# Effect of electron-electron collisions on the phase transition and kinetics of nonequilibrium superconductors

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Moscow Engineering Physics Institute (Submitted 30 June 1982) Zh. Eksp. Teor. Fiz. 84, 223–229 (January 1983)

An explicit expression is obtained for the distribution function of excess quasiparticles, taking into account electron-electron collisions in nonequilibrium superconductors. It is shown that the character of the phase transition may change at a definite ratio of the electron-electron and electron-phonon interaction constants: the dependence of the order parameter on the power of the source becomes single-valued. In addition, diffusion instability and paramagnetism of the superconductors arise. The multiplication factor of the excess quasiparticles due to electronelectron collisions and to reabsorption of phonons is calculated.

PACS numbers: 74.20. - z

## INTRODUCTION

An electromagnetic field and tunnel injection lead to the generation of excess quasiparticles in superconductors. These quasiparticles can change the character of the phase transition.<sup>1,2</sup> Actually, as shown previously,<sup>3,4</sup> the dependence of the order parameter  $\Delta$  on the pump power  $\beta$  becomes doubly valued above some critical value  $\beta_c$ , while the transition becomes of first order and is accompanied by the formation of an inhomogeneous state due to coherence instability. The theoretical model (the so called  $E \mod 2^{2,5}$  describing these effects takes into account only the electronphonon energy relaxation of the quasiparticles, neglecting the electron-electron (*e-e*) collisions.

The effect of the *e-e* collisions on the form of the quasiparticle distribution function (QDF) near  $\Delta = 0$  ( $\Delta < \Delta_0, \Delta_0$ is the value of the order parameter  $\Delta$  at  $\beta = 0$ ) has been studied in Ref. 6, <sup>1)</sup> where attention was called to the possibility of a change in sign of the coherent increment of the QDF at sufficiently large values of C (C is the ratio of the constants of the *e-e* and electron-phonon interactions).

In the present work we have found the explicit form of the QDF both analytically and numerically with a computer over a wide range of C and have shown that in the case of C that is much greater than a certain value ( $C_{\rm cr} \sim 6.5$ ) the coherent increment changes sign, i.e., the so-called Ne model is realized.<sup>2,5</sup>

The character of the phase transition changes in this case; the dependence of  $\Delta$  on  $\beta$  becomes single-valued (Fig. 1) and conditions are established for the onset of the diffusion instability predicted in Refs. 9 and 10, and for the corresponding inhomogeneous state.

Suitable superconductors with  $C > C_{cr}$  are W, Ir, and Hf. Such superconductors as Ru, Ti, Os, Mo, Al, Zn, and Cd have high values of C ( $\sim 2-5$ ).

In addition, the coefficients of quasiparticles multiplication due to electron-electron and electron-phonon collisions are found; these coefficients determine the critical source power  $\beta_c$ .

### **1. BASIC EQUATIONS. STATEMENT OF THE PROBLEM**

The character of the phase transition of nonequilibrium superconductors is determined by the form of the excess-

quasiparticle distribution function  $n(\xi)$  ( $\xi$  is the energy of the quasiparticles, measured from the Fermi surface). Near the transition point ( $\Delta = 0$ ),  $n(\xi)$  can be represented in the form.<sup>3,6</sup>

$$n(\xi) = n_0(\xi) + n_1(\xi).$$
 (1)

Here  $n_0(\xi)$  is the QDF at the transition point  $(\Delta = 0, \beta = \beta_c), n_1(\xi)$  is the coherent correction, which is proportional to  $\Delta$  and is governed by the coherent factors in the kinetic equation.

As shown in Ref. 3, the equation for  $\Delta$  is

$$\frac{\beta - \beta_c}{\beta_c} = 2\zeta \int_{0}^{\infty} \frac{n_1}{\varepsilon} d\xi, \quad \zeta \sim 1,$$
(2)

and the sign of the quantity  $N_S$  (the number of superconducting electrons, which determines the stability of the system)

$$N_{s} = 1 + 2 \int_{0}^{\infty} \frac{dn}{d\varepsilon} d\xi = 2 \int_{0}^{\infty} \frac{dn_{1}}{d\varepsilon} d\xi$$
(3)

is determined by the sign and form of the function  $n_1$ . Here  $\varepsilon = (\xi^2 + \Delta^2)^{1/2}$ .

Upon neglect of the electron-electron collisions,  $n_1$  is negative, so that  $d\Delta / d\beta > 0$  [i.e., there is a non-single-valued  $\Delta (\beta)$  dependence] and  $N_S > 0.^3$ 

Account of the electron-electron collisions has been given in Ref. 6, where it was shown that the coherent correction takes the form



FIG. 1. Dependence of the order parameter on the source power  $\beta$ : 1—  $C < C_{cr}$ ; 2— $C = C_{cr}$ , 3— $C > C_{cr}$ .

$$n_{1} = \frac{\Psi}{2\tilde{a}_{2}}, \quad \Psi = -\frac{\Delta^{2}a_{1}}{\epsilon\pi}(\varphi_{ef} - C\varphi_{ec}), \quad (4)$$

$$\varphi_{ee} = \iint_{0}^{\infty} dx_1 \, dx_2 \, \frac{x_1^2 + x_2^2 + x_1 x_2}{(x_1 + x_2) \, x_1 x_2} \, \varphi_{ce}(x_1, x_2) \,, \tag{5}$$

$$\varphi_{ee}(x_1, x_2) = n_0(x_1 + x_2) (1 - n_0(x_1)) (1 - n_0(x_2)) - n_0(x_1) n_0(x_2) (1 - n_0(x_1 + x_2)),$$
(6)

$$a_{i} = \int_{0}^{\infty} d\xi \xi^{i} n_{0}(\xi), \quad \tilde{a}_{i} = a_{i} + CS_{ee}^{+}(n_{0}, 0, 0), \quad (7)$$

$$\varphi_{e_{i}} = \int_{0}^{\infty} dx x [n_{0}(x) - N_{0}(x) (1 - 2n_{0}(x))], \qquad (8)$$

$$x = \xi/a_1^{\prime/_2}, \quad C = b\omega_D^2/\pi\lambda E_F a_1^{\prime/_2},$$
 (9)

where a and b are dimensionless quantities of the order of unity, dependent on the matrix elements of the *e-e* interaction,  $\lambda$  is the constant of the electron-phonon interaction,  $\omega_D$ is the Debye frequency,  $E_F$  is the Fermi energy,  $N_0(x)$  is the equilibrium distribution function of the phonons; for  $S_{ee}^+$ , see Eq. (12).

The function  $\varphi_{ee}(x_1, x_2) > 0$  is proportional to the difference in the rates of recombination and creation of quasiparticles via the *e-e* interaction. In equilibrium, when  $n_0 = (e^x + 1)^{-1}$ ,  $\varphi_{ee}$  vanishes (just as  $\varphi_{ef}$ ). For the case  $C \ll 1$ , realized for a number of superconductors (see Ref. 1), the influence of the *e-e* collisions on  $n_1$  can be neglected.<sup>6</sup> If, however, C > 1 it is necessary to take into account  $\varphi_{ee}$ , which is determined by the function  $n_0(x)$  that satisfies the following equation (here, for simplicity, we have set  $N_0(x) = 0$ ,  $\varphi_{ef} = 1$ )<sup>11</sup>:

$$(1-n_0(x))S^+-n_0(x)S^-=Q(x), \quad \beta=\int_0^\infty dx Q(x), \quad (10)$$

$$S^{\pm} = S_{ef}^{\pm} + CS_{ee}^{\pm}, \quad S = S^{+} + S^{-},$$
 (11)

$$S_{ee}^{+} = \iiint_{0}^{\infty} dx_{1} dx_{2} dx_{3} \{n_{1}n_{2}n_{3}\delta(x-x_{1}-x_{2}-x_{3}) + 3n_{1}n_{2}(1-n_{3})\delta(x-x_{1}-x_{2}+x_{3}) + 3n_{1}(1-n_{2})(1-n_{3})\delta(x-x_{1}+x_{2}+x_{3}) \}, \qquad (12)$$

$$S_{cc} = \iiint dx_1 dx_2 dx_3 \{ (1-n_1) (1-n_2) (1-n_3) \delta(x-x_1-x_2-x_3) \}$$

$$+3(1-n_{1})(1-n_{2})n_{3}\delta(x-x_{1}-x_{2}+x_{3})$$
  
+3(1-n\_{1})n\_{2}n\_{3}\delta(x-x\_{1}+x\_{2}+x\_{3})\}, (13)

$$S_{ef}^{+} = \int_{x}^{x+\omega} dx_1 (x-x_1)^2 n_1, \qquad (14)$$

$$S_{ef}^{-} = \int_{L(x)}^{x} dx_1 (x - x_1)^2 (1 - n_1) + \int_{0}^{\omega - x} dx_1 (x + x_1)^2 n_1;$$
(15)  

$$\tilde{\omega} = \omega_D / a_1^{'h}, \quad n_i \equiv n_0 (x_i), \quad L(x) = \begin{cases} 0, & x < \tilde{\omega} \\ x - \tilde{\omega}, & x > \tilde{\omega} \end{cases}.$$

We obtain the expression for the critical power  $\beta_0$  with

account of the reabsorption of phonons  $[N(x) \neq 0]$  by integrating (10) with respect to x:

$$\boldsymbol{\beta}_{e} = \int_{0}^{\tilde{\omega}} dx_{1} \int_{0}^{\tilde{\omega} - x_{1}} dx_{2} (x_{1} + x_{2})^{2} \{ n_{0}(x_{1}) n_{0} (x_{2}) - N (x_{1} + x_{2}) (1 - n_{0} (x_{1}) - n_{0} (x_{2})) \} - \int_{0}^{\infty} dx_{1} \{ S_{ee}^{+} (1 - n_{0} (x_{1})) - S_{ee}^{-} n_{0} (x_{1}) \}.$$
(16)

Here N(x) is the nonequilibrium phonon distribution function.

## 2. QUASIPARTICLE-DISTRIBUTION FUNCTION

It has been shown earlier<sup>6,12</sup> that the function  $n_0(x)$  is very close to the equilibrium value  $n_T$  in the range  $x \leq 3$ . At  $x \geq 3$ , the falloff of  $n_0(x)$  with the energy becomes slower. In this range of interest to us, the integral of the *e-e* collisions can be represented in the form

$$S_{ec} = n_0 S_{ec}^{-} - (1 - n_0) S_{ec}^{+} = \frac{1}{2} x^2 n_0(x) - 3 \int_x^{\infty} dx_1 (x_1 - x) n_0(x_1)$$
(17)

with accuracy to terms of order  $(x)^{-2}$ . Then Eq. (10) takes the form

$$n_{0}(x) \int_{L(x)}^{x} dx_{1}(x-x_{1})^{2} - \int_{x}^{\tilde{\omega}+x} dx_{1}(x-x_{1})^{2} n_{0}(x_{1}) + C \left[ n_{0}(x) x^{2}/2 - 3 \int_{x}^{\infty} dx_{1}(x_{1}-x) n_{0}(x_{1}) \right] = Q(x).$$
(18)

Analysis of Eq. (18) shows that at energies  $x < \tilde{x}$ ,

$$\widetilde{x} = (2E_F \omega_D \pi \lambda / 3ba_i)^{\frac{1}{2}}, \tag{19}$$

we can neglect the *e*-*e* interaction, and at  $x > \tilde{x}$  we can neglect the electron-phonon interaction.

In the case of a broad source

$$Q(x) = \alpha \theta(\omega - xa_i^n) \tag{20}$$

[which is realized for a high-frequency electromagnetic field and tunnel injection to the symmetric junctions (SiS junctions)<sup>2</sup>] the solution of Eq. (18) has the following form:

a) at  $\omega > \tilde{x}$  (here and below,  $\omega \equiv \omega/a_1^{1/2}$ )

$$n_{0}(x) = \begin{cases} n_{\tau}(x), & 0 < x \leq 3, \\ \eta \omega^{2}(\widetilde{\omega}/x)^{4}, & 3 < x < \widetilde{\omega}, \\ 4\chi(\widetilde{x}-x) + \eta \omega^{2}, & \widetilde{\omega} \leq x < \widetilde{x}, \\ \eta \widetilde{x}^{4} \left(\frac{2x}{3\omega^{3}} + \frac{\omega^{2}}{x^{4}}\right), & \widetilde{x} \leq x \leq \omega, \\ n_{0}(\omega) \exp(\omega - x), & \omega < x < \infty; \end{cases}$$
(21)

b) at  $\omega < \tilde{x}$ ,

Here

$$n_{0}(x) = \begin{cases} n_{\tau}(x), & 0 < x \leq 3, \\ 4\chi\omega(\vec{\omega}/x)^{4}, & 3 < x < \vec{\omega}, \\ \chi[4(\omega-x)+3\vec{\omega}], & \vec{\omega} \leq x \leq \omega, \\ n_{0}(\omega)\exp(\omega-x), & \omega < x < \infty. \end{cases}$$
(22)

$$\eta = \frac{12\alpha E_F}{5aa_1 \tilde{x}^4}, \quad \chi = \frac{2\alpha}{\pi \lambda \tilde{\omega}^2 a_1^{\nu_1}}, \quad 1 \ll \tilde{\omega} \ll \omega \quad (\omega < \tilde{x}),$$
$$1 \ll \tilde{\omega} \ll \tilde{x} \ll \omega \quad (\omega > \tilde{x}).$$

The solutions (22) at  $3 < x < \omega$  are identical with those obtained previously in Ref. 7, since the case  $\omega < \tilde{x}$  represents account of only the electron-phonon interaction in the solution of the kinetic equation.

The solutions (21) are essentially different from (22), since in the case  $\omega > \tilde{x}$  there is an energy interval  $\tilde{x} \le x \le \omega$ , where only *e-e* interactions are operative. Here we must solve the following equation [see (17)]:

$$S_{ee}(n_0(x)) = \frac{2\alpha E_F}{aa_1} \theta(\omega - x).$$
<sup>(23)</sup>

The boundary condition for (23) has the form

$$n_0(x \ge \omega) = 0. \tag{24}$$

The boundary condition (24) is connected with the fact that the rapid exponential decay of  $n_0(x)$  behind the source makes a small contribution to the equation in comparison with the slowly falling power-law solutions at  $x < \omega$ ; therefore, we can neglect  $n_0(x)$  behind the source with high accuracy.

We shall show that behind the source  $(x > \omega) n_0(x)$  decays exponentially. In this energy region, the *e-e* interactions are responsible for the formation of the QDF since the probability of absorption of a phonon by a high-energy quasiparticle followed by "overshooting" it behind the source is small  $(\sim n_0^{-2}(x), x > 1)$ . The principal term in the distribution function is due the interactions between quasiparticles with energy  $x \sim 1$  and quasiparticles with energy  $x \sim \omega$ . The probability of the remaining *e-e* interactions is proportional to  $n_0^2(x)$ , where x > 1.

We keep in (10) those terms that describe the most probable energy relaxation processes in correspondence with what was said above. We set  $n_0(x) = n_T(x)$  at  $x \sim 1$  and integrate  $S^{\pm}$  with account of  $n_0(x) \leq 1$ , x > 1, where we obtain

$$n_0(x) = n_0(\omega) e^{\omega - x}, \quad x \ge \omega.$$
(25)

The function  $n_0(x)$  has been found numerically as well, by solution of Eq. (10) for the case of a broad source.

Figure 2 shows the QDF for various values of the parameter C. It is seen that the QDF falls off more slowly than  $n_T$  at x > 3. The numerical solution agrees with good accuracy with the theoretical dependence (21) over the entire range of variation of x [at  $x > \omega$ , the numerical solution also falls off exponentially, just as in (21)].

### **3. CALCULATION OF THE COHERENT CORRECTION**

The solution (21) and the numerically calculated  $n_0(x)$ , were substituted in  $\varphi_{ee}$  (5). The results are shown in the table. It is seen that the numerical values of  $\varphi_{ee}$  are close to the values calculated with the help of (21). We note that  $\varphi_{ee}$ depends weakly on C at  $C \sim C_{cr}$ . The point is that the form of  $n_0(x)$  undergoes weak changes when C increases in the interval of x that are significant for  $\varphi_{ee}(0 < x < \tilde{\omega})$  at  $C \sim C_{cr}$  (it can be shown that the contribution of the QDF to  $C\varphi_{ee}$  at



FIG. 2. Dependence of the quasiparticle distribution function on the energy at  $\Delta = 0$ ;  $x = \xi / a_1^{1/2}$ . 1—C = 0; 2—C = 7.7, 9.1, 11.9; 3—equilibrium QDF— $n_T$ .

 $x > \bar{\omega}$  is of the order of  $\tilde{\omega}^{-2} \lt 1$ ). Therefore we obtain approximately from (4)

$$\varphi = 1 - C \varphi_{ee} \approx 1 - 0.15C.$$
 (26)

We then obtain the result that the function  $\varphi$ , and consequently,  $n_1$ , changes sign at  $C = C_{\rm cr} \approx 6.5$ . It therefore follows from (2) and (3) that  $d\Delta / d\beta < 0$  (i.e.,  $\Delta (\beta)$  becomes a single-valued function) and  $N_S < 0$  (i.e., the appearance of a diffusion instability and of paramagnetism is possible).

## 4. MULTIPLICATION COEFFICIENTS OF THE EXCESS QUASIPARTICLES

For the quasiparticles created by the source transfer their energy to the electron gas and to the lattice. In the first case, the energy of the quasiparticles is expected in the increase in energy of the existing quasiparticles and in the creation of new ones, thereby increasing the total number of quasiparticles. This process is characterized by a quasiparticle multiplication coefficient  $r_{ee}$ . In the second case, the energy of the quasiparticles is expended in the creation of nonequilibrium phonons, which manage to reabsorbed during their life in the superconducting film, and which also create new quasiparticles with a multiplication coefficient  $r_{ef}$ . The total multiplication coefficient<sup>2</sup> is  $r = r_{ee} + r_{ef}$ .

The critical power  $\beta_c$  depends on the multiplication coefficients of the quasiparticles,  $r_{ee}$  and  $r_{ef}$ :

$$\beta_{c}(1+r_{ee}+r_{ef}) = \int_{0}^{\tilde{\omega}} dx_{1} n_{1} \int_{0}^{\tilde{\omega}-x_{1}} dx_{2} (x_{1}+x_{2})^{2} n_{2}, \quad (27)$$

where

TABLE I. Values of the expression  $\varphi_{ee}$  calculated numerically with the help of the function (21).

	С				
	0	1,8	7,7	9,1	11,9
$\varphi_{ee}$ , calculated according to	0.10	0.11	0.15	0.15	0.16
(21) $\varphi_{ee}$ , calculated numerically.	0.11	0.11	0,15	0.16	0.16

$$r_{ef} = \frac{1}{\beta_c} \int_{0}^{\tilde{\omega}} dx_1 \int_{0}^{\tilde{\omega} - x_1} dx_2 (x_1 + x_2)^2 N (x_1 + x_2) (1 - n_1 - n_2),$$
(28)

$$r_{ee} = \frac{1}{\beta_e} \int_{0}^{\infty} dx [S_{ee}^+(1-n_0(x)) - S_{ee}^-n_0(x)]$$
(29)

[compare Eqs. (27)–(29) with Eq. (16)]. Here N(x) is the distribution function for the nonequilibrium phonons, which satisfies the corresponding kinetic equation (Refs. 2 and 8). The function N(x) for the case  $\Delta = 0$  and T = 0 (T is the temperature) has the following form<sup>13</sup>:

$$N(x) = \frac{\gamma/\bar{\Delta}_{0}}{1 + \gamma x/\bar{\Delta}_{0}} \left[ 2\int_{x}^{\infty} n_{0}(x_{1}) \left(1 - n_{0}(x_{1} - x)\right) dx_{1} + \int_{0}^{\infty} n_{0}(x_{1}) n_{0}(x - x_{1}) dx_{1} \right],$$
(30)

where  $\tilde{\Delta}_0 = \Delta_0 / a_1^{1/2} \approx 1.7$  and for  $\gamma$  see below.

Substituting Eq. (21) and (30), we calculate (28) and (29), retaining the principal terms (which depend on  $\omega$ ). We then have for a broad source:

$$r_{ef} = \begin{cases} \gamma \frac{\omega}{\overline{\Delta}_{0}}; & \omega < \tilde{x}, \quad \gamma \ll 1 \\ \frac{6}{5} \gamma \frac{\omega \tilde{\omega}}{\overline{\Delta}_{0} \tilde{x}}; & \omega > \tilde{x}, \quad \gamma \ll 1 \\ \frac{4}{3} \frac{\omega}{\tilde{\omega}}; & \omega < \tilde{x}, \quad \gamma \gg 1 \\ \frac{8}{5} \frac{\omega}{\tilde{x}}; & \omega > \tilde{x}, \quad \gamma \gg 1 \end{cases}$$
(31)

Here  $\gamma$  is a parameter that characterizes the relative probability of reabsorption and departure of phonons from the film:

$$\gamma = \tau_{es} / \tau_B, \tag{32}$$

where  $\tau_{es} = 4d/s$ , d is the thickness of the film, s is the speed of sound,

 $\tau_B^{-1} = \pi \lambda \Delta_0 \omega_D / 2E_F.$ 

The quasiparticle distribution function itself depends weakly on  $\gamma$ ,<sup>14</sup> therefore the solutions (21) obtained at  $\gamma = 0$ are valid even at  $\gamma \neq 0$ . The case  $\gamma \ll 1$  corresponds to a thin film. It follows from (31) that in this situation  $r_{ef}$  depends linearly on  $\gamma$ . In the case of a thick film  $(\gamma \gg 1) r_{ef}$  does not depend on  $\gamma$ :

$$r_{ee} = \begin{cases} \frac{2\omega^{3}}{9\tilde{\omega}\tilde{x}^{2}}, & \omega < \tilde{x} \\ \frac{8}{5}\frac{\omega}{\tilde{x}} & \omega > \tilde{x} \end{cases}$$
(33)

In the case  $\omega < \tilde{x}$  (33) coincides with the results of Ref. 7 (since this is the case when the electron-phonon interaction is taken into account in the solution of the kinetic equation). In the case  $\omega > \tilde{x}$  the coefficient  $r_{ee}$  depends linearly on  $\omega$ , in contrast with Ref. 7.

Equation (31) and (33) are valid also in the case  $\Delta \neq 0$ , since the principal role in the multiplication coefficients is played by the quantity  $n_0(x)$  at higher energies, where  $x \ge \Delta / a_1^{1/2}$ .

Recently, Mitsen<sup>14</sup> measured the multiplication coefficients of the quasiparticles of lead films. The results agree with good accuracy with the calculation carried out using Eqs. (31) and (33).

- <sup>1)</sup> The *e-e* collisions were taken into account in Refs. 7 and 8 for the case of weak pumping, when  $(\Delta_0 \Delta)/\Delta_0 \leq 1$ .
- <sup>2)</sup> There is a misprint in Ref. 2—in Eq. (23)  $r_e r_f$  should read  $r_e + r_f$ .
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