Relaxation of the Superconducting Order Parameter*

G. LUCAS† AND M. J. STEPHEN
Physics Department, Yale University, New Haven, Connecticut
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The creation and annihilation of quasiparticles in a superconductor through their interaction with the phonon field is examined. At low frequencies this appears to be the main mechanism leading to relaxation of the order parameter in a superconductor. The relaxation time becomes long both close to T_c and at low temperatures. Close to T_c, a diffusion equation for the order parameter is obtained. The correlation in time of the fluctuations of the order parameter is examined. A weak coupling between the amplitude and phase of the order parameter is shown to lead to a slow diffusion of the phase of the order parameter.

1. INTRODUCTION

THE BCS[1] theory of superconductivity provides an excellent description of the equilibrium state of a superconductor. We consider here the mechanism by means of which equilibrium is established. At a finite temperature in a superconductor, a dynamic equilibrium exists between the quasiparticles and condensed pairs. Condensed pairs are continually breaking up, forming quasiparticles above the gap, and quasiparticles are recombining to form pairs. When a superconductor is disturbed from equilibrium, e.g., by application of a magnetic field or by heating or cooling, these processes bring the number of excitations and hence the energy gap Δ (order parameter) to their new equilibrium values. In this paper, we examine the creation and annihilation of quasiparticles through their interaction with the phonon field as a mechanism leading to relaxation in the superconductor. This mechanism has been considered by Schrieffer and Ginsberg[2] and by Parmenter.[3] The mean recombination time of excitations τ is obtained and a rate equation for the quasiparticles is derived. The situation is very similar to that in a mixture of fluids which can react chemically. Close to the critical temperature T_c, the rate equation reduces to a diffusion equation for the energy gap Δ. At temperatures low compared with T_c, the recombination time τ is proportional to $e^{\beta\Delta}$, when $\beta=(kT)^{-1}$. This is the case considered by Schrieffer and Ginsberg,[2] and it arises because the number of excited electrons available for pairing is proportional to $e^{-\beta\Delta}$.

There are other interactions which lead to recombination of excitations and hence relaxation of the superconductor. Burstein, Langenberg, and Taylor[4] have considered the recombination of two excitations accompanied by the emission of a photon. This leads to a recombination time $\tau=0.4$ sec and is a slow process compared with that due to the phonons. The residual Coulomb interaction between excitations will also lead to recombination, but if this Coulomb interaction is of the same order of magnitude as that between quasiparticles in a normal metal, this is not an important process. Abrahams and Tsuneto[5] have shown that in the presence of time-dependent fields if there are frequency components $\omega>2\Delta$ of the fields (or of the energy gap itself) then pairs will be broken or formed. If the fields vary sufficiently rapidly in space, the above condition on the frequency can be relaxed. Close to T_c they have shown that this also leads to a diffusion equation for the energy gap. However, this process is slower than that considered above.

The recombination time τ is of interest in the case of superconductors in some nonequilibrium situations. At the present time there is little direct experimental evidence relating to this process. However, τ does have a significant effect on the electronic transport properties of a superconductor, e.g., thermal conductivity.[6]

The recombination of quasiparticles (phonons and rotons) in superfluid He II has been considered by Khalatnikov.[7] His method of deriving the rate equation is followed in this paper. We also make a comparison between the processes taking place in a superconductor and those in He II.

2. THEORY

In a superconductor at a finite temperature, a dynamic equilibrium exists between quasiparticles and condensed pairs. In certain respects a superconductor can be regarded as a mixture of two fluids and the condition of equilibrium of the quasiparticles and condensed pairs can be derived in the same way as for two fluids that can react chemically. If n_n is the density of excitations and n_s is the superfluid density, then the equilibrium condition is that the corresponding chemical potentials μ_n and μ_s be equal. These potentials are

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† Present address: Physics Department, Yeshiva University, New York.

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determined in the usual manner from the energy (or any other thermodynamic potential) by

$$dE = TdS + \mu_n dn_n + \mu_s dn_s. \quad (1)$$

It is more convenient to regard E as depending on $n = n_s + n_n$ and n_s, so that

$$dE = TdS + \mu_n dn + (\mu_s - \mu_n) dn_s. \quad (2)$$

The equilibrium condition of the normal fluid and superfluid is then

$$\mu_s - \mu_n = (\partial E / \partial n_s)_{S,n} = 0. \quad (3)$$

It is interesting to note that this equation is precisely the Ginzburg-Landau equation of superconductivity in the absence of current flow if we identify n_s with $|\psi|^2$, where ψ is the order parameter.[8]

Now let us consider how the equilibrium between quasiparticles and condensate is established. It is interesting first to examine the case of He II which has been studied by Khalatnikov.[7] The quasiparticles in He II are phonons (p) and rotons (R) and Khalatnikov considered the following processes which change the number of excitations:

$$p_1 + p_2 + p_3 \rightleftharpoons p_4 + p_5, \quad (4a)$$

$$p + R_1 \rightleftharpoons R_2 + R_3. \quad (4b)$$

In Eq. (4a), three phonons collide producing two, or vice versa, and in (4b) an energetic phonon collides with a roton producing two rotons, or vice versa. There are also scattering processes in which the number of excitations of one kind does not change, and these serve to bring the excitations into equilibrium among each other. The processes (4) give rise to the second-viscosity coefficients in the two-fluid model of He II which in turn accounts for the damping of first and second sound.

In a superconductor, the phonons play an important role in bringing the quasiparticles into equilibrium with the condensed pairs. The electron-phonon interaction in the conventional form is

$$H_{ep} = \sum_{\mathbf{k},\mathbf{q},\sigma} (V_q b_{\mathbf{q}} C_{\mathbf{k}+\mathbf{q},\sigma}^{\dagger} C_{\mathbf{k},\sigma} + \text{c.c.}). \quad (5)$$

Part of this interaction has already been used in forming the superconducting state but, as pointed out by BRT, the part that leads to real transitions remains. When written in terms of quasiparticle creation and annihilation operators $\gamma_{k0,1}^{\dagger}$ and $\gamma_{k0,1}$, Eq. (5) becomes

$$H_{ep} = \sum_{\mathbf{k},\mathbf{q}} V_q b_q [(u_{k+q} u_k - v_{k+q} v_k) \times (\gamma_{\mathbf{k}+\mathbf{q}0}^{+} \gamma_{\mathbf{k}0} + \gamma_{\mathbf{k}1}^{+} \gamma_{\mathbf{k}+\mathbf{q}1}) + (u_{k+q} v_k + u_k v_{k+q}) \times (\gamma_{\mathbf{k}+\mathbf{q}0}^{+} \gamma_{\mathbf{k}1}^{+} + \gamma_{\mathbf{k}+\mathbf{q}1} \gamma_{\mathbf{k}0})] + \text{c.c.} \quad (6)$$

[8] We are grateful to Dr. L. Mittag for valuable discussion on this point.

The first two terms lead to scattering of the quasiparticles by phonons, but the third and fourth terms give rise to processes in which the number of quasiparticles is changed. These processes can be represented by the equations (where ν is a phonon)

$$E_{\mathbf{k}+\mathbf{q}} \rightleftharpoons E_{\mathbf{k}} + \nu_{\mathbf{q}}, \quad (7a)$$

$$E_{\mathbf{k}+\mathbf{q}} + E_{\mathbf{k}} \rightleftharpoons \text{pair} + \nu_{\mathbf{q}}. \quad (7b)$$

To determine the rate of change of the number of quasiparticles we can use the Boltzmann equation derived by BRT[6] in connection with the thermal conductivity. In the absence of spatial variations the Boltzmann equation is

$$\partial f_k / \partial t = I_a + I_b, \quad (8)$$

where f_k is the distribution function of the quasiparticles of momentum $\mathbf{k}$ and the collision terms I_a and I_b arise from the scattering processes (7a) and (7b), respectively. The rate of change of the number density n_n of quasiparticles is now

$$\frac{\partial n_n}{\partial t} = \frac{\partial}{\partial t} \frac{2}{V} \sum_k f_k = \frac{2}{V} \sum_k I_b. \quad (9)$$

The collision term I_a does not contribute in (9) because the number of quasiparticles is conserved in this process.

We now consider a superconductor in which the number of quasiparticles is not in equilibrium with the value of the energy gap Δ. In this case, as discussed above, we introduce an extra chemical potential μ_n' which determines the number of quasiparticles. It is most convenient to use a thermodynamic potential $\Phi(T,\mu,\Delta,\mu_n')$, as in the original work of BCS, except that now the extra chemical potential μ_n' appears. The electron density ρ and quasiparticle density n_n are determined from

$$d\Phi = -SdT - \rho d\mu - n_n d\mu_n'. \quad (10)$$

It should be noted that μ_n' corresponds to $\mu_n - \mu_s$ in (2). The energy gap is determined from

$$(\partial \Phi / \partial \Delta)_{T,\mu,\mu_n'} = 0. \quad (11)$$

It is possible to write down Φ explicitly by starting with a Hamiltonian $\mathcal{H} - \mu_n' \hat{n}_n$, instead of the BCS Hamiltonian $\mathcal{H}$, where $\hat{n}_n$ is an operator for the number of quasiparticles in the superconductor. It is not difficult to see that this amounts to replacing $f(E_k)$ by

$$f(E_k - \mu_n') = (1 + e^{\beta(E_k - \mu_n')})^{-1} \quad (12)$$

in the expression for Φ given by BCS. This indicates that the modified gap, Eq. (11), is now

$$1 = \frac{g}{V} \sum_k \frac{1 - 2f(E_k - \mu_n')}{2E_k}, \quad (13)$$

where g is the coupling constant and the number of quasiparticles is

$$n_n=\frac{2}{V}\sum_k f(E_k-\mu_n'). \tag{14}$$

The chemical potential μ_n' vanishes in equilibrium and measures the departure from equilibrium of the superconductor.

We now take (12) to be an approximate solution to the Boltzmann equation, Eq. (8). This requires the assumptions that the quasiparticles are in equilibrium among each other but not with the condensate and that the phonons are in equilibrium and hence merely act as a reservoir. These assumptions are not necessary for the further development of the theory but should often correspond to the physical situation. On substituting (12) in (9) and only retaining terms linear in μ_n' on the right-hand side we find

$$\partial n_n/\partial t=-\Gamma\mu_n'. \tag{15}$$

This equation relates the approach to equilibrium to the departure from equilibrium. The cross section Γ is easily evaluated using the method of BRT. We will only give the results at temperatures close to T_c and at low temperatures.

$$\Gamma=ACT^3,\qquad \beta\Delta\ll 1 \tag{16a}$$

$$=4\pi C(\Delta/k)^3e^{-2\beta\Delta},\quad \beta\Delta\gg 1 \tag{16b}$$

where

$$C=2mk^3\eta/\pi u^2p_F\hbar^4$$

and

$$A=\int_0^\infty dxdx'\frac{(x+x')^2e^{x+x'}}{(e^{x+x'}-1)(e^x+1)(e^{x'}+1)}\simeq 5.4.$$

Here η is the dimensionless electron-phonon coupling constant, u is the sound velocity, p_F is the Fermi momentum, and k is the Boltzmann constant. After performing one integration, A was evaluated numerically. Using typical values for the constants, $C\simeq 10^{42}$ erg^{-1} cm^{-3} sec^{-1}. At T_c, Eq. (16a) reduces to the electron-hole recombination time and the T^3 dependence arises from the phonon density. At low temperatures, Γ becomes exponentially small because of the small number of quasiparticles present.

Close to T_c it is more convenient to use Δ as a parameter than μ_n'. Then, expanding Eq. (13) in powers of Δ and retaining terms linear in μ_n', we find

$$\mu_n'=(2/\beta)[(a-b\Delta^2)/D(\Delta)], \tag{17}$$

where

$$a=\frac{T_c-T}{T_c},\quad b=\frac{7\zeta(3)}{8\pi^2}\beta^2,\quad D(\Delta)=\ln\frac{4}{\Delta\beta}+I.$$

I is the integral

$$I=2\int_0^\infty dx\,\ln x\,\text{sech}^2x\,\tanh x\simeq 0.1,$$

which was evaluated numerically. Note that μ_n' vanishes in equilibrium because the equilibrium value of Δ is given by $\Delta_{eq}^2=a/b$.

The density of excitations n_n in (15) varies most rapidly with Δ and μ_n' close to T_c rather than through the explicit dependence on T and μ. Equation (15) then becomes an equation for the rate of change of the energy gap Δ:

$$\left(\frac{\partial n_n}{\partial\Delta}+\frac{\partial n_n}{\partial\mu_n'}\frac{\partial\mu_n'}{\partial\Delta}\right)\frac{\partial\Delta}{\partial t}=-\Gamma\mu_n'. \tag{18}$$

The derivatives on the left-hand side are easily calculated from (14) in the limit $\beta\Delta\ll 1$ and on substituting in (18) we find

$$\frac{\beta^2\Delta}{D(\Delta)}\left[D^3(\Delta)+\frac{7\zeta(3)}{\pi^2}D(\Delta)-\frac{4}{\beta^2\Delta^2}(a-b\Delta^2)\right]\frac{d\Delta}{dt}=\frac{2\Gamma}{N_F}(a-b\Delta^2), \tag{19}$$

where N_F is the density of states of one spin at the Fermi surface and $D(\Delta)$ has been given above.

If $T<T_c$ and a is positive, Eq. (19) can be linearized around the equilibrium value of Δ:

$$\Delta(t)=(a/b)^{1/2}+\Delta_1(t).$$

The equation determining Δ_1 is then

$$\partial\Delta_1/\partial t=-(1/\tau)\Delta_1, \tag{20}$$

where

$$\frac{1}{\tau}=\frac{7\zeta(3)}{2\pi^2}\left(\frac{\Gamma}{N_F}\right)\frac{1}{D^2(\Delta_{eq})+7\zeta(3)/\pi^2},\quad(\beta\Delta\ll 1). \tag{21}$$

The superconductor relaxes exponentially in a time τ to its equilibrium state. The relaxation time τ behaves like $[\ln(T_c-T)/T_c]^2$ close to T_c and becomes long.

If $T>T_c$ and a is negative, the equilibrium value of Δ is zero. But if a gap was to form its decay, from (20), it would be determined by the equation

$$d\Delta/dt=-(\Gamma/2N_F)\Delta D(\Delta), \tag{22}$$

with the solution

$$\beta\Delta=\exp[\ln 4+I-ce^{\gamma t}], \tag{23}$$

where c is a constant and $\gamma=\Gamma/2N_F$. The order of magnitude of the relaxation time in each case is, from (16a),

$$\Gamma/N_F\simeq 10^8T^3\ \text{sec}^{-1}. \tag{24}$$

The theory may also be extended to low temperatures by linearizing (15) around the equilibrium value of the energy gap. An equation identical to (20) is obtained but with τ given by

$$1/\tau=(2\pi)^{1/2}(C/N_F)(\beta\Delta)^{5/2}e^{-\beta\Delta},\quad(\beta\Delta\gg 1), \tag{25}$$

where C is given below Eq. (16).

3. DISCUSSION

This relaxation process will be one contribution limiting the lifetime of a quasiparticle. Other contributions will arise from ordinary scattering processes. The broadening of the energy levels $\sim\hbar/\tau$ of the quasiparticles due to this relaxation is much smaller than Δ except very close to $T_c[(T_c-T)/T_c\simeq 10^{-6}]$.

These results can be used to examine the correlation in time of the fluctuations of the magnitude of the order parameter. Thus, from (20), the correlation function

$$\langle\Delta_1(t)\Delta_1(0)\rangle=\langle\Delta_1^2(0)\rangle e^{-t/\tau}, \quad (26)$$

and decays exponentially. The amplitude $\langle\Delta_1^2(0)\rangle$ can be calculated by a thermodynamic argument[9]:

$$\langle\Delta_1^2(0)\rangle=kT(\partial^2\Phi/\partial\Delta^2)^{-1}=kT/4N_FVb\Delta_{eq}^2, \quad (27)$$

where V is the volume of the superconductor. The amplitude of the fluctuations given by (27) is very small. For a specimen with $V=1$ cm^3 it is found that $\langle\Delta_1^2(0)\rangle=\Delta_{eq}^2$ only when $(T_c-T)/T_c\simeq 10^{-9}$. Such fluctuations would lead to a lack of sharpness in the transition temperature of a small specimen.

An interesting question which arises is how rapidly the phase of the order parameter in a superconductor diffuses. We will consider here the phase diffusion due to the coupling of the amplitude and the phase of the order parameter. Other sources of phase diffusion are electromagnetic, density, and temperature fluctuations.

As shown by Gor'kov,[10] the time rate of change of the phase is determined by the superfluid chemical potential μ_s. Thus if

$$\Delta(t)=|\Delta(t)|e^{i\omega(t)}, \quad (28)$$

then

$$\hbar\partial\omega/\partial t=2\mu_s. \quad (29)$$

Close to T_c the superfluid density is given by

$$n_s=2nb|\Delta|^2,$$

where n is the total electron density and b is given below Eq. (17). Thus

$$\mu_s=\mu+(1/2nb)\partial\Phi/\partial|\Delta|^2$$
$$=\mu+(3m/4bp_F^2)(-a+b|\Delta|^2). \quad (30)$$

Substituting Eq. (30) into (29) and linearizing the right-hand side by writing $|\Delta|=\Delta_{eq}+\Delta_1(t)$, where Δ_{eq} is the equilibrium value of $|\Delta|$, we find

$$\hbar\partial\omega/\partial t=2\mu+(3m\Delta_{eq}/p_F^2)\Delta_1(t). \quad (31)$$

As we are neglecting temperature and density fluctuations, μ is a constant and $\omega=2\mu t/\hbar+\omega'(t)$, where

$$\hbar\partial\omega'/\partial t=(3m\Delta_{eq}/p_F^2)\Delta_1(t). \quad (32)$$

To determine the correlation function

$$\langle\exp\{i[\omega(t_1)-\omega(t_2)]\}\rangle=\exp[(2i\mu/\hbar)(t_1-t_2)] \times\langle\exp\{i[\omega'(t_1)-\omega'(t_2)]\}\rangle,$$

we will make a Gaussian approximation and replace this by

$$\exp[(2i\mu/\hbar)(t_1-t_2)]\exp[-\tfrac{1}{2}\langle[\omega'(t_1)-\omega'(t_2)]^2\rangle]. \quad (33)$$

From Eq. (32),

$$\langle[\omega'(t_1)-\omega'(t_2)]^2\rangle=\left(\frac{3m\Delta_{eq}}{\hbar p_F^2}\right)^2\int_{t_2}^{t_1}dt\int_{t_2}^{t_1}dt' \times\langle\Delta_1(t)\Delta_1(t')\rangle. \quad (34)$$

Substituting from (26) and supposing that $(t_1-t_2)\gg\tau$, we find

$$\langle[\omega'(t_1)-\omega'(t_2)]^2\rangle=D|t_1-t_2|, \quad (35)$$

where

$$D=2(3m\Delta_{eq}/\hbar p_F^2)^2\langle\Delta_1^2(0)\rangle\tau. \quad (36)$$

When $\langle\Delta_1^2(0)\rangle$ is substituted from (27) it is seen that this result is independent of Δ_{eq}. Substituting numerical values for the parameters appearing in (36), we find

$$D\simeq 10^{-7}\tau/V, \quad (37)$$

when V is the volume of the superconductor. For $\tau=10^{-8}$ sec, we then get a slow diffusion of the phase.

The relaxation equation (19) has the form of a diffusion equation. In the presence of fields a term arising from the kinetic energy of the supercurrents proportional to $(\nabla-(2ie/c)\mathbf{A})^2\Delta$ must be included on the right-hand side. It is natural to expect a first time derivative because then the future state of the system is determined by the value of Δ and other thermodynamic variables. It has the unphysical property that disturbances propagate with an infinite velocity. It has been shown that second time derivatives of Δ do appear,[5,11] but because of the large damping, a diffusion equation is a good approximation.

Close to T_c, an equation similar to (20) has been derived by Abrahams and Tsuneto[5] in which the relaxation of the quasiparticles is brought about by time-dependent fields. The relaxation time for this process is

$$\frac{1}{\tau_a}=\frac{14\zeta(3)kT_c}{\pi^3\hbar}\left(\frac{\Delta_{eq}}{kT_c}\right)^2, \quad \beta\Delta\ll 1, \quad (38)$$

with the restriction that the frequency ω of the disturbance must be greater than $2\Delta_{eq}$. Comparison with (21) shows that for low-frequency disturbances the phonon mechanism of relaxation will dominate. The mechanism considered by Abrahams and Tsuneto would be responsible for infrared absorption in the superconductor.

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It is interesting to note that there is a close similarity between a superconductor and a laser and that many of the equations appearing here are analogous to the equations used in the description of lasers.[12] In that case the role of Δ is played by the electric field $\mathbf{E}$. Both systems can in many respects be regarded as nonlinear oscillators.[13] At its operating point the impedance of a self-sustaining oscillator is zero and the vanishing of the resistance and the reactance correspond, respectively, to Eqs. (19) and (29).

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Isospin Formulation of the Theory of a Granular Superconductor

R. H. Parmenter*
RCA Laboratories, Princeton, New Jersey
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The properties of a granular superconductor are studied with the aid of the isospin formulation of the microscopic theory of superconductivity. The system consists of grains of homogeneous superconductor separated by insulating but tunnelable barriers (Josephson junctions). The general nonlinear equations of motion are set up for the isospins, "spin up" representing the absence, and "spin down" the presence, of a given Cooper pair. These equations are like torque equations for each isospin moving in an effective pseudomagnetic field due to all the other isospins. Linearized solutions result in various single-particle and collective excitations. A certain class of nonlinear solutions is shown to satisfy a Ginzburg-Landau-like differential equation. The effects of electric fields (within the junctions) and real magnetic fields are studied, one result being that there are bulk electromagnetic modes, analogous to the surface modes known to be associated with a single isolated Josephson junction. Consequences of changes in temperature and changes in effective electron-electron interaction are studied.

I. INTRODUCTION

In this paper we wish to examine the properties of a particular kind of granular superconductor; namely, one where each grain consists of a homogeneous superconductor, but at each grain boundary there is a thin insulating layer (e.g., oxide). Each layer is thin enough that it can be tunneled by the Cooper pairs of the superconductor; in other words, we have a Josephson junction[1] at each grain boundary. For simplicity, we assume that the junctions take up a negligible fraction of the total volume of material.

For such a superconductor, the energy density of the BCS theory[2] is augmented by a tunneling-energy density, the latter being directly proportional both to the linear density of tunnel junctions[3] and to the Cooper-pair transition amplitude for an average junction of unit area. We are free to imagine the tunneling-energy density as large or as small as we like, because of variations in the number of junctions per unit length. We cannot, however, let the tunneling energy be either too large or too small because of the tunneling transition probability. The upper limit is set by the limitation of second-order perturbation theory (the Cooper-pair tunneling being visualized as a two-step process,[4] the intermediate step involving the virtual state where only one of the two electrons composing the pair has tunneled). When the tunneling transition probability is too high, perturbation theory breaks down.

The lower limit to the tunneling transition probability is set by a physical process that has nothing to do with superconductivity per se; it is the value of the tunneling probability at which the normal-metal conductivity of the system (at temperatures where the normal phase is thermodynamically stable) switches over to *insulating* behavior, because there is a thermal activation energy associated with electron tunneling.[5] This activation energy is the energy required to change two neighboring,

* Present address: Department of Physics, University of Arizona, Tucson, Arizona.
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