ENHANCEMENT OF SUPERCONDUCTIVITY BY EXTRACTION OF NORMAL CARRIERS

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One can ask the question: Why does a sufficiently high temperature always lead to quenching of superconductivity in a metal? An examination of the Bardeen-Cooper-Schrieffer (BCS) theory of superconductivity¹ shows that the answer to this question is as follows: The phonon-induced electron-electron attraction between electrons leads to a resonant, or coherent, interaction between the one-electron states of the metal. The greater the number of states interacting, the greater will be the resonant interaction, and thus the greater the lowering of the energy of the assembly of superconducting electrons of the metal. As the temperature is raised, electrons are thermally excited out of this sea of superconducting electrons, thus giving rise to normal electrons and holes. The one-electron states occupied by these normal carriers are no longer available for the resonant interaction (because of the exclusion principle), thereby decreasing the strength of the resonant depression of the energy of the system. At a sufficiently high temperature (the superconducting transition temperature), there are sufficient normal carriers present to quench the resonance completely, whereupon the metal becomes normal.

This suggests that, if there were some method of electrically extracting the normal carriers as soon as they are thermally generated, then it might be possible to maintain the metal superconducting above the usual transition temperature. As a result of recent work,² it is known that it is indeed possible to inject or extract normal carriers from a superconductor. (It is this phenomenon which insures the success of the operation of the superconducting tunnel diode.) This is done by making contact to another superconductor through a thin (10-20 angstroms) insulating layer. the latter serving to pass normal carriers under suitable bias (by tunneling), but to block superconducting electrons completely. It appears that an insulating barrier more than a few angstroms thick is effective in preventing current flow involving the cooperative motion of many electrons (i.e., the superconducting current) while at the same time allowing some current flow involving uncorrelated motions of individual carriers (i.e., the normal current).

Let us suppose we wish to extract normal car-

riers from superconductor A having transition temperature T_{cA} . We assume that the operating temperature T is close to (but smaller than) T_{cA} in order that there be appreciable numbers of normal carriers in A under equilibrium conditions. Through a thin insulating layer, we make contact with superconductor B having transition temperature T_{CB} sufficiently greater than T for there to be negligible numbers of normal carriers present in it. By now biasing such a diode with a voltage equal to one-half the energy-gap difference of the two superconductors, we can extract either normal electrons or normal holes (depending on the polarity of the bias) from A. By having A in the form of a film sandwiched between contacts (electrodes) of superconductor B (along with the thin insulating layers), we can extract both signs of normal carriers by putting a bias between the two B contacts equal to or greater than the energygap difference $2(\epsilon_{0B} - \epsilon_{0A})$. If the bias is made greater than the energy-gap sum $2(\epsilon_{0B} + \epsilon_{0A})$, however, we get an additional effect due to generation of isoenergetic electron-hole pairs at each insulating interface-this effect leading to a double injection of normal carriers into A. This injection apparently will overbalance the previously discussed double extraction, so that for biases greater than $2(\epsilon_{0B} + \epsilon_{0A})$, there will be a net injection rather than extraction. Figure 1 illustrates the energy-level diagrams for such a device under conditions of zero, $2(\epsilon_{0B}-\epsilon_{0A})$, and $2(\epsilon_{0B} + \epsilon_{0A})$ bias.

We designate by τ the lifetime against normal electron-hole recombination and by n' the density of normal electrons (or holes) in a superconductor under thermal equilibrium conditions. Thus n'/τ is the normal electron-hole pair generation rate per unit volume in the superconductor. (It is also the recombination rate.) Under conditions of extraction in the superconductor, we shall, for the moment, assume the normal pair generation rate is unchanged (i.e., that it remains n'/τ). The recombination rate, however, will be reduced to $(n''/n')^2(n'/\tau)$, where n'' is the reduced density of normal electrons resulting from the extraction process. The factor $(n''/n')^2$ is a consequence of the fact that we are dealing with a bimolecular recombination process. We designate by L the thickness of the superconducting film



FIG. 1. Energy level diagrams for a system of a superconducting film of thickness *L* and energy gap $2\epsilon_{0A}$ separated by insulating layers from superconducting contacts having energy gap $2\epsilon_{0B}$. (a) Zero bias. (b) Bias = $2(\epsilon_{0B} - \epsilon_{0A})$. (c) Bias = $2(\epsilon_{0B} + \epsilon_{0A})$.

being subjected to extraction, by γ the mean tunneling transition probability per collision of the normal carriers striking the insulating barriers, and by v_F the velocity of the normal carriers (this being the Fermi velocity). The pair extraction rate per unit volume of superconducting film is $\frac{1}{3}v_F(n''/2)(\gamma/L)$. Upon setting the difference between the generation rate and the recombination rate equal to the extraction rate (i.e., dynamic equilibrium), we get

$$\left[1 - \left(\frac{n''}{n'}\right)^2 - \left(\frac{\gamma\lambda}{6L}\right)\left(\frac{n''}{n'}\right)\right]\left(\frac{n'}{\tau}\right) = 0, \qquad (1)$$

where $\lambda = v_F \tau$ is the mean free path against pair recombination under thermal equilibrium conditions. If $\gamma \lambda / 6L \gg 1$, then

$$n''/n' = 6L/\gamma\lambda \ll 1, \qquad (2)$$

and the extraction process is very efficient. An

order-of-magnitude lower limit to λ is given by the normal-state bulk electrical conductivity mean free path λ_n as limited by lattice vibrations, λ_n being a lower limit since it is harder to generate normal pairs thermally in a superconductor than in the corresponding normal metal. The real phonon absorbed in the generation process must have at least the gap energy in the case of the superconductor. Thus if the operating temperature is appreciably below the usual transition temperature of the superconductor, we can expect $\lambda \gg \lambda_n$. Since λ_n may be 10⁻² cm and γ $\sim 10^{-2}$ - 10^{-3} under typical conditions, Eq. (2) shows that L, the superconducting film thickness, may be 100 angstroms or more and still have $(n''/n') \ll 1.$

In carrying out the analysis of the extraction process, we have assumed spatial uniformity of n'' in the superconducting film. This will be an excellent approximation as long as $L \ll \lambda$. We also assumed the pair generation rate to be unaffected by the extraction process. As we shall see in a moment, however, the superconducting energy gap may increase under conditions of extraction. If so, this implies a decrease of the generation rate with increasing extraction (fewer phonons are energetic enough to create pairs). Such a decrease of the generation rate obviously does not invalidate the conclusions of the preceding paragraph.

A third assumption is that the density of normal carriers in the contacts is negligible compared with n'', the density in the superconducting film, so that there is negligible tunneling of normal carriers from the contacts back into the film. This implies that there is an efficient method of getting rid of the normal carriers injected into the contacts. One way of accomplishing this is to plate each contact with a certain amount of normal metal. The latter will serve as an efficient sink for normal carriers injected into the contact, whenever the distance from the tunnelable barrier facing the contact is less than a mean free path (for normal carriers in S_B) from the normal metal of the contact. The normal metal should be separated from the tunnelable barrier by a thickness of superconductor B of at least something of the order of the Pippard coherence distance (10^{-4} cm) , and the normal metal itself should have minimum thickness of similar size. This obviates the possibility that the normal metal is made superconducting by being in contact with S_B , or conversely that S_B has its superconductivity quenched by being in contact with a

normal metal.³

It should be pointed out that the superconductor S_B in each contact contains injected normal electrons or normal holes, but not both. (One contact will have electrons, and the other holes.) The resulting space charge in each contact will be compensated by an adjustment in the density of superconducting electrons (a shift of the energy of the bottom of the conduction band in S_B relative to the Fermi level). This will lead to a minute shift in the transition temperature of each contact.⁴ Such a shift will be negligible compared with the shift of transition temperature of the superconducting film S_A due to extraction of normal carriers of both signs without change of the density of superconducting electrons.

The situation we are considering in this note represents a nonequilibrium condition. There are two general types of nonequilibrium conditions in a superconductor: (1) those where the lack of equilibrium is due to the superconducting electrons, so that dissipation processes do not occur; (2) those where the lack of equilibrium is due to the normal carriers, so that dissipation processes do occur (i.e., finite entropy production). Time-independent current flow in a superconducting wire is an example of the first type.⁵ Because of the lack of dissipation, it is possible to define a free energy, despite the lack of equilibrium. Specifically, one modifies the equilibrium free energy by adding a term equal to the product of the quantity being constrained times a Lagrangian multiplier. (The constraint, set by boundary conditions, is what causes disequilibrium.) For the example of dc current flow in a superconducting wire, the constraint is that imposed on the net current due to the superconducting electrons.⁶

The extraction of normal carriers represents a nonequilibrium situation of the second type where dissipation processes occur. There is a certain amount of thermoelectric cooling in the superconducting film being exhausted of normal carriers, and a still greater amount of heating in the contacts. For example, under the bias conditions of Fig. 1(b), heat is being removed from the film at the rate $I(2\epsilon_{0A}/e)$ and being liberated in each contact at the rate $I(\epsilon_{0B}/e)$ when extraction is efficient. (Here I is the total current, and e is the electronic charge.) The removal of heat in the film occurs when phonons are absorbed in making normal hole-electron pairs. Because of backflow of heat from the contacts into the film, the actual temperature drop of the film is probably small enough to ignore. Nevertheless, the

presence of dissipation associated with this thermoelectric process makes it difficult to define any sort of free energy F such that the minimization of F will lead to a description of steadystate equilibrium.

A generalization of the BCS theory to such a nonequilibrium situation can be made in the following manner. A Boltzmann transport equation is set up for the distribution function f_k associated with the normal carriers (normal electrons for $k > k_F$, and normal holes for $k < k_F$, \vec{k} being the wave vector labeling the single-particle states, and k_F being the Fermi wave vector). At the same time, the internal energy U is minimized with respect to the parameters h_k appearing in the BCS many-electron wave functions.⁷ Such a minimization leads to the equation

$$[N(0)V]^{-1} = \int_{0}^{\hbar\omega} (\epsilon_{k}^{2} + \epsilon_{0}^{2})^{-1/2} (1 - 2f_{k}) d\epsilon_{k}.$$
 (3)

Here N(0)V and $\hbar\omega$ are constraints characteristic of the superconductor, ϵ_k is the single-electron Bloch energy (measured with respect to the Fermi level), $2\epsilon_0$ is the energy gap, and $(\epsilon_k^2 + \epsilon_0^2)^{1/2}$ is the energy of a normal carrier in the superconductor (also measured with respect to the Fermi level). Equation (3) is formally the same as that of the BCS theory; the only difference lies in the fact that the f_k contained therein is obtained from a Boltzmann transport equation rather than from a minimization of a free energy with respect to f_k .

Of course, it is possible to have a nonequilibrium situation where both the superconducting electrons and the normal carriers contribute to the disequilibrium. For such a situation, one should solve a Boltzmann equation for f_k and at the same time minimize the internal energy U with respect to h_k subject to the constraint on the superconducting electrons causing their disequilibrium. Such a situation arises in the particular physical problem of the superconducting film with extracting electrodes if we choose to make a supercurrent flow in the film parallel to the plane of the film at the same time that we are extracting normal carriers from the film.

For the moment, we return to the case where there is no supercurrent in the superconducting film. Boltzmann's equation for our problem reduces to the statement that the total time rate of change of f_k vanishes, the three processes of normal-pair thermal generation, normal-pair recombination, and double extraction of normal carriers all contributing to (df_k/dt) . The extraction process is effective in lowering f_k for all orientations of \vec{k} , despite the fact that only carriers moving nearly normal to the insulating interface have appreciable probability of tunneling. This lowering of f_k for all orientations is a consequence of the fact that those carriers reflected at the insulating interface are undoubtedly reflected diffusely, thus leading to rapid equilibration between isoenergetic carriers moving in different directions.

It is not really necessary to solve Boltzmann's equation for f_k . Under reasonable physical conditions, it appears that whenever extraction is efficient as a whole, i.e., whenever $(n''/n') \ll 1$, it will also happen that

$$f_k \ll 1. \tag{4}$$

This will hold for all \vec{k} if the bias is adjusted such that normal carriers of all energies can tunnel [e.g., as in Fig. 1(b)]. [In contrast, under noextraction conditions where n' is appreciable, there will be a number of f_k comparable with $\frac{1}{2}$ and thus not satisfying (4).] It should be noted that the only way the temperature T can affect the energy gap $2\epsilon_0$, obtained by solving Eq. (3), is through the temperature dependence of f_k . However, as long as (4) holds true, then (3) will be independent of T, and we will obtain an ϵ_0 equal to the BCS value at the absolute zero of temperature, even when T is above the usual superconducting transition temperature.

It would appear that, by means of electrical extraction, we can raise the transition temperature of the superconducting film. An upper limit to this new transition temperature will, of course, be set by T_{cB} , the transition temperature of the contacts. As the operating temperature approaches T_{cB} , the extraction efficiency will drop off because of the presence of thermally generated normal carriers in the contacts which may tunnel through the insulating layers into the superconducting film. It appears plausible that one should be able to start with the film in the normal state and by means of extraction cause the film to go superconducting, the operating temperature being less than T_{cB} but greater than T_{cA} (the usual transition temperature of the film). Conversely, we should be able to lower the transition temperature of the film by injection of normal carriers as shown in Fig. 1(c). By lowering the transition temperature below the operating temperature, we can drive the superconducting film normal by electrical injection. Using either injection or extraction, we have an electrical (nonmagnetic) method of controlling a superconducting current in the film (the current flowing parallel to the plane of the film).

¹J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. <u>108</u>, 1175 (1957).

²J. Nicol, S. Shapiro, and P. H. Smith, Phys. Rev. Letters <u>5</u>, 461 (1960); I. Giaever, Phys. Rev. Letters <u>5</u>, 464 (1960); I. Giaever and K. Megerle, Phys. Rev. <u>122</u>, 1101 (1961).

³R. H. Parmenter, Phys. Rev. <u>118</u>, 1173 (1960); P. H. Smith, S. Shapiro, J. L. Miles, and J. Nicol, Phys. Rev. Letters <u>6</u>, 686 (1961).

⁴R. E. Glover, III, and M. D. Sherrill, Phys. Rev. Letters 5, 248 (1960).

⁵J. Bardeen, Phys. Rev. Letters <u>1</u>, 399 (1958).

⁶In reference 5, it appears that the constraint is being imposed on the normal current rather than the superconducting current. This is because the calculation, for convenience, is being carried out in a coordinate system moving with the mean drift velocity of the superconducting electrons. In the laboratory coordinate system, the constraint is being imposed on the supercurrent, not the normal current.

⁷The internal energy *U* is formally the same as in the BCS theory, i.e., Eq. (3.16) of BCS after removal of the term *-TS*. In the BCS theory, the free energy F = U - TS, rather than *U*, is minimized with respect to h_k . This difference is of no consequence, since the entropy *S* is independent of h_k . The minimization of *U* with respect to h_k appears to be the only reasonable way of generalizing the BCS variational approach to a nonequilibrium situation. Such a minimization nevertheless represents an assumption.