New instability in superconductors under external dynamic pair breaking*

Jhy-Jiun Chang and D. J. Scalapino
Department of Physics, University of California, Santa Barbara, California 93106
(Received 13 May 1974)

For superconducting films exposed to uniform laser radiation, it is found that the uniform distribution of quasiparticles is unstable when the system is driven sufficiently far from equilibrium. For $T/T_c < 0.7$, this instability occurs at lower laser intensities than the first-order supernormal transition for the uniform system and can drive the system into an intermediate state (dissipative structure).

A few years ago, Testardi[1] found that the transition temperature of a superconducting thin film was suppressed by laser light. He gave arguments which showed that this was not simply a heating effect and suggested that it represented a dynamic modification of the electrons. Following this, Owen and Scalapino[2] proposed a simple model in which they assumed the quasiparticles attained thermal equilibrium with respect to the lattice, but remained out of chemical equilibrium with respect to the pairs. This leads to a new distribution for the quasiparticles governed by an effective chemical potential μ^* which is a function of the number of excess quasiparticles as well as the lattice temperature. The excess quasiparticles due to the "bottleneck" in the recombination process interfere with the coherent scattering of the electrons and depress the gap. For a system constrained to have a uniform quasiparticle density, this model leads to a first-order supernormal phase transition at $T< T_c$. In this article we investigate the stability of this model with respect to quasiparticle density fluctuations. We find that over the major part of the temperature region of interest ($T/T_c<0.7$), an instability with respect to quasiparticle density fluctuations occurs at laser intensities less than those necessary to produce the first-order supernormal transition.

Consider a sample with a uniform quasiparticle distribution of density N_Q. We imagine a long-range fluctuation in the density so that half of the sample has density $N_Q+\epsilon$, while the other half has density $N_Q-\epsilon$, where ϵ is an infinitesimal quantity. For a sufficiently large sample the surface energy can be neglected, therefore, if the uniform configuration is stable, the relation

$$F(N_Q,T)-\tfrac{1}{2}[F(N_Q+\epsilon,T)+F(N_Q-\epsilon,T)] = -\frac{\partial^2 F(N_Q,T)}{\partial N_Q^2}\epsilon^2 = -\frac{\partial\mu^*(N_Q,T)}{\partial N_Q}\epsilon^2<0 \qquad (1)$$

must be satisfied. Here the free energy $F(N_Q,T)$ for the nonequilibrium system has the same form as in the BCS theory[3] except for the replacement of the quasiparticle energy E by $E-\mu^*$ in the Fermi function and the introduction of a modified gap. Equations for the modified gap and the effective chemical potential μ^* are given in Ref. 2. Figure 1 shows a plot of μ^* as a function of the *excess* quasiparticle density n [in units of $4N(0)\Delta_0$] for various temperatures. We note that for sufficiently large n, $\partial\mu^*/\partial n$ becomes negative. Since $(\partial\mu^*/\partial N_Q)_T=(\partial\mu^*/\partial n)_T(\partial n/\partial N_Q)_T$ and $(\partial n/\partial N_Q)_T$ is positive, the system is unstable when $(\partial\mu^*/\partial n)_T$ becomes negative. The fact that μ^* decreases at large n is due to the rapid decrease of the energy gap. Whether this instability will affect the nature of the supernormal phase transition depends on whether it occurs at a level of laser intensity higher or lower than that for the supernormal transition at the same temperature.

Figure 2 represents a phase diagram with the excess quasiparticle density n plotted as the ordinate and the reduced temperature T/T_c plotted as the abscissa. The solid curve represents the locus of points for which

$$\left(\frac{\partial\mu^*}{\partial n}\right)_T=0\ . \qquad (2)$$

For $(n, T/T_c)$ parameters associated with points below the solid curve, the uniform system is stable. The solid curve marks the boundary above which the system is unstable with respect to long-wavelength small-amplitude density fluctuations. We will refer to this as a density instability. The long-dashed curve represents the points at which

$$F(N_Q,T)-F_n(T)=0\ , \qquad (3)$$

where $F_n(T)$ is the free energy of the system in the normal state. Since it is assumed in the model that the thermal relaxation time is short, F_n is not affected by the application of laser light. The long-dashed curve marks a boundary associated with the first-order supernormal phase transition. We will refer to this as the SN phase transition. For temperatures $T/T_c<0.7$, the density instability occurs at a value of n smaller than that of the SN phase transition so that in this region the system will not undergo a first-order SN transition. At high temperatures ($T/T_c > 0.7$), the system first becomes unstable with respect to the SN phase transition. However, in this region the quasiparticle recombination time is no longer much larger than the quasiparticle thermalization time, and the model may not be accurate in describing the system.

It is not totally surprising to have a density instability occur in the present system. Many nonlinear systems are known to exhibit instabilities in their uniform steady-state solutions when operated far from thermodynamic equilibrium.[4] Some of them, e.g., laser thresholds,[5] are unstable with respect to small fluctuations, while others, such as chemical autocatalytic systems which show dissipative structures,[6] are unstable with respect to large amplitude fluctuations. These instabilities resemble, respectively, second-order and first-order phase transitions. It is well known that a liquid-gas system goes through a first-order transition before it hits the supercooling (or superheating) curve. The supercooling curve is given by $-(\partial P/\partial V)_T = 0$, which resembles $(\partial \mu^*/\partial N_Q)_T = 0$ in our case. Therefore, an instability with respect to large amplitude fluctuations can happen before the instability with respect to small amplitude fluctuations occurs. In order to pursue this for the present problem, consider the free energy $F(n, T)$ which can be obtained by integrating μ^* with respect to N_Q [Eq. (1)]. The resulting free energy $F(n, T)$ is concave upward for $n < n_i$ and concave downward for $n > n_i$, where n_i satisfies Eq. (2). If n_i is smaller than n_{sn} which satisfies Eq. (3), there exists an n^* ($< n_i$) such that a mixed state consisting of separate regions with densities n_{sn} and n^* ($n_{sn} > n \geq n^*$) will have less free energy than a uniform system with the same total number of quasiparticles. The excess quasiparticle density n^* is set by N_Q^*, which has the property that the tangent

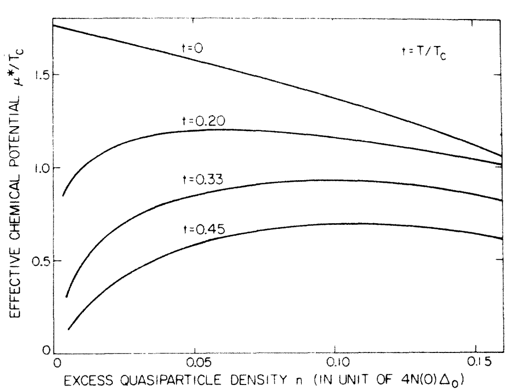

FIG. 1. Effective chemical potential μ^* as a function of the excess quasiparticle density n (measured in units of $4N(0)\Delta_0$) for temperatures $T/T_c = 0$, 0.2, 0.33, and 0.45.

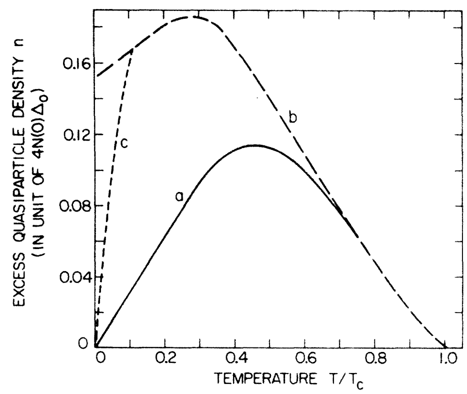

FIG. 2. "Phase diagram" for nonequilibrium superconductors. The solid curve marks the locus for density instabilities, the long-dashed curve represents where the system would undergo a first-order supernormal phase transition if there were no density instability, and the short-dashed curve shown is where the phonon attenuation coefficient $\alpha(\omega = 2\Delta)$ for the uniform system becomes negative.

of $F(N_Q, T)$ at N_Q^* passes through the point $F(n_{sn}, T)$. Therefore, the boundary above which the system is unstable with respect to large amplitude density fluctuations lies below the solid curve in Fig. 2. As n gets larger, since the quasiparticles are created uniformly, we expect the system to show a dissipative structure in which quasiparticles diffuse from the superconducting regions to the normal regions. At the same time electrons in the normal regions combine with the quasiparticles in the superconducting regions to form pairs and thus maintain local charge neutrality.

While we do not have a theory of this new intermediate state, it is possible to make some general comments. It appears that there are two basic types of intermediate states possible: one in which the system shows a stationary configuration with either superconducting regions embedded in a normal background or normal regions embedded in a superconducting background; the other in which these regions change in time[7,8] as well as in space. In order to reduce the cost of surface energy, the size of the embedded region should be governed by the coherence length, while the distance between the two-nearest embedded regions is of the order of the diffusion length to allow the quasiparticle to diffuse.

Recently the authors calculated[9] the ultrasonic attenuation coefficient $\alpha(\omega)$ for the nonequilibrium system assuming a uniform quasiparticle density. It was found that at sufficiently low temperatures and high quasiparticle concentrations the ultrasonic attenuation becomes negative for $\omega \sim 2\Delta$. This would mean that the system was unstable with respect to lattice vibrations at $\omega = 2\Delta$. The curve for $\alpha(\omega = 2\Delta, n, T) = 0$ is shown as the short-dashed curve in Fig. 2. However, since this curve lies in the region above the solid curve, the system will no longer be in the uniform state, and this lattice instability is unphysical.

To compare the theory with experimental results one has to notice that, because of the reduced specific heat and thermal conductivity at low temperatures, heating effects can be important[10,11] and hence must be determined and removed from the data before analysis. It is interesting to note that the tunneling measurements on a superconductor-insulator-superconductor junction by Parker and Williams[12] appear to stop near the point $(\partial\mu^*/\partial n)_T = 0$. Recent experiments such as the dc and the microwave resistance measurements by Langenberg *et al.*[8] and the tunneling measurements by Dynes *et al.*[11] indicate that the measured quantities change continuously to their values in the normal state as the laser intensities are increased. The evolvement of a dissipative structure triggered by the density instability may be responsible for these observed effects.

We thank Dr. Y. Imry for helpful discussions, and Dr. L. R. Testardi for private correspondence.

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*Research supported in part by the U. S. Army Research Office, Durham, North Carolina.

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