

# Nonequilibrium state of superconductors with optical excitation of quasiparticles

V. F. Elesin

Moscow Engineering-Physics Institute

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The energy distribution of the nonequilibrium quasiparticles in a superconductor is investigated with the aid of the kinetic equation. The quasiparticles are produced in the absorption of an electromagnetic field whose frequency is more than double the superconducting gap. The values of the superconducting gap are calculated in the nonequilibrium state. The gap decreases as the amplitude of the field increases, and in certain cases it may vanish at the critical field. It is shown that the nature of the quasiparticle distribution function is essentially determined by the specific form of the dependence of the electron-phonon interaction matrix element on the wave vector. For the well-known dependences  $M_q^2 \sim q$  (atomic crystal),  $M_q^2 \sim \text{const}$  (deformation potential), and also  $M_q^2 \sim 1/q$  the distribution function is less than one-half ( $n_\epsilon < 1/2$ ). This implies that population inversion is not present at any amplitudes of the field.

## 1. INTRODUCTION

Recently there has been a great deal of interest in the investigation of nonequilibrium processes in superconductors.<sup>[1,2]</sup> The departure from equilibrium may be due to the influence of an electromagnetic field. In a certain sense the situation is analogous to the situation in semiconductors when they are exposed to light with a frequency greater than the width of the forbidden band. In fact, the field produces quasiparticles in the superconductor (analogous to electrons and holes in a semiconductor), and these possess excess energy and recombine with the emission of phonons. In addition, the field may warm up the quasiparticles.<sup>[2]</sup>

The investigation of nonequilibrium processes in superconductors is of interest, on the one hand, as a convenient method for studying the properties of superconductors and, on the other hand, in view of the great sensitivity of the effects to relatively weak fields superconductors are very appropriate objects to use in order to investigate nonequilibrium states. This is explained by the smallness of the superconducting gap in comparison with, for example, the Debye frequency. In fact, even for a small number of quasiparticles created by the field, their energy distribution turns out to have an exceptionally strong effect on the superconducting gap. Thus, it was shown in the articles by Eliashberg<sup>[2]</sup> that a change in the energy distribution of the quasiparticles due to heating by the field can lead to an increase of the gap. Conversely, the production of nonequilibrium quasiparticles should decrease the superconducting gap. This problem was considered in the article by Owen and Scalapino.<sup>[3]</sup> The authors of<sup>[3]</sup> assumed that the distribution function  $n_\epsilon$  of the nonequilibrium quasiparticles is described by a Fermi function with a nonvanishing chemical potential. When the temperature is equal to zero, such a function becomes equal to unity over a certain range of energies. This means that population inversion occurs for the quasiparticles ( $n_\epsilon > 1/2$ ). Population inversion would lead to a number of unusual properties.

The present article is devoted to an investigation of the energy distribution of the quasiparticles created in superconductors by an electromagnetic field, and also to the determination of the magnitude of the gap in the nonequilibrium state. In this connection major attention is paid to the case when the number of nonequilibrium quasiparticles greatly exceeds the number of equilibrium

particles, and the distribution function  $n_\epsilon$  is not small in comparison with unity. Under these conditions  $n_\epsilon$  differs substantially from the quasiequilibrium distribution function  $n_\epsilon^0$  (i.e., the Fermi distribution function with a nonvanishing chemical potential) and, as is shown in this article, cannot exceed 1/2. The latter assertion is proved under the assumption that the superconducting gap  $\Delta$  is much smaller than the Debye frequency  $\omega_D$ , and well known expressions ( $M_q^2 \sim q^k$ ,  $k = 1, 0, -1$ ) are used for the matrix elements of the electron-phonon interaction.

The physical reason for the absence of population inversion under these conditions is as follows. The quasiparticles produced by the light lose their excess energy and recombine at roughly the same rate, since scattering and recombination proceed by means of single-phonon processes. Therefore, the rate of loss of quasiparticles due to recombination and their rate of increase due to scattering are approximately the same (the recombination rate is faster for the matrix elements used here) and, consequently,  $n_\epsilon$  does not exceed 1/2.

We wish to emphasize that we are talking about states for which the distribution function of the nonequilibrium quasiparticles is not small in comparison with unity. In the opposite case, when  $n_\epsilon \ll n_\epsilon^0 \ll 1$ , the recombination time  $\tau_R \sim e^{\Delta/T}$  is large in comparison with the energy relaxation time and one can use the quasiequilibrium distribution function to describe  $n_\epsilon$  (but it is necessary to remember that  $n_\epsilon \ll 1$ ). The transference of this estimate to the case when  $n_\epsilon$  is not small, which is made by Owen and Scalapino in<sup>[3]</sup>, is unjustified, as is the conjecture about the quasiequilibrium form of the distribution function and the consequences which follow from here: About population inversion, concerning the specific form of the dependence of the gap on the number of quasiparticles, and so forth.

We have investigated the kinetic equation for quasiparticles<sup>[2]</sup> interacting with phonons, found analytic solutions for certain cases, and determined the magnitude of the gap in the nonequilibrium state.

## 2. GENERAL PROPERTIES OF THE DISTRIBUTION FUNCTION FOR NONEQUILIBRIUM QUASIPARTICLES

We shall confine our attention to an investigation of the superconducting states at  $T = 0$ . The kinetic equations and the equation for the gap have the following form<sup>[2,4]</sup> in the nonequilibrium state:

$$-(1-n_\epsilon)S^+(\epsilon) + n_\epsilon S^-(\epsilon) + n_\epsilon S^R(\epsilon) = Q(\epsilon); \quad (1)$$

$$\begin{pmatrix} S^+ \\ S^- \\ S^R \end{pmatrix} = \int_{\Delta}^{\infty} d\epsilon' \int_0^{\omega_D} \frac{d\omega_s \omega_q M^2(\omega_s) \epsilon \epsilon'}{(\epsilon^2 - \Delta^2)^{1/2} (\epsilon'^2 - \Delta^2)^{1/2}} \times \begin{pmatrix} n' \delta(\epsilon' - \epsilon - \omega_q) \left(1 - \frac{\Delta^2}{\epsilon \epsilon'}\right) \\ (1-n') \delta(\epsilon - \epsilon' - \omega_q) \left(1 - \frac{\Delta^2}{\epsilon \epsilon'}\right) \\ n' \delta(\epsilon + \epsilon' - \omega_q) \left(1 + \frac{\Delta^2}{\epsilon \epsilon'}\right) \end{pmatrix};$$

$$\epsilon = (\xi^2 + \Delta^2)^{1/2}, \quad \xi = p^2/2m - p_0^2/2m;$$

$$1 = \lambda \int_{\Delta}^{\omega_D} \frac{1-2n_\epsilon}{(\epsilon^2 - \Delta^2)^{1/2}} d\epsilon, \quad \lambda = g^2 \frac{m p_0}{2\pi^2}, \quad \hbar = 1, \quad (2)$$

where M is the matrix element of the electron-phonon interaction:

$$M^2 = \frac{\pi \lambda}{2s p_0} \left(\frac{\omega_q}{s p_0}\right)^k, \quad (3)$$

where  $k = 1$  corresponds to an atomic crystal,  $k = 0$  corresponds to the deformation potential method,  $\omega_D$  is the Debye frequency,  $p_0$  is the momentum at the Fermi surface,  $\Delta_0$  is the gap in the absence of the field, and  $s$  is the speed of sound.

The right-hand side of Eq. (1),  $Q(\epsilon)$ , describes the interaction of the quasiparticles with the electromagnetic field:<sup>[2]</sup>

$$Q(\epsilon) = 2\alpha [U_-(n_\epsilon - n_{\epsilon-\omega}) - U_+(n_\epsilon - n_{\epsilon+\omega}) + V(1 - n_\epsilon - n_{\epsilon-\omega})];$$

$$U_{\mp} = \frac{\epsilon(\epsilon \mp \omega) - \Delta^2}{(\epsilon^2 - \Delta^2)^{1/2} [(\epsilon \mp \omega)^2 - \Delta^2]^{1/2}} \theta(\epsilon \mp \omega - \Delta); \quad (4)$$

$$V = \frac{\epsilon(\omega - \epsilon) - \Delta^2}{(\epsilon^2 - \Delta^2)^{1/2} [(\omega - \epsilon)^2 - \Delta^2]^{1/2}} \theta(\omega - \epsilon - \Delta);$$

$\alpha = E^2 e^2 p_0 l / \omega^2 m$ , where  $E$  is the field amplitude,  $l$  is the electron mean free path, and  $\omega$  is the frequency of the field.

The quantity  $Q(\epsilon)$  only depends on  $\epsilon$ , i.e., it is an even function of  $\xi$ . Therefore, the distribution function  $n$  will only depend on the energy and, as a consequence of this, the terms which are odd in  $\xi$  are omitted from the collision integral in Eq. (1).

It is necessary to add the condition

$$\int_{\Delta}^{\infty} d\epsilon n_\epsilon S^R(\epsilon) = \int_{\Delta}^{\infty} d\epsilon Q(\epsilon) = 2\alpha \int_{\Delta}^{\infty} d\epsilon V(\epsilon) (1 - 2n_\epsilon). \quad (5)$$

to Eq. (1); this equation is obtained if (1) is integrated with respect to energy. The first two terms in Eq. (1) describe the scattering of the quasiparticles;  $S^+$  refers to arrival,  $S^-$  refers to departure, and the third term  $S^R$  refers to recombination. The recombination term is quadratic in  $n_\epsilon$  and small for  $n_\epsilon \ll 1$ . However, for  $n_\epsilon \sim 1$  it is comparable with the first two terms.

In order to investigate the general properties of  $n_\epsilon$ , it is convenient to write Eq. (1) in the following form:

$$n_\epsilon = \frac{S^+(\epsilon) + Q(\epsilon)}{S^+(\epsilon) + S^-(\epsilon) + S^R(\epsilon)}. \quad (6)$$

Let us examine  $n_\epsilon$  for  $\epsilon = \Delta$ , when  $n_\epsilon$  assumes its maximum value. It is clear from Eq. (6) that  $n_\Delta$  is determined by the ratio of  $S^+(\Delta)$  to  $S^R(\Delta)$  ( $S^-(\Delta) = 0$ ). The quasiparticles produced by the light gather together above (below) the gap and occupy a certain energy range  $[\Delta, \tilde{\epsilon}]$ . If  $\tilde{\epsilon} \ll \omega_D$  (which is usually realized), then the limits of integration are identical in  $S^+$  and  $S^R$ , and for the matrix element (3) with  $k = 0, 1$  the quantity  $S^R(\Delta)$  turns out to be larger than  $S^+(\Delta)$ . The coherence factors also increase  $S^R$ . Thus,  $n_\Delta < 1/2$ .

To verify this directly, let us calculate  $S^+(\Delta)$  and  $S^R(\Delta)$  for the function  $n_\epsilon$ , which has the form of the Fermi step-function (at  $T = 0$ ):

$$n_\epsilon = \begin{cases} 1, & \epsilon < \mu \\ 0, & \epsilon > \mu \end{cases},$$

where  $\mu$  is related to the dimensionless number of quasiparticles  $\bar{n}$  by the equation

$$\mu = (\Delta^2 + \Delta_0^2 \bar{n}^2)^{1/2}, \quad \bar{n} = \frac{1}{\Delta_0} \int_{\Delta}^{\infty} \frac{\epsilon n_\epsilon d\epsilon}{(\epsilon^2 - \Delta^2)^{1/2}}. \quad (7)$$

Precisely such a function was used in the article by Owen and Scalapino.<sup>[3]</sup> We shall assume for simplicity that the intensity of the source is not strong, that is,  $\bar{n} \ll 1$ , so that

$$\Delta \approx \Delta_0 (1 - 2\bar{n}), \quad \mu - \Delta \approx 2\Delta_0 \bar{n}^2. \quad (8)$$

In this case the integrals are easily evaluated:

$$S^+(\Delta) \approx \frac{2(\mu - \Delta)^{1/2}}{7\sqrt{\Delta}}, \quad S^R(\Delta) \approx 8\Delta^3 \left[ \frac{2(\mu - \Delta)}{\Delta} \right]^{1/2}$$

and for the ratio  $S^+/S^R$  we obtain

$$\frac{S^+(\Delta)}{S^R(\Delta)} \approx \frac{1}{28\sqrt{2}} \left( \frac{\mu - \Delta}{\Delta} \right)^3 = \frac{\bar{n}^6}{224\sqrt{2}} \ll 1, \quad (9)$$

i.e., the recombination is considerably stronger than the arrival term. This means that the distribution function is not described by the Fermi function and  $n_\epsilon$  is much smaller than 1.

Therefore, it is necessary to solve the kinetic equation (1) in order to determine  $n_\epsilon$ . Below we examine the problem separately for the following two cases: 1) A "wide source" with frequency  $\omega \gg \Delta_0$ , which produces quasiparticles in a broad range of energies,  $\Delta < \epsilon < \omega - \Delta \gg \Delta_0$ ; 2) a "narrow source" with frequency  $\omega - 2\Delta_0 \ll \Delta_0$ .

### 3. THE CASE OF A WIDE SOURCE

In this case  $Q$  can be omitted from Eq. (1) (that is, in the interval  $\Delta < \epsilon < \tilde{\epsilon} \ll \omega$ ), and the connection between  $n_\epsilon$  and  $Q$  can be found from condition (5). Taking  $Q$  into account in Eq. (1) leads to the appearance of small corrections (see below). We shall find the solution of Eq. (1) with the matrix element (3) given by the expression

$$M^2 = \frac{\pi \lambda}{2p_0 s} \left(\frac{p_0 s}{\omega_q}\right). \quad (10)$$

After integrating with respect to  $\omega_q$  in Eq. (1) we obtain

$$-(1-n_\epsilon) \int_{\Delta}^{\omega_D} n_\epsilon' \left(1 - \frac{\Delta^2}{\epsilon \epsilon'}\right) \frac{\epsilon' d\epsilon'}{(\epsilon'^2 - \Delta^2)^{1/2}} + n_\epsilon \int_{\Delta, \epsilon - \omega_D}^{\epsilon} (1-n_\epsilon') \times \left(1 - \frac{\Delta^2}{\epsilon \epsilon'}\right) \frac{\epsilon' d\epsilon'}{(\epsilon'^2 - \Delta^2)^{1/2}} + n_\epsilon \int_{\Delta}^{\omega_D - \epsilon} n_\epsilon' \left(1 + \frac{\Delta^2}{\epsilon \epsilon'}\right) \frac{\epsilon' d\epsilon'}{(\epsilon'^2 - \Delta^2)^{1/2}} = 0. \quad (11)$$

We are interested in the distribution of the quasiparticles for source intensities such that  $n_\epsilon$  is of the order of unity in the interval  $[\Delta, \tilde{\epsilon}]$ . Under these conditions  $\tilde{\epsilon} \sim \Delta_0 \bar{n} \sim \Delta_0$ , but  $\Delta$  decreases and becomes small in comparison with  $\Delta_0$ , so that the inequality

$$\Delta \ll \tilde{\epsilon} \ll \omega_D. \quad (12)$$

is satisfied.

Equation (11) can be simplified by using relation (12)<sup>[1]</sup>

$$-(1-n) \int_{\Delta}^{\omega_D} n' d\epsilon' + n \int_{\Delta}^{\epsilon} (1-n') d\epsilon' + n \int_{\Delta}^{\omega_D} n' d\epsilon' = 0. \quad (13)$$

We introduce the new function:

$$\chi(\varepsilon) = \int_{\Delta}^{\varepsilon} (1-n') d\varepsilon', \quad \chi(\Delta) = 0, \quad (14)$$

which obeys the equation

$$\frac{d\chi}{d\varepsilon} (2\chi + 2\bar{n}\Delta_0 - \varepsilon + \Delta) = \chi + \bar{n}\Delta_0. \quad (15)$$

The solutions for  $\chi$  and consequently for  $n_{\varepsilon}$  have the form

$$\chi(\varepsilon) = 1/2(\varepsilon - \Delta) + [(\varepsilon - \Delta)^2/4 + |c|]^{1/2} - \bar{n}\Delta_0, \quad (16)$$

$$|c| = \bar{n}^2\Delta_0^2,$$

$$n_{\varepsilon} = 1 - \frac{d\chi}{d\varepsilon} = \frac{1}{2} \left( 1 - \frac{\varepsilon - \Delta}{[(\varepsilon - \Delta)^2 + 4\Delta_0^2\bar{n}^2]^{1/2}} \right). \quad (17)$$

The dimensionless concentration of quasiparticles is found from condition (5):

$$\bar{n}^2 \approx \beta \left( 1 - \frac{2\Delta}{\omega} - 2\bar{n} \frac{\Delta_0}{\omega} \right) \approx \beta, \quad \beta = \frac{4\alpha\omega}{\pi\lambda\Delta_0^2}. \quad (18)$$

The maximum value of the function  $n_{\varepsilon}$  (given by Eq. (17)) is attained at  $\varepsilon = \Delta$  and is equal to  $1/2$ .  $n_{\varepsilon}$  decreases with increasing values of  $\varepsilon$ , and it becomes small for  $\varepsilon > \Delta_0\bar{n}$ , as was assumed above. The physical reason for the absence of population inversion lies in the equality (for the present model) of the recombination and scattering rates.

Now let us generalize the obtained results. With the aid of Eq. (6) we can trace the dependence of the function  $n_{\varepsilon}$  on the specific form of the matrix element  $M_{\mathbf{q}}^2$ . It is clear from Eq. (6) that for  $k = 0, 1$  the term  $S^+$ , corresponding to arrival, is diminishing in comparison with the recombination term  $S^R$ . Therefore,  $S^R > S^+$  and the distribution function  $n_{\varepsilon}$  becomes smaller than  $1/2$  at  $\varepsilon = \Delta$ .<sup>2)</sup> If  $Q$  is explicitly taken into account in Eq. (13), i.e., if the source

$$Q \approx \beta_1(1 - 2n_{\varepsilon})\theta(\omega - \varepsilon - \Delta), \quad \beta_1 = 4\alpha/\pi\lambda,$$

is added to the right-hand side, then we obtain

$$n_{\varepsilon} \approx \frac{1}{2} \left( 1 - \frac{\varepsilon - \Delta}{[(\varepsilon - \Delta)^2 + 4(\Delta_0\bar{n} + \beta_1)^2]^{1/2}} \right). \quad (19)$$

instead of expression (17). With allowance for the equality (18) we find

$$\beta_1/\Delta_0\bar{n} \approx \Delta_0\bar{n}/\omega \ll 1,$$

i.e.,  $\beta_1$  is small in comparison with  $\Delta_0\bar{n}$  and can be neglected in the approximation we have adopted.

#### 4. THE SUPERCONDUCTING GAP IN THE NONEQUILIBRIUM STATE

We substitute the function (17) into (2) and obtain the following equation for the gap at  $T = 0$ :

$$1 = \lambda \int_{\Delta}^{\omega} \frac{d\varepsilon}{(\varepsilon^2 - \Delta^2)^{1/2}} \frac{\varepsilon - \Delta}{[(\varepsilon - \Delta)^2 + 4\Delta_0^2\bar{n}^2]^{1/2}}. \quad (20)$$

This equation enables us to determine the dependence of the gap on  $\bar{n}$ , and, consequently, the dependence on the field amplitude. We can find the critical value  $\bar{n}_c$  for which  $\Delta = 0$  from the equation

$$1 = \lambda \int_0^{\omega} \frac{d\varepsilon}{(\varepsilon^2 + 4\bar{n}_c^2\Delta_0^2)^{1/2}}.$$

It turns out to be given by

$$\bar{n}_c = 1/2. \quad (21)$$

In the case  $\bar{n} \lesssim \bar{n}_c$  (i.e.,  $\Delta \ll \Delta_0$ ) the dependence of  $\Delta$  on  $\bar{n}$  is of the form

$$\Delta \approx \Delta_0(1 - 2\bar{n}) \left( 1 + \ln \frac{4\Delta_0}{\Delta} \right)^{-1}. \quad (22)$$

In contrast to the prediction of Owen and Scalapino,<sup>[3]</sup> the gap monotonically tends to zero. This is due to the fact that the difference  $1 - 2n_{\varepsilon}$  does not change sign (also see<sup>[5]</sup>).

Now let us estimate the intensity  $E_c^2$  at which the gap vanishes. Taking (18) into account we find

$$E_c^2 \approx \frac{\omega\Delta_0^2}{16\pi\lambda\nu_0 l e^2}. \quad (23)$$

If we assume  $\Delta_0 \approx 10^\circ \text{K}$ ,  $\lambda = 0.1$ ,  $\omega = 10^{12} \text{sec}^{-1}$ ,  $\nu_0 = 10^8 \text{cm/sec}$ , and  $l = 10^{-5} \text{cm}$ , we then obtain  $E_c \approx (1 \text{ to } 10) \text{V/cm}$ .

#### 5. THE CASE OF A NARROW SOURCE

If the frequency of the source satisfies the inequality

$$\omega - 2\Delta_0 \ll \Delta_0, \quad (24)$$

then at  $T = 0$  the quasiparticles are localized in a narrow range of energies,  $\tilde{\varepsilon} - \Delta_0 \approx \omega - 2\Delta_0 \ll \Delta_0$ . In this case the recombination rate for  $M_{\mathbf{q}}^2 \sim q$  is much faster than the scattering rate (see Sec. 2), so the terms  $S^+$  and  $S^-$  in Eq. (1) can be neglected.

Then the equation for  $n_{\varepsilon}$  can be written in the form

$$\frac{\varepsilon n_{\varepsilon}}{(\varepsilon^2 - \Delta^2)^{1/2}} \int_{\Delta}^{\omega} \frac{\varepsilon' n_{\varepsilon'} d\varepsilon'}{(\varepsilon'^2 - \Delta^2)^{1/2}} = \frac{\alpha}{4\pi\lambda} \left( \frac{sp_0}{\Delta} \right)^2 V(\varepsilon). \quad (25)$$

We have set  $n_{\varepsilon} = 0$  in  $Q$ . This approximation is valid for a weak field. From Eq. (25) we obtain the following expression for the distribution function:

$$n_{\varepsilon} = \frac{1}{\bar{n}} \frac{\beta}{16\omega} \left( \frac{sp_0}{\Delta} \right)^2 (\varepsilon^2 - \Delta^2)^{1/2} V(\varepsilon), \quad (26)$$

where  $\bar{n}$  is determined from Eq. (5):

$$\bar{n}^2 = \frac{\pi\beta}{16} \left( \frac{sp_0}{\Delta} \right)^2 \frac{\omega - 2\Delta}{2\omega}. \quad (27)$$

With the aid of Eqs. (27) and (2) we find that the gap is given by

$$\Delta \approx \Delta_0(1 - 2\bar{n}), \quad \bar{n} \sim \beta^{1/2} \ll 1. \quad (28)$$

One interesting property should be noted concerning the nonequilibrium state of a superconductor upon excitation by light of frequency  $\omega \gtrsim 2\Delta_0$ . As the light intensity increases, the number of quasiparticles increases and the gap decreases which, in turn, leads to an increase in the number of quasiparticles produced (since the effective range of the source increases). This may cause instability of the state. In fact, let the number  $\bar{n}$  increase, then  $\Delta$  decreases which leads, in turn, to a further increase of  $\bar{n}$ , and so forth.

An experimental study of the photoexcitation of quasiparticles in nonequilibrium superconductors is described in the article by Parker and Williams.<sup>[6]</sup> The dependence of the gap on the intensity which was observed at low temperatures is in agreement with expression (28). However, the most interesting region,  $\bar{n} \sim 1$ , still remains uninvestigated.

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<sup>1)</sup>An analogous equation was studied by V. M. Galitskii and the author in connection with an investigation of semiconductors in a strong electromagnetic field.

<sup>2)</sup>It should be noted that if  $k < -1$  the function  $n_{\varepsilon}$  may generally become larger than  $1/2$ .

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