On the role of electron–electron collisions in the formation of nonequilibrium distributions in superconductors

A. G. Aronov and B. Z. Spivak

B. K. Konstantinov Leningrad Institute of Nuclear Physics, Academy of Sciences of the USSR
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The effect of the electron–electron collisions in superconductors on the nonequilibrium distributions that arise upon the injection of quasiparticles is investigated. It is shown that a quasiequilibrium distribution of quasiparticles with a negative chemical potential arises in the superconductor at high injection rates. The results of the Willenson-Gray experiment [Phys. Rev. Lett. **41**, 812 (1978)] are discussed.

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In a recent investigation Willenson and Gray[1] observed on injecting nonequilibrium quasiparticles into a superconducting Al film a quasiparticle distribution of the form

$$n_\varepsilon = \exp\left(\frac{\nu-\varepsilon}{kT}\right) \tag{1}$$

with temperature T different from the thermostat temperature T_0 and a chemical potential, ν, that became negative at high injection rates. In Ref. 2 the consequences of the existence of a negative chemical potential of quasiparticles are discussed, and it is shown that so long as $\nu<0$, the energy gap $\Delta>0$, even if the temperature of the excitation is higher than the critical temperature. This is precisely what has been experimentally observed.

In Refs. 3–5 the nonequilibrium quasiparticle distributions that arise during injection, when the electron-phonon collisions are the principal energy-relaxation mechanism, are theoretically investigated. However, none of the existing theories predicts the possibility of the existence of a state with $\nu<0$. At the same time, the observation has repeatedly been made (see, for example, Refs. 6–8) that the electron-electron collisions can play an important role in the energy relaxation of electrons in metals with a high Debye phonon energy, in particular, in Al.

In the present paper we show that if the electron-electron collisions are important, then their consideration allows us to account for the results of the Willenson–Gray experiment.[1]

The electron-electron collision operator in a superconductor[9] has the form

$$J_{ee}=\frac{1}{2\mu}\int_\Delta^\infty \frac{d\varepsilon_1\,d\varepsilon_2\,d\varepsilon_3}{\varepsilon(\varepsilon_1^2-\Delta^2)^{1/2}(\varepsilon_2^2-\Delta^2)^{1/2}(\varepsilon_3^2-\Delta^2)^{1/2}}\{[(1-n)n_1n_2n_3$$
$$-n(1-n_1)(1-n_2)(1-n_3)]M_1\delta(\varepsilon-\varepsilon_1-\varepsilon_2-\varepsilon_3)$$
$$+3M_3[n_1(1-n)(1-n_2)(1-n_3)-(1-n_1)nn_2n_3]\cdot\delta(\varepsilon+\varepsilon_2+\varepsilon_3-\varepsilon_1)$$
$$+3M_2[nn_1(1-n_2)(1-n_3)-(1-n)(1-n_1)n_2n_3]\delta(\varepsilon+\varepsilon_1-\varepsilon_2-\varepsilon_3)\}, \tag{2}$$

$$M_1=a_1(\varepsilon\varepsilon_1\varepsilon_2\varepsilon_3-\Delta^4)-{}^1/_3a_2\Delta^2(\varepsilon_1\varepsilon_2+\varepsilon_1\varepsilon_3+\varepsilon_2\varepsilon_3-\varepsilon^2),$$

$$M_2=-M_1(-\varepsilon_3),\quad M_3=M_1(-\varepsilon_2,-\varepsilon_3),$$

$$a_i=-\pi N(0)\int\frac{d\Omega_p\,d\Omega_{p_1}\,d\Omega_{p_2}}{(4\pi)^3}\delta\left(\frac{\mathbf{p}-\mathbf{p}_1-\mathbf{p}_2}{p_F}-1\right)A_i,$$

$$A_1=V_{p-p_1}V_{p-p_2}-2|V_{p-p_2}|^2,$$

$$A_2=V_{p-p_1}V_{p-p_2}+V_{p_1+p_2}V_{p-p_2}-2|V_{p_1+p_2}|^2+V_{p-p_1}V_{p_1+p_2}.$$

Here μ is the chemical potential of the normal metal, $N(0)=mp_F/\pi^2$ is the density of states at the Fermi surface, p_F is the Fermi momentum, $\Omega_p=\mathbf{p}/p_F$, and V_q is the Fourier transform of the electron-electron potential. Connected with the electron-electron collisions in superconductors are two processes.

1. The scattering of the quasiparticles on each other with conservation of the total number of quasiparticles and the total energy [the third term in (2)]. The characteristic time of these processes is given by the formula

$$\frac{1}{\tau_s^{ee}}\approx(3a_1-a_2)\frac{\Delta^2x}{\mu},\qquad x=\frac{\sum_{p\sigma}n_{p\sigma}}{4N(0)\Delta}=\left(\frac{\pi kT}{\Delta}\right)^{1/2}\exp\left(\frac{\nu-\Delta}{kT}\right), \tag{3}$$

where x is the dimensionless quasiparticle concentration.

2. Recombination (and impact ionization), when one quasiparticle with $\varepsilon\gtrsim 3\Delta$ is created instead of three quasiparticles with $\varepsilon\approx\Delta$. The characteristic time of this process is given by the relation

$$1/\tau_r^{ee}\approx a_2\Delta^2x^2/\mu. \tag{4}$$

The electron-electron collisions are important if $\tau_s^{ee}<\tau_r^{ph}$, where $\tau_r^{ph}=(4\lambda\Delta^3x/\Theta^2)^{-1}$ is the characteristic time of the quasiparticle recombination with emission of a phonon, λ is the electron-phonon coupling constant, and Θ is the Debye energy. This condition can be written in terms of the parameter η:

$$\eta=\frac{8}{\pi}\frac{\lambda}{3a_1-a_2}\frac{\Delta\mu}{\Theta^2}\approx\frac{\tau_s^{ee}}{\tau_r^{ph}}<1. \tag{5}$$

Experimentally, this parameter is not well known, largely because of the absence of data on the quantities a_1 and a_2. In Al, $\lambda\Delta\mu/\Theta^2\approx 0.2$, and therefore it may be inferred that the parameter $\eta<1$. We shall consider the case when $x\ll 1$. If $\eta\ll 1$, then the electron-electron scattering that conserves the total energy and the quasiparticle number turns out to be the fastest process. It establishes the distribution (1), which makes the scattering part of the operator J_{ee} vanish at arbitrary ν and T. The recombinational part of the J_{ee} operator is less than the scattering part according as the parameter $x\ll 1$.

As in Ref. 5, we find the law of conservation of the quasiparticle number to have the form

$$c\frac{I}{d\Delta}=\frac{2N(0)\Delta}{\tau_{ph}^r}\left\{x-x_0+\frac{x^2}{\eta}\frac{a_2}{3a_1-a_2}\frac{\pi}{6\sqrt{2}}(1-e^{-2\nu/kT})\right\}, \tag{6}$$

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where x_0 is the dimensionless equilibrium quasiparticle concentration at T_0 and I is the pump intensity. The number c (~1) in (6) takes account of the quasiparticle multiplication during the injection, as well as the fact that not the entire absorbed energy goes into the production of quasiparticles with $\varepsilon \approx \Delta$. If $x < \eta < 1$, then the electron recombination [the last term in (6)] can be neglected, and the quasiparticle concentration is determined by the phonon recombination, i.e., $x \sim I^{1/2}$ (see, for example, Ref. 3).

In order to find the energy conservation law, let us use a method similar to the one used in Ref. 5, and introduce the energy ε_1 ($\varepsilon_1 - \Delta > kT$) below which the electron-electron collisions become more effective than the electron-phonon collisions, i.e., below which

$$\tau_*^{ee}(\varepsilon_1) \approx \tau_*^{ph}(\varepsilon_1)$$

or

$$\varepsilon_1 - \Delta = \beta\Delta (x/\eta)^{2/7}; \tag{7}$$

where β is a number of the order of unity.

We shall call the energy region $\Delta \leqslant \varepsilon \leqslant \varepsilon_1$ the active region. The quasiparticles that are created during the injection fall into the active region through the emission of phonons. Here frequent electron-electron collisions establish a quasi-equilibrium distribution, conserving the total energy in the process. If $\tilde{d} = d/t \ll l_>$, where d is the film thickness, t is the coefficient of penetration of the phonons into the substrate, and $l_>$ is the mean free path of the phonons with energy higher than 2Δ, then the phonon distribution is an equilibrium distribution with temperature equal to T_0, and the energy leaves the sample in the form of low-frequency phonons. Each phonon on the average carries away an amount of kinetic energy of the order of kT. As a result, the energy conservation law has the form[5]

$$\beta x \frac{\varepsilon_1 - \Delta}{kT} = \left(\frac{kT}{\Delta}\right)^{7/2} J\left(\frac{T}{T_0}\right), \tag{8}$$

$$J(t) = \frac{t^{7/2}}{4} \int_0^\infty dy\, y^3 \frac{e^{-y} - e^{-ty}}{1 - e^{-ty}} \int_0^\infty dx e^{-x} \frac{2x+y}{[x(x+y)]^{1/2}}. \tag{8a}$$

When $x \ll \eta < 1$, the expressions (6) and (8) determine the dependence of ν and T on the pump intensity. [It follows from (6) that $\nu \to 0$ when $x > \eta$.]

At low pump intensities, so long as the quasiparticle concentration $x < (kT_0/\Delta)^{7/2}\eta^{2/9}$, the temperature $T = T_0$, as can be seen from (8), and the chemical potential increases monotonically with increasing pump intensity.

If, on the other hand, $x > (kT_0/\Delta)^{7/2}\eta^{2/9}$, then we have from (8) the relation between density of the excitations and the temperature:

$$x \approx (kT/\Delta)^{7/2}\eta^{2/9}. \tag{9}$$

We have omitted the numerical factor, since because of the small powers with which they enter into the expression (9), it is with a high degree of accuracy equal to unity. It follows from (9) that

$$\frac{\nu}{\Delta} = 1 + \frac{kT}{\Delta} \ln\left[\left(\frac{kT}{\Delta}\right)^3 \eta^{2/9}\right]. \tag{10}$$

Since $x \sim I^{1/2}$, it follows from the expression (10) that,

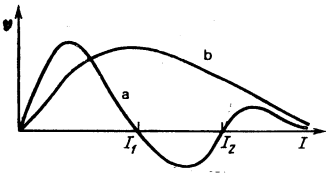

FIG. 1. Dependence of the chemical potential of the excitations on the pump intensity.

as the pump intensity increases, the temperature of the excitations increases, while ν decreases. Furthermore, for $\eta < 1$, there always exists a temperature region, $T_1 \leqslant T \leqslant T_2$, in which $\nu < 0$. The temperatures T_1 and T_2 are defined as the roots of the equation

$$(kT_i/\Delta)^{-3} \exp(-\Delta/kT_i) = \eta^{2/9}. \tag{11}$$

Furthermore, the solution to (11) vanishes only when $\eta > 3.7$, i.e., outside the limits of applicability of the theory. If even $\eta = 1$, $\Delta/kT_1 \approx 4.5$ and $\Delta/kT_2 \approx 1.8$. Correspondingly, the concentrations at these points are $x_1 \approx 10^{-1}$ and $x_2 \approx 5 \times 10^{-3}$, and are much smaller than unity.

Let us briefly discuss what happens when the film thickness is increased. If $l_> < \tilde{d} < l_>/x$, then the effective lifetime of the quasiparticles becomes $\tau^r_{ph}\tilde{d}/l_>$ (Ref. 5), instead of τ^r_{ph}. As a result, $\eta^{2/9}$ in the expressions (9) and (10) will be replaced by the quantity $\eta^{2/9}\tilde{d}/l_>$, and therefore the conditions under which distributions with negative chemical potentials can be observed become complicated, and, in general, the region in which $\nu < 0$ vanishes when $\eta^{2/9}\tilde{d}/l_> > 1.35$.

The dependence $\nu(I)$ is shown in Fig. 1 for two cases: a) $\eta^{2/9}\tilde{d}/l_> < 1.35$ and b) $\eta^{2/9}\tilde{d}/l_> > 1.35$.

It is interesting to attempt to estimate the quasiparticle concentration when the temperature of the nonequilibrium excitations becomes equal to T_c. From (9) we have $x(T_c) \approx 0.1\eta^{2/9}$, and the change $\delta\Delta/\Delta_0 \approx 2x \approx 0.2\eta^{2/9}$.

Thus, for thin films and sufficiently small values of η, the results of the theory are valid right down to the nonequilibrium-excitation temperatures close to the critical temperature.

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