with K. Lassmann, J. Buck, and W. Forkel.

[1]L. R. Testardi, Phys. Rev. B 4, 2189 (1971).
[2]W. H. Parker and W. D. Williams, Phys. Rev. Lett. 29, 924 (1972).
[3]G. A. Sai-Halasz, C. C. Chi, A. Denenstein, and D. N. Langenberg, Phys. Rev. Lett. 33, 215 (1974).
[4]M. Goetze, M. Nover, and O. Weis, Z. Phys. B25, 1 (1976).
[5]I. Giaever, Phys. Rev. Lett. 5, 147 (1960).
[6]Corresponding calculations with respect to phonon emission spectra are H. Kinder, K. Lassmann, and W. Eisenmenger, Phys. Lett. 31A, 475 (1970); A. H. Dayem and J. J. Wiegand, Phys. Rev. B 5, 4390 (1972); M. Welte, thesis, Universität Stuttgart, 1976 (unpublished).
[7]C. S. Owen and D. J. Scalapino, Phys. Rev. Lett. 28, 1559 (1972).
[8]A. Rothwarf and B. N. Taylor, Phys. Rev. Lett. 19, 27 (1967).
[9]W. H. Parker, Phys. Rev. B 12, 3667 (1975).
[10]P. Hu, R. C. Dynes, and V. Narayanamurti, Phys. Rev. B 10, 2786 (1974).
[11]The gap instability is also observed in the Sn-I-Sn I-V characteristic. After a first strong steplike voltage increase follow a few closely spaced smaller steps until complete normal conductance is reached.
[12]H. J. Trumpp, K. Lassmann, and W. Eisenmenger, Phys. Lett. 41A, 431 (1972).
[13]Quasiparticles at the gap edge have maximal depairing influence.
[14]By evaluating the tunneling integral for the Sn-I-Pb junction we found that the simple thermal calibration of the current minimum for determing the quasiparticle density in the injection case, in our measurement range, leads to systematic errors of less than +4%. The more accurate calibration with equal energy gaps in the thermal and in the injection case (Fig. 3) and assuming proportionality between tunneling currents and quasiparticle densities is subject to systematic errors of less than +1.5%.
[15]W. Eisenmenger and A. H. Dayem, Phys. Rev. Lett. 18, 125 (1967); W. Eisenmenger, in *Physical Acoustics*, edited by W. P. Mason and R. N. Thurston (Academic, New York, 1976), Vol. XII, p. 79.
[16]1000-Å Sn films show less than 3% reduction of the quasiparticle recombination lifetime in a parallel field of 50 Oe (W. Eisenmenger, unpublished) and less than 1% energy gap reduction [R. C. Dynes and V. Narayanamurti, Phys. Rev. B 6, 5143 (1972)].
[17]B. K. Miller and A. H. Dayem, Phys. Rev. Lett. 18, 1000 (1967).
[18]W. Eisenmenger, K. Lassmann, H. J. Trumpp, and R. Krauss, Appl. Phys. 12, 163 (1977).
[19]M. Welte, to be published; the gap instablity of the μ^* model follows also directly from the solution of the proper rate equations.
[20]J. J. Chang and D. J. Scalapino, Phys. Rev. B 10, 1447 (1974).
[21]V. S. Elesin, Zh. Eksp. Teor. Fiz. 66, 1755 (1974) [Sov. Phys. JETP 39, 862 (1974)].

Stability of Radiation-Stimulated Superconductivity

Albert Schmid*
Institut für Physik, Universität Dortmund, West Germany
(Received 10 May 1976)

Recent observations of radiation-stimulated superconductivity above the thermodynamic transition temperature reveal the existence of a first-order phase transition between a normal and a superconducting state. In this Letter, a Langevin equation for the order parameter is derived, which is appropriate for the nonequilibrium steady-state process under consideration, and which explains the observed discontinuous phase transition.

The prediction of Eliashberg[1] on radiation-stimulated superconductivity seems now to have found acceptance from the experimental point[2,3] of view, particularly since superconductivity above the transition temperature has been observed[3] in a homogeneous material. It is well known that thermally excited electrons and holes oppose superconductivity; this explains the phase transition at finite temperatures. Less known, however, is the fact that excitations near the gap edge are more detrimental to superconductivity than those which are off the edge. Quite generally, a classical radiation field tends to spread the energy distribution of the excitations and thus remove them from the gap edge. In essence, this is the explanation of radiation-stimulated superconductivity.

In a certain range of temperatures, Eliashberg's theory allows three solutions of the gap equation, a situation which reminds us of van der Waal's theory of real gases. Furthermore, Klapwijk, van der Berg, and Mooij[3] have observed a discontinuous transition between the normal and superconducting states, including hysteresis, which is similar in appearance to a first-order phase transition. However, standard thermodynamic stability theory cannot be applied, as these are nonequilibrium steady states,[4] and therefore it is necessary to resort to techniques which can be found illuminatingly reviewed in a recent article by Haken.[5] This means that we have to construct a Langevin equation for the order parameter which includes time dependence and random force, both of which can be found by generalizing Eliashberg's theory. The origin of the random force is the fluctuations of the quasiparticle occupation numbers. This equation allows us to calculate the stationary probability distribution of the order parameter, which will be written in the form $W(\Delta)\propto \exp[-\mathfrak{F}(\Delta)/T_c]$. Obviously, the quantity $\mathfrak{F}$ generalizes the free energy concept to nonequilibrium steady states, and allows us to answer questions on the stability of nonequilibrium steady states in a most familar way.

As an introduction, I briefly review the theory of Eliashberg. This theory is based on the BCS gap equation,

$$\Delta/\lambda=\int d\epsilon_p(\Delta/2E_p)[1-2\langle n_{\vec{p}}\rangle], \qquad (1)$$

where $E_p=(\epsilon_p{}^2+\Delta^2)^{1/2}$. Furthermore $n_{\vec{p}}$ is the quasiparticle distribution function and $\langle n_{\vec{p}}\rangle$ its angular average. Introducting $n_p=n_0(E_p)+\delta n_{\vec{p}}$, where n_0 is the Fermi function, and

$$\chi=-\int d\epsilon_p(1/E_p)\langle\delta n_{\vec{p}}\rangle, \qquad (2)$$

one can, provided that $\Delta \ll T_c$, transform Eq. (1) into an equation of the Ginzburg-Landau type,

$$[(T_c-T)/T_c-0.106(\Delta/T_c)^2+\chi]\Delta=0, \qquad (3)$$

where $0.106=7\zeta(3)/8\pi^2$. The above relation demonstrates clearly that a shift of quasiparticles to higher energies does, indeed, stimulate superconductivity.

In the stationary case (and also for a spatially homogeneous situation), Eliashberg calculates n_p from a rate equation (Boltzmann equation) of the form

$$\dot{n}_{\vec{p}}=(\dot{n}_{\vec{p}})^{\text{coll}}+(\dot{n}_{\vec{p}})^{\text{rad}}=0, \qquad (4)$$

where the time rates of change take into account the interaction of the quasiparticles with phonons (which also play the role of a heat reservoir) and with an electromagnetic radiation field. Note that the time rates of change include scattering as well as generation and recombination of quasiparticles.

Close to T_c, spectacular phenomena can be observed even for small radiation power, and hence for small $\delta n_{\vec{p}}$ as well. Assuming this to be the case, we may linearize the collision integral, $(\dot{n}_{\vec{p}})^{\text{coll}}$, with respect to $\delta n_{\vec{p}}$, and we may also retain only the leading contribution to the radiative term, $(\dot{n}_{\vec{p}})^{\text{rad}}$, which is then a functional of $n_0(E_p)$. Thus, Eq. (4) becomes an inhomogeneous, linear, integral equation. It is important now to realize that the most important part of $\delta n_{\vec{p}}$ is confined to a rather narrow energy range $E_p \lesssim O(\Delta)\ll T_c$. In such a case, it is possible to approximate the collision integral by the relaxation approximation,

$$(\dot{n}_{\vec{p}})^{\text{coll}}=-\delta n_{\vec{p}}/\tau_0, \qquad (5)$$

where τ_0 is the inelastic electron-phonon collision time at the Fermi level (and at T_c).

Inserting the solution $\delta n_{\vec{p}}$ of Eq. (4) into Eq. (2), we obtain $\chi^{(\text{rad})}$, which is proportional to the absorbed radiation power [and which is denoted by $(\Delta/T_c)F$ in the second article of Ref. 1].

The normal state with $\Delta=0$ is always a solution of the Ginzburg-Landau equation (in which χ has been replaced by the definite form $\chi^{(\text{rad})}$). Superconducting-state solutions with $\Delta\neq 0$ can be found from a graphical construction as shown in Fig. 1. There is one solution for $T<T_c$, two solutions for $T_c<T<T_M$, and no solutions for other temperatures.[6]

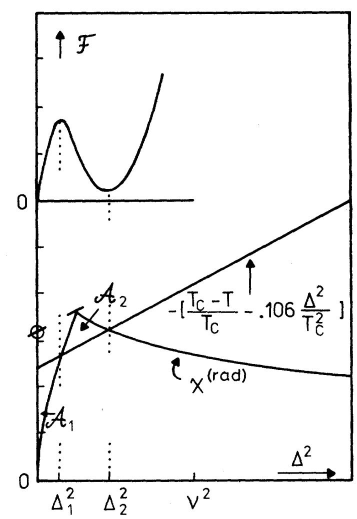

FIG. 1. The intersections of the straight line $(T_c-T)/T_c-0.106\Delta^2/T_c$, with the curve $\chi^{(\text{rad})}$ determine the solutions Δ_1 and Δ_2 of the stationary Ginzburg-Landau equation. Note the kink in $\chi^{(\text{rad})}$ at $2\Delta=\nu$, where ν is the radiation frequency. For a given radiation power, there are superconducting solutions only if $T<T_M$, where $(T_M-T_c)/T_c$ is marked by φ on the ordinate. The inset shows the free energy $\mathfrak{F}$ which reveals that the normal state is globally stable. Equibalent to $\mathfrak{F}(\Delta_2)>0$ is the condition $\mathcal{A}_1>\mathcal{A}_2$ for the two areas.

Using this theory of Eliashberg as a foundation, I discuss the stability of the solutions found above. This necessitates that the quantities depend on time. For instance, $\dot{n}_{\vec{p}}$ of Eq. (4) now differs from zero; specifically,

$$\dot{n}_{\vec{p}} = n_0'(E_p)(\Delta\dot{\Delta}/E_p) + \delta\dot{n}_{\vec{p}}. \tag{6}$$

In the following, I will exploit in an essential way the fact that close to the transition temperature the order parameter changes only slowly in time. For instance, its relaxation time in the vicinity of thermal equilibrium states,[7]

$$\tau_R = 3.7(T_c/\Delta)\tau_0, \tag{7}$$

is very long in comparison with the quasiparticle relaxation time τ_0. Therefore, we may neglect $\delta\dot{n}_{\vec{p}}$ in Eq. (6); this corresponds to the situation where the quasiparticle occupation numbers $n_{\vec{p}}$ follow adiabatically the changes of the order parameter.[8]

Mathematically, the term $n_0'(\Delta\dot{\Delta}/E_p)$ plays the role of an inhomogeneous term in the linearized Boltzmann equation. This term is rather compact with respect to its energy dependence. This allows us to solve the linearized Boltzmann equation in the relaxation approximation and to calculate easily

$$\chi^{(\Delta)} = -(\pi/4T_c)\tau_0\dot{\Delta}. \tag{8}$$

Inserting $\chi = \chi^{(\mathrm{rad})} + \chi^{(\Delta)}$ into Eq. (3), we obtain a time-dependent Ginzburg-Landau equation which we may use to check Eq. (7). Also, it is not difficult to prove that in the temperature range $T_c < T < T_M$ the state with order parameter Δ_1 is absolutely unstable, since any infinitesimal fluctuation will grow exponentially in time. On the other hand, the normal state with $\Delta_0 = 0$ and the superconducting state with Δ_2 are locally stable, by which I mean that infinitesimal fluctuations decay in time.

A criterion for absolute stability can only be found if one includes the finite fluctuations of $\hat{\chi}$. (Instantaneous values of fluctuating quantities are marked by a caret, and the average value by a bracket. Hence, $\langle\langle\hat{\chi}\rangle\rangle = \chi$). Since for fermions[9]

$$\langle\langle \hat{n}_{\vec{p}} n_{\vec{p}'}\rangle\rangle = \begin{cases} n_{\vec{p}}, & \text{if } \vec{p} = \vec{p}', \\ n_{\vec{p}} n_{\vec{p}'}, & \text{if } \vec{p} \neq \vec{p}', \end{cases} \tag{9}$$

we find on the basis of Eq. (2)

$$\langle\langle(\delta\hat{\chi})^2\rangle\rangle = \langle\langle(\hat{\chi})^2\rangle\rangle - \chi^2 = (2N_0\Omega)^{-2}\textstyle\sum_{\vec{p}\sigma} E_p^{-2} n_{\vec{p}}(1-n_{\vec{p}}) = \pi/8N_0\Omega\Delta, \tag{10}$$

where $(2N_0\Omega)^{-1}\sum_{\vec{p}\sigma}\ldots$ has been substituted for $\int d\epsilon_p\ldots$ and where Ω is the volume of the system. Evidently, only states with $E_p \lesssim O(\Delta) \ll T_c$ contribute to this expression and, hence, $n_{\vec{p}} = \frac{1}{2}$, independent of the radiation power within the limits set up previously. Again, we meet here a situation where the relaxation approximation may be used in calculating the time dependence of the fluctuations of the occupation numbers. It follows that τ_0 is also the correlation time of $\delta\hat{\chi}$, which means that its power spectrum is given by

$$\langle\langle|\delta\hat{\chi}|^2\rangle\rangle_\omega = [2\tau_0/(1+\omega^2\tau_0^2)]\langle\langle(\delta\hat{\chi})^2\rangle\rangle. \tag{11}$$

Since $\delta\hat{\chi}$ is a sum of a very large number ($\propto\Omega$) of independently fluctuating quantities, it corresponds to a Gaussian process which is completely specified by Eq. (11). As already pointed out, the order parameter varies slowly in time. Hence, $\omega \sim \tau_R^{-1} \ll \tau_0$, and the term $\omega^2\tau_0^2$ may be neglected in Eq. (11).

Inserting $\chi = \chi^{(\mathrm{rad})} + \chi^{(\Delta)} + \delta\hat{\chi}$ into Eq. (3), we obtain for the instantaneous value $\hat{\Delta}$ of the order parameter an equation which couples the motion of $\hat{\Delta}$ to a random force with an effectively white spectrum. Explicitly, this nonlinear Langevin equation is as follows:

$$(\pi\tau_0/4T_c)\hat{\Delta}\dot{\hat{\Delta}} = [(T_c - T)/T_c - 0.106\hat{\Delta}^2/T_c^2 + \chi^{(\mathrm{rad})}]\hat{\Delta} + (\pi\hat{\Delta}\tau_0/4N_0\Omega)^{1/2}\hat{\eta}(t), \tag{12}$$

where $\langle\langle\hat{\eta}(t)\hat{\eta}(t')\rangle\rangle = \delta(t-t')$.

Following the standard procedure,[10] one obtains the stationary probability distribution $W(\Delta)$ of the order parameter,

$$W(\Delta) = \mathrm{const}(2\tau_0\Delta)^{1/2}\exp[-\mathfrak{F}(\Delta)/T_c], \tag{13}$$

with

$$\mathfrak{F}(\Delta) = -2N_0\Omega\int_0^\Delta d\Delta'[(T_c - T)/T_c + 0.106\Delta'^2/T_c^2 + \chi^{(\mathrm{rad})}]\Delta'. \tag{14}$$

Obviously, the quantity $\mathfrak{F}$ represents a generalization of the Ginzburg-Landau free energy to nonequilibrium steady-state situations, and it agrees with the usual expression when $\chi^{(\mathrm{rad})} = 0$, i.e., in the absence of radiation.

Since $\mathfrak{F}$ is proportional to the volume Ω, the prefactor $(2\tau_0\Delta)^{1/2}$ is without importance, and that state is almost certainly realized which has the smallest value of $\mathfrak{F}$. We may call such a state one of global stability. In the example of Fig. 1, we have $\mathfrak{F}(\Delta_0=0)=0<\mathfrak{F}(\Delta_2)$, which means that the normal state is globally stable. At the same time, the superconducting state is locally, but not globally stable, and such a state is usually called a metastable state. For temperatures somewhat lower, the stability assignment for these two states is just reversed.

It is well known that the existence of metastable states does lead to a hysteretic behavior in the transition between two phases; in other words, the phenomena of supercooling and superheating are intimately connected with the appearance of metastable states. This is what has been observed by the authors of Ref. 3.

For the sake of completeness, I wish to add three remarks. Firstly, in their theory of a laser-irradiated superconductor, Owen and Scalapino[11] have assumed that the recombination rate of quasiparticles is so small that it can be neglected. Then a thermodynamic theory can be constructed where the quasiparticle number appears as an additional variable. In the present case, however, recombination processes occur at a large rate $\sim 1/\tau_0$ which exclude the application of such a theory. Secondly, electromagnetic radiation will stimulate superconductivity even above the thermodynamic transition temperature, since the troublesome generation of quasiparticles (which is possible if 2Δ is less than the energy of the radiation quanta) is suppressed by the coherence factor $1-\Delta^2/EE'$. In contrast, the coherence factor for acoustic radiation is $1+\Delta^2/EE'$, which favors quasiparticle creation. Thirdly, it is possible to generalize the considerations to cases where the order parameter also varies in space. Then the Ginzburg-Landau equation acquires the usual "kinetic energy" term, and the stochastic force is δ-correlated in both space and time.[12]

In conclusion, I wish to remark that radiation-stimulated superconductivity provides an additional example of phase transitions in a nonequilibrium steady-state system.

I acknowledge gratefully a stimulating discussion with Dr. T. M. Klapwijk and Dr. J. E. Mooij who brought my attention to this problem.

*Present address: Institut für Theorie der Kondensierten Materie, Universität Karlsruhe, D-7500 Karlsruhe, West Germany.

[1]G. M. Eliashberg, Pis'ma Zh. Eksp. Teor. Fiz. 11, 186 (1970) [JETP Lett. 11, 114 (1970)]; see also B. I. Ivlev, S. G. Lisitsyn, and G. M. Eliashberg, J. Low Temp. Phys. 10, 449 (1973).

[2]T. J. Tredwell and E. H. Jackson, Phys. Rev. B 13, 2931 (1976).

[3]T. M. Klapwijk, J. N. van den Berg, and J. E. Mooij, J. Low Temp. Phys. 26, 385 (1977).

[4]I wish to distinguish clearly between nonequilibrium steady state and nonequilibrium. For general information on problems of both kinds in superconductivity, I refer to D. N. Langenberg's article in *Low Temperature Physics—LT14*, edited by M. Krusius and M. Vuorio (North-Holland, Amsterdam, 1975), Vol. 5, p. 223.

[5]H. Haken, Rev. Mod. Phys. 47, 67 (1975).

[6]Various effects which are insignificant for the question of stability have been omitted. In principle they can be included by introducing an apparent electronic temperature $T^*>T$.

[7]This is the relaxation time of the longitudinal mode, which has been discussed in A. Schmid and G. Schön, J. Low Temp. Phys. 20, 207 (1975).

[8]Haken (Ref. 5) calls this "the enslavement of the many degrees of freedom by the order parameter."

[9]Equation (9) is evidently correct for thermal equilibrium. In other situations it is correct when correlations are neglected. This, however, is in the spirit of the Boltzmann equation approach ("molecular chaos"). The interpretation of the Boltzmann equation in the sense of a probabilistic equation has been discussed, for instance, by M. Kac, in *The Boltzmann Equation*, edited by E. G. D. Cohen and W. Thirring (Springer, New York, 1973).

[10]R. L. Stratonovich, *Topics in the Theory of Random Noise* (Gordon and Breach, New York, 1963), Vol. 1.

[11]C. S. Owen and D. J. Scalapino, Phys. Rev. Lett. 28, 1559 (1972).

[12]I consider here only fluctuations in the magnitude of the order parameter. Phase fluctuations require a separate treatment.