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Peierls state far from equilibrium

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We show that a Peierls insulator under strong dynamic excitation of electron-hole pairs exhibits an instability into a mixed metal-insulator phase. This problem is analogous to the inhomogeneous nonequilibrium BCS state discussed by Scalapino and Huberman. We calculate the threshold parameters for typical Peierls insulators and discuss some organic systems that might be likely candidates for experimental verification. In contrast to the nonequilibrium superconductor, the mixed state in Peierls systems can be easily detected by light scattering techniques.

I. INTRODUCTION

Condensed-matter systems far from equilibrium are of interest because they can undergo transitions into states which are not usually attainable in their equilibrium state. As the examples of Rayleigh-Benard instabilities in fluids, spinodal decomposition in alloys, and nonequilibrium superconductors show, new qualitative features can appear as a system is driven far from equilibrium. Thus, it is possible to obtain dissipative structures characterized by a high degree of regularity in which the properties of the system are modulated spatially and/or temporally.

Recently, considerable attention has been devoted to the properties of BCS superconductors far from equilibrium.[1] The experimental impetus was provided by Testardi's observation[2] of a nonthermal suppression of T_c by laser irradiation in thin superconducting films, which he attributed to a dynamic modification of the electronic density of states by the incoming photons. Soon afterwards, Owen and Scalapino[3] proposed a simple model in which the photogenerated quasiparticles are in equilibrium with respect to the lattice but not with respect to the Cooper pairs. This possibility leads to a new "quasiequilibrium" distribution for the quasiparticles characterized by an effective chemical potential μ^*. For a system constrained to have a uniform quasiparticle density such a model leads to a suppression of T_c and a first-order supernormal phase transition into the normal state. Furthermore, the possibility exists that at lower laser intensities than those needed to produce the supernormal phase transition the superconducting film might be driven into some kind of mixed state. Two forms for this mixed state have been proposed: a stationary inhomogeneous state consisting of separate superconducting and normal regions, and a temporary varying one which oscillates between the superconducting and normal phases. Most recently, Scalapino and Huberman[4] have discussed a phenomenological model for a nonequilibrium superconducting film which exhibits an instability towards a stationary inhomogeneous state with periodic spatial variations in the excess quasiparticle density and the order parameter.

The model of Owen and Scalapino makes use of the BCS theory with its characteristic pairing of electron states into bound Cooper pairs. A key quantity in the model is thus the gap equation, which arises out of this pairing. It is well known that pairing is not a unique feature to superconductivity. Systems as varied as, for example, spin-density waves,[5] ferroelectric semiconductors,[6] charge-density waves,[7] and Peierls states[8] can be described in a form similar to that of the BCS superconductor. With this analogy in mind it is tempting to speculate about the nonequilibrium properties of these other systems along the lines we have discussed above.

We have chosen to investigate the fate of a Peierls insulator driven far from equilibrium by dynamic photoproduction of electron-hole pairs. As we will show, it is possible for such a system to undergo a transition into a mixed metal-insulator structure characterized by the presence of coexisting regions of both phases. In this paper the general conditions for attaining such an instability are discussed and some possible experimental candidates are suggested. The choice of a Peierls insulator was dictated by several reasons: (i) current interest in these solids has led to a thorough understanding of the basic mechanisms

that are operational in many of the organic conductors known to exhibit a Peierls insulating phase; (ii) there exist systems where a mean-field description seems to be appropriate in spite of their quasi-one-dimensional character; and (iii) we believe that the existence of a mixed metal-insulator state in such solids can be detected quite simply by light scattering techniques.

Throughout this paper we will restrict ourselves to a simple mean-field treatment of the gap equations. Although it is known that the effect of fluctuations is important for quasi-one-dimensional systems, we believe that a mean-field model may be sufficient to speculate on some of their nonequilibrium properties. In addition, the nonequilibrium Peierls state might be a potentially useful system for probing such fluctuations.

In Sec. II the homogeneous case of a quasiequilibrium Peierls insulator is described and the phase diagram as a function of excitation number is obtained. Section III deals with the onset of spatial inhomogeneities and the characteristic length scales to be expected. Possible experimental tests of these ideas are discussed and some likely candidates proposed.

II. HOMOGENEOUS PEIERLS INSULATOR

In this section we study a homogeneous Peierls insulator within the mean-field approximation ignoring the spatial variations of the order parameter. For this purpose we make use of a Frölich electron-phonon (e-p) Hamiltonian for a linear chain,[9]

$$H=\sum_k \epsilon_k a_k^\dagger a_k+\sum_q \hbar\omega_q b_q^\dagger b_q + \frac{1}{\sqrt{N}}\sum_k\sum_q g(q)a_{k+q}^\dagger(b_q+b_{-q}^\dagger) \tag{2.1}$$

in which $a_k^\dagger$ and $b_q^\dagger$ denote annihilation operators for the electron Bloch states, of energies ϵ_k, and unperturbed longitudinal phonons, of frequencies ω_q, respectively. N is the number of ions in the chain of lattice constant a, and $g(q)$ is the e-p coupling constant. For the unperturbed one-electron dispersion relation we choose for simplicity the tight-binding expression $\epsilon_k=-\epsilon_B\cos(ak)$ with $|k|\leq\pi/a$. For the summations over crystal momenta in Eq. (2.1) electron spin is to be included.

As discussed by Peierls,[10] Frölich,[11] and Kuper,[12] for $T<T_c$ the wave numbers $\pm q_0=2k_F$ which connect the two points of the Fermi surface become macroscopically occupied. For the special case of a half-filled band, a case that we will restrict ourselves to in the following, the lattice undergoes a distortion in which the period of the lattice doubles. As shown by Rice and Strassler[13] the Hamiltonian of Eq. (2.1) may then be approximated by the simplified, static form

$$H=\left(\frac{N\hbar\omega u^2}{2}\right)+\sum_k[\epsilon_k a_k^\dagger a_k+ug(a_{k+q_0}^\dagger a_k+a_{k-q_0}^\dagger a_k)] \tag{2.2}$$

where $\omega=\omega_{q_0}$ and u is a dimensionless parameter related to the phonon amplitude through $\langle b_q\rangle=\langle b_q^\dagger\rangle=\frac{1}{2}\sqrt{N}\,\delta_{q,q_0}$. This approximation amounts to including only the electron entropy in the free energy. It is convenient to define an "inner" Brillouin zone (in-BZ) by $|k|\leq k_F$ with $k_F=\pi/2a$. The Hamiltonian

$$H=\sum_{k\in(\text{in-BZ})}[\epsilon_k a_k^\dagger a_k+\epsilon_{k+q_0}a_{k+q_0}^\dagger a_{k+q_0}+2gu(a_{k+q_0}^\dagger a_k + a_k^\dagger a_{k+q_0})]+\frac{N\hbar\omega u^2}{2} \tag{2.3}$$

is then easily diagonalized by means of the canonical transformation

$$\begin{aligned}\alpha_k&=v_k a_k+u_k a_{k+q_0}\ ,\\ \beta_k&=-u_k a_k+V_k a_{k+q_0}\ ,\end{aligned} \tag{2.4}$$

where $u_k^2+v_k^2=1$. One thus obtains

$$H=\sum_{k\in(\text{in-BZ})}(E_k^\alpha\alpha_k^\dagger\alpha_k\,E_k^\beta\beta_k^\dagger\beta_k)+\frac{N\hbar\omega u^2}{2}\ . \tag{2.5}$$

For a symmetric band, $\epsilon_k=-\epsilon_{k+q_0}$, the quasiparticle energies for the two bands, α and β, are

$$E_k^{\alpha,\beta}=\pm(\epsilon_k^2+\Delta^2)^{1/2}\ , \tag{2.6}$$

where $\Delta=ug$. The gap Δ is determined by a minimization of the electronic free energy, which gives rise to the gap equation[13]

$$\sum_{k\in(\text{in-BZ})}\frac{[n_\alpha(k)-n_\beta(k)]}{(\epsilon_k^2+\Delta^2)^{1/2}}=\frac{N\hbar\omega}{g^2}\ . \tag{2.7}$$

In the case of thermal equilibrium the occupation numbers are $n_{\alpha,\beta}=1/[\exp(\beta E_k^{\alpha,\beta})+1]$ where $\beta=1/k_BT$ is the inverse temperature. Because the band is symmetric around $E=0$ the equilibrium chemical potential μ equals zero at all temperatures. In the weak coupling limit it follows[13] from Eq. (7) that $\Delta_0\simeq 4\epsilon_B e^{-2b}$ at $T=0$, where $b=\hbar\omega/2g^2N(0)\gtrsim 1$. [$N(0)$ is the density of states per atom including spin.] Above the critical temperature $k_BT_c\simeq 2.27\epsilon_B e^{-2b}$ there is only one trivial solution $\Delta=0$. One thus has the following well known picture of the Peierls distortion. Above T_c the regular lattice structure of the one-dimensional conductor, and hence its metallic state, is stable. At T_c a lattice distortion occurs giving way to an insulating state. With further decrease of T the gap at the Fermi surface increases in a BCS fashion. With these preliminaries we may now consider the quasiequilibrium state.

As described in Sec. I we shall assume that the system is prepared in such a way that there is an excess number of particles in the upper band. This excess number of electrons can be in principle produced by laser pumping across the gap Δ. We shall also assume that the time scale is such that a quasiequilibrium situation will have time to develop, i.e., the electrons in the upper band thermalize down to the bottom of the band in times that are short when compared to recombination times. In the formalism above, we shall therefore have to add the subsidiary condition that the number of excess particles remains fixed.[3,14] This will result in an additional chemical potential μ^* such that the occupation numbers in Eq. (7) now become

$$n_{\alpha,\beta}(k,\mu^*) = 1/\{\exp[\beta(E_k^{\alpha,\beta} \pm \mu^*)] + 1\} \ .$$

In order to show the behavior of the Peierls gap as a function of excess electron-hole pairs we have solved the gap equation with μ^* included by numerical methods for a set of temperatures less than T_c. Selected results are displayed in Fig. 1, where the number of excess particles in the upper band of the insulating state is defined as

$$\Delta n = [N_\beta(\mu^*,T) - N_\beta(o,T)]/N \tag{2.8}$$

with

$$N_\beta(\mu^*,T) = \sum_{k\epsilon(\text{in-BZ})} n_\beta(k,\mu^*) \tag{2.9}$$

As in the case of the quasiequilibrium BCS state[3,14] we note that the gap is double valued. Comparing free energies it turns out, however, that the "small" gap solution is unfavorable (as it is in the BCS case). Hence, we may disregard it in the following. Figure 2 shows the chemical potential μ^* as a function of

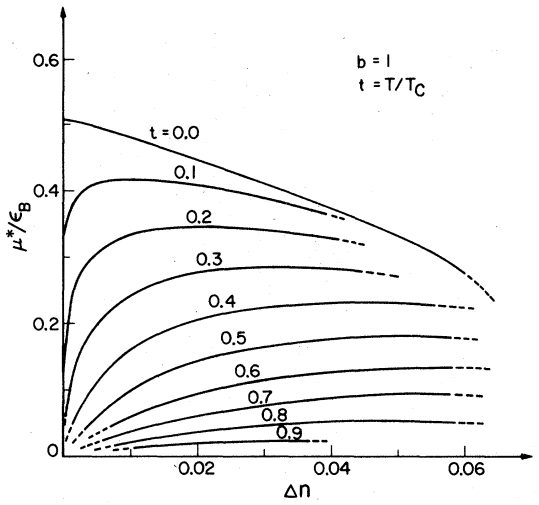

FIG. 2. Effective chemical potential μ^* as a function of number of excess carriers at different temperatures.

Δn, results which should also be compared with the quasiequilibrium BCS state. Since conditions for thermodynamic stability require that $(\partial\mu^*/\partial\Delta n)_T > 0$,[15] there is a critical value of Δn beyond which the system becomes unstable. This instability is used to define the approximate phase boundary between regions (a) and (b) in the "phase diagram" of Fig. 3. (A more accurate determination of the phase boundary requires a tangential construction as discussed in Ref. 7.) The phase diagram is completed by considering the difference in electronic free energies between the condensed and normal, metallic phases, $\Delta F = F_c - F_N > 0$ as shown in Fig. 4. Since it is assumed that the thermal relaxation time is

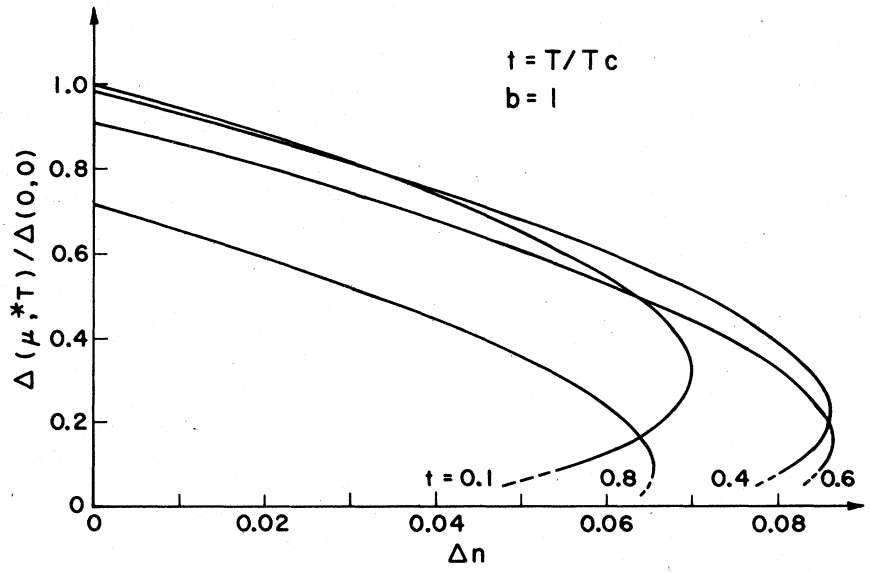

FIG. 1. Gap $\Delta(\mu^*,T)$ as a function of number of excess carriers, Δn, at different temperatures $t = T/T_c$ and with $b = 1$.

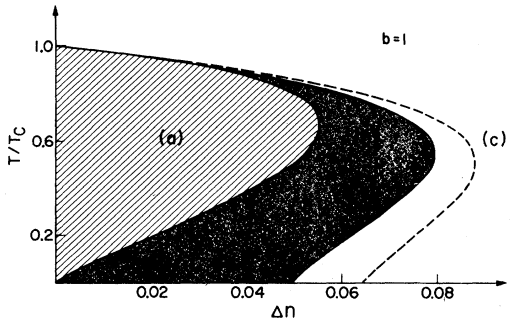

FIG. 3. "Phase diagram" for a nonequilibrium Peierls system: (a) denotes a homogeneous region in which $\Delta(\mu^*,T)\neq 0$ and $(\partial\mu^*/\partial\Delta n)_T=0$; (b) defines a region of quasiparticle density instabilities with $(\partial\mu^*/\partial\Delta n)_T<0$; and (c) is a region in which the normal, metallic state is stable, i.e., $\Delta F\geq 0$. At the dashed curve the gap equation ceases to have nontrivial solutions.

short, F_N is not affected by the laser field. In conclusion, we obtain for the quasiequilibrium Peierls insulator a behavior, which is quite similar to its BCS analog. In region (a) of Fig. 3 the system remains homogeneous and insulating. As Δn increases at constant T, however, region (b) of Fig. 3 is reached which implies that the system becomes unstable against large fluctuations in Δn. The system will then gain free energy by forming some kind of mixed state consisting of separate regions with different quasiparticle concentrations. It is, however, hard to predict the detailed nature of this mixed phase, but in the simplest case, it will consist of a mixture of insulating and normal regions, which may change in space as well as in time, as mentioned in Sec. I. With further increase of Δn at constant T the normal phase is reached [region (c) in Fig. 3].

III. MIXED STATE

As we showed in Sec. II (Fig. 3), for $T<T_c$ there exists a critical concentration of excited electrons, Δn_c, for which the Peierls insulator becomes thermodynamically unstable. In other words, as the excess electron density increases, the system exhibits a bifurcation into a mixed state characterized by the coexistence of small regions with different gaps. Since so far in our treatment we have only looked at homogeneous fluctuations, it is impossible within that context to study the spatial characteristics of the bifurcating state. In what follows, we will consider the contribution to the free energy and rate equations of a nonlocal chemical potential. As we will show, it then becomes possible to study the onset of a spatially periodic structure characterized by the coexistence of droplets of different gaps. This situation, under stong pumping conditions (or far enough from equilibrium) will ultimately lead to a mixed metallic-insulating phase in a nonequilibrium Peierls insulator.

Consider the rate equations for electron-hole photoproduction. If I is the rate at which electrons are pumped across the gap and τ their recombination time we can write

$$\frac{\partial n}{\partial t}=I-\frac{n}{\tau}-\vec{\nabla}\cdot\vec{J} \tag{3.1}$$

with J the electron current, which in terms of the effective chemical is given by

$$\vec{J}=-N(0)D\vec{\nabla}\mu^* \ , \tag{3.2}$$

with D the diffusion coefficient of the electrons. In order to obtain an expression for the dependence of μ^* on the excess quasiequilibrium electron density we expand the chemical potential around Δn_c, as sug-

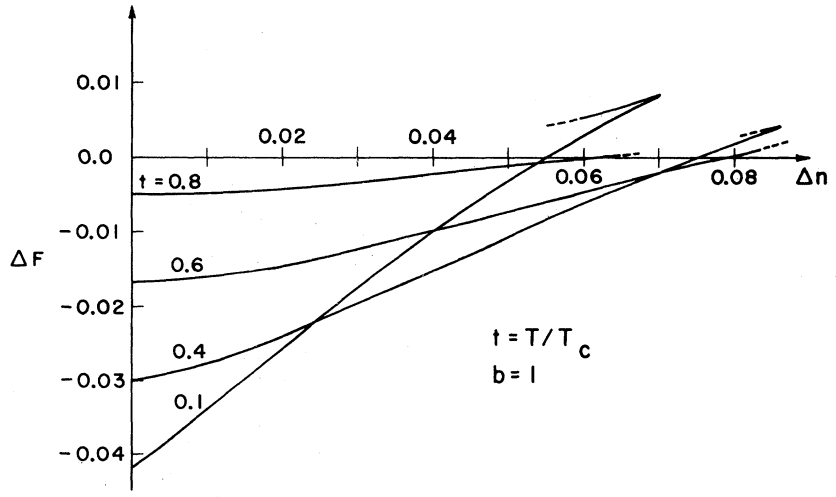

FIG. 4. Difference in electronic free energies, $\Delta F=F_C-F_N$, between the condensed and the normal, metallic phases (in units of E_B).

gested by Fig. 2. We then obtain

$$\mu^*(\Delta n) = \mu^*(\Delta n_c) - A(\Delta n_c - \Delta n)^2/N(0)\Delta_0 \ , \quad (3.3)$$

with A a constant of order unity.

The nonlocal contribution to $\mu^*(\Delta n)$ arises from considering the contribution from spatial fluctuations in the gap order parameter to the free energy. If the latter possesses a Ginzburg-Landau form the correlation energy can be written $\int N(0)\xi_0^2|\nabla\Delta|^2 d\vec{r}$ with ξ_0 the zero-temperature coherence length, which is given by[13]

$$\xi_0^2 = 7\zeta(3)\hbar^2 V_F^2/16\pi^2 K_B^2 T_c^2 \ , \quad (3.4)$$

with V_F the Fermi velocity of the electrons. Assuming that the gap is renormalized by the excess electrons as $\Delta = \Delta_0(1 - \frac{1}{2}\Delta n)$ and since $\mu^*(\vec{r}) = \delta Fn/\delta n(r)$ we obtain, together with Eq. (3.3)

$$\mu^*(\Delta n, \vec{r}) = \mu^*(\Delta n_c) - \frac{A(\Delta n_c - \Delta n)^2}{N(0)\Delta_0} - \frac{\xi_0^2}{2N(0)}\nabla^2(\Delta n) \quad (3.5)$$

so that Eq. (3.1) becomes

$$\frac{\partial \Delta n}{\partial t} = I - \frac{\Delta n}{\tau} + \frac{2DA}{\Delta_0}(\Delta n_c - \Delta n)\nabla^2(\Delta n) - \tfrac{1}{2}D\xi_0^2\nabla^4(\Delta n) \quad (3.6)$$

For $\Delta n_c > \Delta n$ and when properly linearized, Eq. (3.6) reduces to a well-known diffusion equation with sources, in which a gradient in the quasiparticle concentration generates a corresponding current that flows from high- to low-concentration regions. However, for $\Delta n_c < \Delta n$ an instability can take place in which an effective reversal of the diffusion coefficient leads to the growth of certain Fourier components of the fluctuations which, at later times, tend to be stabilized by the last term of Eq. (3.6).

In order to study the bifurcations of Eq. (3.6) into a mixed state, it is most convenient to perform a linear-stability analysis around the homogeneous steady-state solution, which we will call Δn_0. We write $\Delta n(r,t)$ in the form

$$\Delta n(\vec{r},t) = \Delta n_0 + \Delta n_1(\vec{r},t) \ , \quad (3.7)$$

with $\Delta n_1(\vec{r},t) \ll \Delta n_0$ for small t.

Replacing Eq. (3.7) into Eq. (3.6) and keeping only the linear terms in $\Delta n_1(\vec{r},t)$ we obtain

$$\frac{\partial \Delta n_1(\vec{r},t)}{\partial t} = I - \frac{(\Delta n_0 + \Delta n_1)}{\tau} + \frac{2DA}{\Delta_0}(\Delta n_c - \Delta n_0) \times \nabla^2(\Delta n_1) - \tfrac{1}{2}D\xi_0\nabla^4(\Delta n_1) \ . \quad (3.8)$$

Fourier analyzing, we obtain the following solution for Eq. (3.8)

$$\Delta n_1(r,t) = B\sum_q e^{\alpha(q)t}\cos q\, r \ , \quad (3.9)$$

with q the wave vector, B a constant, and $\alpha(q)$ the amplification factor, which is given by

$$\alpha(q) = -\left[\frac{1}{\tau} + \frac{2DA}{\Delta_0 N(0)}(\Delta n_c - \Delta n_0)q^2 + \frac{D\xi_0^2 q^4}{2}\right] \ . \quad (3.10)$$

For $\Delta n_0 < \Delta n_c$, $\alpha(q)$ is negative for all values of q and fluctuations in Δn will decay exponentially in time. However, if at sufficiently high excitation levels the condition $\Delta n_0 > \Delta n_c$ is fulfilled, the amplification factor can become positive for a range of wave vectors. This nonequilibrium condition will then lead to the growth of an inhomogeneous but regular structure characterized by a length λ_m. From the point of view of Eqs. (3.7) and (3.9) as $\alpha(q)$ crosses the origin the system exhibits a bifurcation into a new state characterized by a nonzero value of $\Delta n(\lambda_m)$.[16]

For small values of t, where linear-stability theory is applicable, it is possible to determine the threshold conditions and the length scale of the mixed state. Since $\alpha(q)$ appears in an exponential the particular wave-vector value for which α is a maximum will effectively set the length scale. From Eq. (3.10) and for $\Delta n_0 > \Delta n_c$ the condition $\partial\alpha(q)/\partial q = 0$ gives

$$q_m^2 = 2A(\Delta n_0 - \Delta n_c)/N(0)\Delta_0\xi_0^2 \ . \quad (3.11)$$

On the other hand, since the bifurcation occurs at $\alpha(q_m) = 0$, we can determine the threshold value for the excess electron density as

$$(\Delta n_0 - \Delta n_c)_{\text{th}} = N(0)\Delta n_0/\xi_0 A(2D\tau)^{1/2} \ . \quad (3.12)$$

Replacing this expression in Eq. (3.11) we obtain for the characteristic length λ_m the following expression (with $A \simeq 1$)

$$\lambda_m = \sqrt{2}\pi(2\tau D\xi_0^2)^{1/4} \quad (3.13)$$

which coincides with that of a nonequilibrium superconductor when in the latter the pair breaking lifetime of the phonons is the same as their escape time.[4]

For an organic conductor where band theory is applicable $D = V_F\Lambda$ with Λ the electron mean free path. Using Eq. (3.4) we can then write

$$\lambda_m = \sqrt{2}\pi[14\tau V_F^3\Lambda\hbar^2\zeta(3)/16\pi^2(k_B T_c)^2]^{1/4} \ . \quad (3.14)$$

If $\epsilon_F \simeq 0.5$ eV, $\Lambda \simeq 10$ Å, and $T_c \simeq 100$ °K an electron-hole recombination time of 10^{-11}–10^{-12} sec will give $\lambda_m \simeq 1000$ Å for the characteristic length of the mixed state.

The existence of the mixed gap state can be detected by light scattering techniques. For a system of small droplets with dielectric constant $\epsilon_m(\omega) = \epsilon_m^R(\omega) + i\epsilon_m^I(\omega)$ dispersed in a medium with dielectric constant $\epsilon_p(\omega)$, the absorptive cross section

for electromagnetic waves is given by Mie theory as[17]

$$\sigma = \frac{12\pi\omega r^3}{c} \frac{\epsilon_m^I(\omega)/\epsilon_p^I(\omega)}{[\epsilon_m^R(\omega)/\epsilon_p^R(\omega)+2]^2 + [\epsilon_m^I(\omega)/\epsilon_p^I(\omega)]^2} , \tag{3.15}$$

with r the radius of the droplets, which in our case is of order λ_m, and with the constraint $r > c/\sqrt{\epsilon_m}c$, which can be easily satisfied.

Equation (3.2) shows that as the ratio of the real dielectric constants in the two phases approaches the value -2, a strong absorption peak appears, from which in principle it is possible to obtain a value of λ_m. The considerable amount of uncertainty that surrounds the values of many of the parameters that enter Eqs. (3.2) and (3.3) does not allow us to make more concrete quantitative predictions at this stage. We do have several systems in mind however, that might qualify for a test of our predictions. As reported by Etemad, *et al.*,[18] the organic compound (tetraselenafulvalenium-tetracyanoquinodimethanide) (TSeF-TCNQ) exhibits a semiconducting gap which seems similar to that predicted by the mean-field treatment of the Peierls transition and with a $T_c = 29\,^\circ\mathrm{K}$, which makes it suitable for optical pumping experiments. Also, it has recently been shown by Abrahams, Gorkov, and Kharadze[19] that the series of organic compounds *bis*(tetrathiatetracene)-triiodide $[(TTT)_2I_3]$ can be made to exhibit a cross over from one-dimensional behavior to three-dimensional behavior by varying the iodine concentration. These facts, coupled to the behavior of the Peierls gap with temperature make the possible detection of the inhomogeneous states quite feasible.

IV. CONCLUSIONS

As we have shown, a Peierls insulator driven far from equilibrium by copious photoproduction of electron-hole pairs can undergo a transition into a new mixed state characterized by the coexistence of a lattice of quasimetallic droplets and insulating ones. The parameter values we have used indicate a typical length of several thousand angstroms for the new structure, a range that can easily be probed with light scattering techniques or other means.

Our analysis is based on a bifurcation analysis of the inhomogeneous rate equations that result when the nonlocal fluctuations of the Peierls gap are taken into account. This analysis is only valid for short times and small fluctuations and does not provide quantitative information on the last stages of evolution of the instability. However, the fact that the bifurcation appears for a range of length scales that is bounded both from above and below points to a stable and sharply defined mixed state at all times. Moreover, a recent nonlinear analysis of the nonequilibrium superconductor by Hida[20] indicates that for moderate pumping strengths the mixed state is indeed the stable one.

As we pointed out in Sec. I, this type of phenomena can in principle be found in many other condensed matter systems that possess a gap parameter similar to the BCS one. We chose the Peierls insulator because we believe it may become a more suitable probe of these instabilities than superconductors or charge-density-wave systems. Moreover, the possibility of probing the nonequilibrium fluctuations in quasi-one-dimensional solids might become a reality in those solids that display the crossover characteristics found in potassium cyanoplatinide) (KCP) and $(TTT)_2I_3$.

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[15]For a system to be stable, its free energy must be a minimum, and hence $\partial^2 F/\partial \Delta n^2 > 0$. Finding a situation for which this quantity changes sign amounts to be closer to a maximum of the free energy. Therefore, the system will flow towards a configuration which minimizes it.

[16]Since we have neglected higher-order corrections to the linear-stability analysis, it is not possible to make quantitative predictions concerning the final state of the system. However, the fact that the range of wave vectors, q, for which $\alpha > 0$ is bounded from below indicates that the later stages of the instability will be characterized by a sharply defined periodic structure.

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