \int [v(\epsilon + V/2) - v(-\epsilon + V/2)] \tanh \frac{\epsilon}{2T_e} d\epsilon. \quad (28)$$

We note that at zero-voltage limit one has  $h_0^{(0)} = \tanh(\epsilon/2T_s)$  and for the amplitude  $I_1$  we get usual result [7] for the equilibrium critical current:

$$I_1(V=0) = -\frac{i}{Rr} \int h_S \Im v d\epsilon.$$

In general, the integral (28) can be reduced to the sum over residues to give

$$I_1 = -\frac{2\pi i T_e}{Rr} \sum_{n=1}^{\infty} \frac{1}{q_n \sinh q_n},$$

where

$$q_n = \sqrt{\frac{2(2n-1)\pi T_e - iV}{E_{Th}} + \gamma}.$$

In the limit of  $T_e \gg E_{Th}$  and  $\gamma \leq 1$  we obtain

$$|I_1| = c \frac{T_e}{rR\sqrt{a}} e^{-\sqrt{a}} \begin{cases} a = \frac{2\pi T_e}{E_{Th}} & c = 4\pi, \quad V \ll T_e \\ a = \frac{V}{2E_{Th}} & c = 2\sqrt{2}\pi, \quad V \gg T_e \end{cases}. \quad (29)$$

At very low temperatures  $\tanh(\epsilon/2T_e) \rightarrow \text{sign } \epsilon$  and the integral for the current can be done explicitly:

$$|I_1| = \frac{2E_{Th}}{rR} \left| \ln \cot \left( \frac{1}{2} \sqrt{\frac{iV}{E_{Th}} - \gamma} \right) \right|. \quad (30)$$

The above expression is valid for any voltage if  $T\tau_{in} \ll 1$  and for high voltages  $V\tau_{in} \gg 1$  otherwise.

**Nonequilibrium current.** Calculation starting from Eq.(26) leads to:

$$I_{neq} = \Re[e^{-2iVt}\Phi_1 + e^{-4iVt}\Phi_2], \quad (31)$$

with complex amplitudes  $\Phi_{1,2} = -\frac{\pi E_{Th}}{16Rr^3} \frac{V}{T_e} \phi_{1,2}$ , where  $\phi_{1,2}$  are given by:

$$\begin{aligned} \phi_1 &= \Gamma_0(x_0 - y_0) - \Gamma_V(x_0 + y_0) + (x_V + y_V)(\Gamma_{2V} - \Gamma_V), \\ \phi_2 &= -(x_V + y_V)\Gamma_{2V}, \end{aligned} \quad (32)$$

with

$$x_\epsilon = -\frac{\cot(\kappa_\epsilon/2)}{\kappa_\epsilon}, \quad y_\epsilon = \frac{\tan(\kappa_\epsilon/2)}{\kappa_\epsilon},$$

$$\Gamma_\epsilon = \frac{1}{\sqrt{2(i\epsilon/E_{Th} - \gamma)} \sin \sqrt{2(i\epsilon/E_{Th} - \gamma)}}.$$

The results (32) are applicable for any values of  $V/E_{Th}$  ratio, but in the lowest order over  $V/T_e$ .

Here we analyze the case of weak inelastic scattering  $\gamma \ll 1$ . In the limit of zero voltage,  $|\phi_2| = 2\gamma^{-1}|\phi_1| \gg |\phi_1|$  and the second harmonic dominates. For small voltages  $V \ll E_{Th}$  one has:

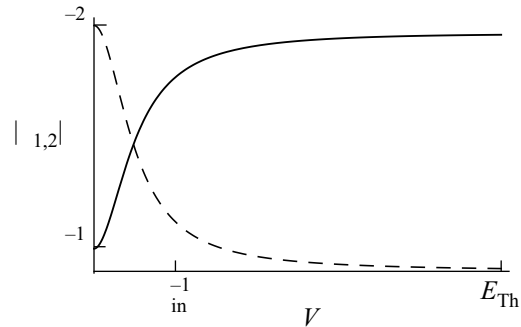
$$\Phi_1 = -\frac{\pi E_{Th}}{16Rr^3} \frac{V}{T_e} \begin{cases} \frac{1}{2}\gamma^{-1} + 4\gamma^{-4}(V/E_{Th})^2, & V \ll \tau_{in}^{-1} \\ -\gamma^{-2}, & \tau_{in}^{-1} \ll V \end{cases} \quad (33)$$

and

$$\Phi_2 = -\frac{\pi E_{Th}}{16Rr^3} \frac{V}{T_e} \begin{cases} \gamma^{-2}, & V \ll \tau_{in}^{-1} \\ -\frac{1}{4}(E_{Th}/V)^2, & \tau_{in}^{-1} \ll V \end{cases}. \quad (34)$$

Comparing first lines of Eqs.(34) and (29) we find that non-equilibrium second harmonics dominates the  $ac$  current at  $T_e \geq E_{Th} \ln^2(\gamma r)$  and low voltages  $V \ll \tau_{in}^{-1}$ . Similar phenomenon was observed in Refs. [14, 15], where subharmonic Shapiro steps with slowly decreasing (upon  $T$  increasing) amplitudes were found. Qualitative theory of this phenomenon was proposed by Argaman [4], who discussed it in terms of time-dependent Andreev bound states with non-equilibrium population. Our result (34) contains the same  $V/T_e$  dependence at low  $V$  as found in Ref. [4]; however, we got  $I_{neq} \sim \chi^{-1} I_{neq}^{Argaman}$ . We expect therefore that the result of [4] is valid (up to numerical factor of order unity) under the condition  $\chi \leq 1$ , which we do not consider here. To understand the origin of the whole effect it is useful to note that the result [4] for the second harmonics is very similar to (9) of Ref. [6] where Debye relaxation contribution to the  $dc$  conductance of SNS junction was estimated. Moreover, preprint version of Ref. [6] contains the same kind of estimation for the SINIS junction, with the result  $\propto \tau_{in}^2$ , very much like our expression for  $\Phi_2$ . We believe therefore that non-equilibrium second harmonics of the current originates from the same Debye relaxation mechanism.

Non-equilibrium  $ac$  current beyond linear in  $V$  approximation never was calculated previously, to the best of our knowledge. Eqs.(33), (34) demonstrate that at not very low voltages non-equilibrium first harmonics of the current  $\Phi_1$  becomes comparable to  $\Phi_2$  and then exceeds it. Note also  $\pi$ -shift in phases of  $\Phi_{1,2}$  with growth of voltage. Technically,  $\Phi_1$  originates from the peaks in nonequilibrium stationary parts of distribution functions,  $h_{0,3}^{(2)}(\epsilon)$  at  $\epsilon = \pm V/2$ . These peaks result from the modulation of the spectral density with the Josephson frequency, cf. Eq.(15). For the particular value of  $\gamma = 0.2$ , the absolute values of amplitudes  $\phi_{1,2}(V)$  at low voltages  $V \leq E_{Th} \ll T_e$  are plotted in the Figure.



Nonequilibrium amplitudes  $|\phi_1(V)|$  (normal line) and  $|\phi_2(V)|$  (dashed line) are calculated with Eq.(32) for  $\gamma = 0.2$  and  $V \ll T_e$

**5. Conclusions.** In this paper we have considered the AC Josephson effect in a long SINIS junction in the case of temperature and voltage low with respect to the bulk gap  $\Delta$ . The main results are given in Eqs.(32)–(34) for non-equilibrium contribution to the current in the high-temperature range  $T_e \gg E_{Th}$ . At high electron temperatures  $T_e$  in the normal wire, the nonequilibrium contribution to the current becomes dominant. We found, that the *ac* current contains two harmonics: the first with basic Josephson frequency  $\omega_J = 2eV/\hbar$  and the second harmonic  $2\omega_J$ . Second harmonic has the largest amplitude at low voltage and high temperature. Note, that calculation of non-equilibrium effects at low temperatures is complicated due to the necessity to account for the time-dependent electric potential in the normal wire; we leave this problem for the future studies.

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#### Appendix

Here we explicitly write out nonstationary Usadel equation and boundary conditions. Retarded component of (1) takes the following form:

$$-D\partial_x(\hat{G}^R \circ \partial_x \hat{G}^R) + \partial_\tau[\hat{\tau}^3, \hat{G}^R] + \frac{1}{2}\partial_T\{\hat{\tau}^3, \hat{G}^R\} + i\varphi_- \hat{G}^R = \hat{I}^R, \quad (35)$$

with  $\hat{I}^R = -i[\hat{\Sigma}^R \circ, \hat{G}^R]$ . It is supplemented with retarded component of boundary condition (3):

$$2R_{SN\sigma N}j_l^R = [\hat{G}_l^R \circ, \hat{G}_r^R]. \quad (36)$$

Since advanced components are identical (with  $R \rightarrow A$  substitution), we proceed with kinetic equation, which results from the Keldysh component of Usadel equation:

$$-D\left[\partial_x\left(\partial_x \hat{h} - \hat{G}^R \circ \partial_x \hat{h} \circ \hat{G}^A\right) + \left(\hat{j}^R \circ \partial_x \hat{h} - \partial_x \hat{h} \circ \hat{j}^A\right)\right] +$$

$$+ \left(\hat{G}_+^R \circ \partial_T \hat{h} + i\hat{G}^R \circ \varphi_- \hat{h}\right) - \left(\partial_T \hat{h} \circ \hat{G}_+^A + i\varphi_- \hat{h} \circ \hat{G}^A\right) = \hat{I}^{St}, \quad (37)$$

with

$$\hat{G}_+^{R(A)} = \frac{1}{2}\left\{\hat{G}^{R(A)}, \hat{\tau}^3\right\},$$

$$\hat{I}^{St} = -i\left(\hat{G}^R \circ \hat{\sigma} - \hat{\sigma} \circ \hat{G}^A\right), \quad (38)$$

$$\hat{\sigma} = \hat{\Sigma}^R \circ \hat{h} - \hat{h} \circ \hat{\Sigma}^A - \hat{\Sigma}^K.$$

The Keldysh component of boundary conditions (3) reads

$$2R_{SN\sigma N}\left(\partial_x \hat{h}_l - \hat{G}_l^R \circ \partial_x \hat{h}_l \circ \hat{G}_l^A\right) = \left(\hat{G}_l^R \circ \hat{u} - \hat{u} \circ \hat{G}_l^A\right), \quad (39)$$

where  $\hat{u} = \hat{G}_r^R \circ \delta \hat{h} - \delta \hat{h} \circ \hat{G}_r^A$ ,  $\delta \hat{h} = \hat{h}_r - \hat{h}_l$ .

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