

that the point in contention may be somewhat academic in the sense that, as a relative contribution to a (moderate to) strong pyroelectric coefficient ($\Pi_{\text{prim}} \approx 10^{-2} \text{ C cm}^{-2} \text{ K}^{-1}$) such terms are probably insignificantly small.

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Onset of an Inhomogeneous State in a Nonequilibrium Superconducting Film

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We discuss a phenomenological theory of the onset of an inhomogeneous state in a nonequilibrium superconducting film. The length scale of the initial instability is given by the geometric mean of the coherence length and an effective quasiparticle diffusion length.

Recent experiments¹⁻⁵ suggest that a superconducting film, driven sufficiently far out of equilibrium, may go into some type of mixed state. Various forms for this have been proposed: a stationary inhomogeneous state consisting of separate superconducting and normal regions,⁶ or regions with different gaps^{4,7}; a temporally varying state which oscillates between the superconducting and normal phases.¹ Here we discuss a phenomenological model for a nonequilibrium film which exhibits an instability towards a stationary inhomogeneous state with spatial variations in the excess quasiparticle density and the order parameter.

The presence of quasiparticles in a superconducting film reduces the gap Δ and hence lowers the superconducting condensation energy. In the absence of a magnetic field, if the quasiparticle distribution is thermal, the film undergoes a second-order transition from the superconducting to normal phase as the quasiparticle density is increased. However, if the quasiparticle distribu-

tion is nonthermal, the system may become unstable with respect to a phase separation in which some regions go normal (or have a smaller gap) and drain off quasiparticles from other regions which can then become more strongly superconducting. Chang and Scalapino⁶ have shown that the μ^* model⁸ for a nonequilibrium superconductor exhibits this type of instability. In order to explore the nature of this instability and to determine its characteristic spatial scale, we consider a phenomenological model similar to one which has been used to describe striations in chemical reactions.⁹ Another closely related problem is that of the inhomogeneous state of ferroelectric photoconductors.¹⁰

Inhomogeneities can naturally arise if the thin films, their thermal contact to the substrate, or the drive are nonuniform so that different portions are driven normal at different drive intensities. Film inhomogeneities were discussed in connection with the laser-irradiation experiments of Ref. 2. Here we are interested in exploring

the possibility of an instability to an inhomogeneous state for a uniform film which is uniformly driven. To the extent that the energy distribution of the quasiparticles in the nonequilibrium state can be described by an effective temperature¹¹ T^* and an effective chemical potential μ^* ,⁸ it is the μ^* part which gives rise to an instability. For this reason we will make use of the μ^* model in guiding our construction of a phenomenological model.

To begin, we assume that the flow of quasiparticles can be described by a relation of the form

$$\mathbf{J} = -N(0)D\nabla\mu^*, \quad (1)$$

where D is the quasiparticle diffusion constant, $N(0)$ is the Bloch single-spin density of states at the Fermi surface, and μ^* is an "effective" chemical potential for the nonequilibrium quasiparticles. The dependence of μ^* on the quasiparticle density will naturally depend upon the detailed form of the quasiparticle energy distribution. Here we will consider the case in which μ^* varies with the excess quasiparticle density relative to thermal equilibrium in the presence of the reduced nonequilibrium gap N_{qp} as shown in Fig. 1. Initially μ^* increases as the excess quasiparticle concentration increases, but then beyond a critical quasiparticle concentration, N_c , the rapid decrease of the gap produced by the excess quasiparticles causes μ^* to decrease.

Based upon the numerical results of Ref. 6, we make an *Ansatz* that in the vicinity of N_c , μ^* varies as

$$\mu^*(N_{qp}) = \mu^*(N_c) - A\Delta_0(N_c - N_{qp})^2/[N(0)\Delta_0]^2. \quad (2)$$

Here the dimensionless parameter A is of order unity, $N(0)$ is the Bloch single-spin density of states at the Fermi surface, and Δ_0 is the zero-temperature gap. When the quasiparticle density

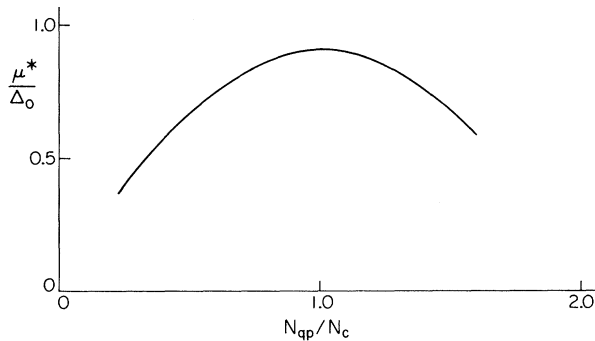


FIG. 1. Assumed form of the effective quasiparticle chemical potential μ^* vs N_{qp} for N_{qp} near N_c .

varies in space there will be an additional term in μ^* proportional to $-\nabla^2 N_{qp}$. Physically this reflects the fact that the order parameter Δ varies with N_{qp} so that $\delta F/\delta N_{qp}(x)$ contains a term

$$\frac{\delta}{\delta N_{qp}(x)} \int N(0)\xi^2 |\nabla\Delta|^2 d^3x = -B \frac{\xi^2}{2N(0)} \nabla^2 N_{qp}, \quad (3)$$

where B is a constant of order unity, and ξ is the zero-temperature coherence length.

Although the values of the parameters in μ^* as well as D depend upon the exact nature of the nonequilibrium state, we believe meaningful estimates¹² can be obtained by taking $A=B=1$, $\xi = 0.085(\xi_0 d)^{1/2}$, and $D = v_F d/3$. Here ξ_0 is the usual BCS coherence length $\hbar v_F/\pi\Delta_0$ and d is the film thickness (or impurity mean free path).

Using the sum of Eqs. (2) and (3) for $\mu^*(N_{qp})$ with $A=B=1$, the expression for the quasiparticle current, Eq. (1), becomes

$$\vec{J} = \frac{-2D}{N(0)\Delta_0} (N_c - N_{qp})\nabla N_{qp} + \frac{D\xi^2}{2} \nabla(\nabla^2 N_{qp}). \quad (4)$$

For N_{qp} less than N_c , the first term gives a quasiparticle current which flows from regions of high to low quasiparticle concentration. However, when N_{qp} exceeds N_c , the first term changes sign and produces an unstable flow in which quasiparticles move from regions of low concentration to regions of high concentration. This flow, being proportional to ∇N_{qp} favors rapid variations in N_{qp} and is the driving term producing the inhomogeneous state. The second term in Eq. (4) acts to oppose rapid spatial changes and stabilizes the spatial variations in N_{qp} .

Adding the divergence of \vec{J} to the Rothwarf-Taylor equation¹³ for the time rate of change of the quasiparticle density, we obtain the phenomenological equation

$$\frac{\partial}{\partial t} N_{qp} = I_{qp} - 2RN_{qp}^2 + 2\beta N_{ph} - \nabla \cdot \vec{J}. \quad (5)$$

Here I_{qp} represents a quasiparticle source, $(2RN_{qp})^{-1}$ is the quasiparticle recombination lifetime τ_R , and β^{-1} is the pairbreaking lifetime τ_B of the phonons. N_{ph} is the nonequilibrium density of phonons (with energies greater than 2Δ). Phonon diffusion is not important so that

$$\frac{\partial N_{ph}}{\partial t} = I_{ph} + RN_{qp}^2 - \beta N_{ph} - \frac{N_{ph} - N_{ph}^e}{\tau_{es}}, \quad (6)$$

with I_{ph} the phonon source, τ_{es} the phonon escape time to the thermal bath, and N_{ph}^e the bath equilibrium phonon density.

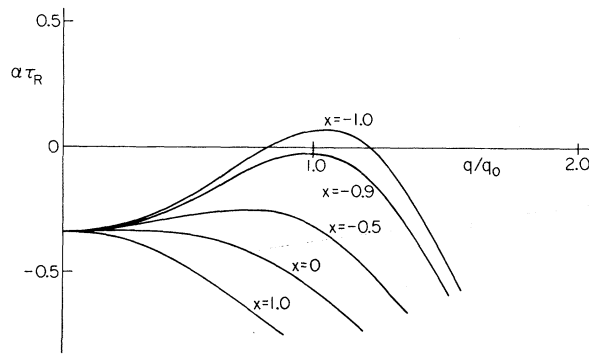


FIG. 2. The instability rate constant $\alpha(q)$ in units of τ_R^{-1} vs q/q_0 with $q_0^2 = (2/\tau_R D \xi^2)^{1/2}$. Here $x = (N_c - N_{qp}^0) \times (2\tau_R D)^{1/2} / N(0)\Delta_0 \xi$ and we have taken $\tau_R = \tau_B = 0.5\tau_{es}$.

Neglecting the dependence¹⁴ of the drive terms I_{qp} and I_{ph} on N_{qp} and N_{ph} , it is straightforward to carry out a linear stability analysis¹⁵ in which

$$\begin{aligned} N_{qp} &= N_{qp}^0 + \delta N_{qp} e^{\alpha(q)t} e^{iqx}, \\ N_{ph} &= N_{ph}^0 + \delta N_{ph} e^{\alpha(q)t} e^{iqx}. \end{aligned} \quad (7)$$

The resulting secular equation for $\alpha(q)$ is

$$\begin{aligned} [\alpha(q) + 2\tau_R^{-1} + D(q)][\alpha(q) + \tau_B^{-1} + \tau_{es}^{-1}] \\ - 2\tau_R^{-1}\tau_B^{-1} = 0, \end{aligned} \quad (8)$$

with

$$D(q) = Dq^2 [2(N_c - N_{qp}^0) / N(0)\Delta_0 + \frac{1}{2}\xi^2 q^2]. \quad (9)$$

Figure 2 shows the solution $\alpha(q)$ of Eq. (8) versus q for various values of $x = (N_c - N_{qp}^0)(2\tau_R D)^{1/2} / N(0)\Delta_0 \xi$. Here $\alpha(q)$ is in units of τ_R^{-1} and q is in units of $q_0 = (\tau_R D \xi^2 / 2)^{-1/4}$. In the figure we have taken $\tau_R = \tau_B = 0.5\tau_{es}$. When N_{qp}^0 is smaller than N_c , corresponding to positive values of x , modes with finite wave vectors are more rapidly damped than the $q=0$ mode. However, when N_{qp}^0 exceeds N_c , corresponding to negative values of x , a maximum of $\alpha(q)$ occurs for a wave vector $q_m = q_0 \times (-x)^{1/2}$. For a particular case shown in Fig. 2, $\alpha(q_m)$ is zero for x slightly less than -0.9 . For an N_{qp}^0 concentration larger than this, a spatial variation with wave vector q_m will develop in N_{qp} and hence also in the gap. The scale of the initial instability is set by $2\pi/q_m$. Note that the roots $\alpha(q)$ are real so that this theory does not lead to a temporally varying inhomogeneity.

It is straightforward to calculate q_m . The maximum in $\alpha(q)$ occurs when $\partial D / \partial q^2$ vanishes and corresponds to $q_m = q_0(-x)^{1/2}$, as noted previously. Then, setting $\alpha(q_m)$ equal to zero in Eq. (8), one

finds that

$$D(q_m)(\tau_B^{-1} + \tau_{es}^{-1}) + 2/\tau_R \tau_{es} = 0, \quad (10)$$

with $D(q_m) = -D\xi^2 q_m^4 / 2$. Solving Eq. (10) for q_m gives, for the characteristic length scale of the instability at threshold,

$$2\pi/q_m = \sqrt{2}\pi [D\tau_R(1 + \tau_{es}/\tau_B)\xi^2]^{1/4}. \quad (11)$$

This length is set by the geometric mean of an effective quasiparticle diffusion length $[D\tau_R(1 + \tau_{es}/\tau_B)]^{1/2}$ and the coherence length ξ . The time scale with which the instability grows is set by $\alpha(q_m)$. Clearly, once the instability onsets it is necessary to include the nonlinear terms in order to determine the ultimate structure of the inhomogeneous state. The nonlinear effects produce a coarsening of the structure as well as a sharper change between the regions with different gap values. Without a more detailed analysis it is not known whether the system will evolve into a mixed state of normal and superconducting regions or possibly regions with two distinct gap values.^{4,7}

For a film whose width is small compared to q_m^{-1} the instability will appear as a periodic variation along the length of the film. We believe that it should be possible to look for this instability in the following way: Imagine a narrow, thin film which forms one side of a tunnel junction. Recent experiments³⁻⁵ indicate that when quasiparticles are injected close to the gap edge, the film will, at some critical injection current I_c , exhibit an instability. For a Sn film, 10^3 \AA thick, we estimate from Eq. (11) that the characteristic wavelength of the initial instability is of order $10 \mu\text{m}$. If a weak spatially varying source, such as a light pattern, were projected on the film, the value of I_c should depend upon the relationship between the length scale L of this source and the characteristic wavelength of the instability. The effect of the weak source should be largest when $L \cong 2\pi/q_m$, so that by changing L one could determine the length scale of the instability. Note that the basic drive must be homogeneous, and it is the variation of the instability threshold in the presence of a weak spatial perturbation which will determine $2\pi/q_m$.

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¹²The length scale of the instability, Eq. (11), actually varies as $A^{1/2}/B^{1/4}$. Thus, to the extent that A and B differ from unity, our result for this length will change. A microscopic theory is needed to determine A and B , but we believe that for $0.3 \leq T/T_c \leq 0.9$ our estimate of unity sets the correct order of magnitude.

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$$[\alpha + 2\tau_R^{-1} + D(q) - \partial I_{qp}/\partial N_{qp}](\alpha + \tau_B^{-1} + \tau_{es}^{-1} - \partial I_{ph}/\partial N_{ph}) - (2\tau_R^{-1} + \partial I_{qp}/\partial N_{qp})(\tau_R^{-1} - \partial I_{ph}/\partial N_{qp}) = 0.$$

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Forced Rayleigh Scattering from Lipid-Water Smectic Phases

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The hydrodynamic relaxation rate for water inhomogeneities in aligned lipid-water smectics is measured for the first time using an improved version of the "forced Rayleigh" technique. Variation of the relaxation rate with water concentration yields a distinction between "bound" and "free" water.

Although part of the motivation for research into the physics of liquid crystals derives from the fact that many biological systems exhibit liquid crystalline morphology, physicists have by and large concerned themselves with studies of liquid crystalline systems that have little or no direct connection with biological systems.^{1,2} On the other hand, it is well known that lipid-water mixtures form smectic liquid crystal phases that have the same symmetries as liquid crystals that have been extensively studied.^{3,4} The lipid molecule possesses a hydrophilic polar head group that is attached to one or more long hydrophobic hydrocarbon chains.⁵ In the presence of water these amphiphilic lipid molecules can form planar bilayers in which the polar head groups locate on the surface so as to shield the hydrophobic chains from the water. Lipids are principal ingredients in biological membranes and lipid bilayers have been extensively studied as model membrane systems. Furthermore, studies on living systems have presented convincing evidence that many of the functions of living membranes are altered at temperatures that correspond to phase transitions

in the smectic lipid-water (SLW) systems.⁵ Interactions between lipid and water are essential to the structure and stability of membranes.⁶ We report here new measurements on the fluidity of water in the SLW phase of dipalmitoyl phosphatidylcholine (DPPC).

SLW systems consist of a stack of planar lipid bilayers separated from each other by a layer of water.⁷ Techniques have recently been developed to produce macroscopic samples ~ 1 cm² in area and ~ 0.1 μ m thick.^{8,9} The size and quality of these samples are comparable to other liquid crystals and crystals that have been studied by physical techniques. These systems also have interesting physical properties without regard to their biological relevance. For example, a unified hydrodynamic theory of crystals, liquid crystals, and liquids has been developed and experiments have been done to observe some of the modes that have been predicted for single-component, nonbiological liquid crystals.^{1,2,10} de Gennes and Brochard have applied these general ideas to the specific case of the binary lipid-water smectic and made predictions concerning an overdamped