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Long-time behavior of Ginzburg-Landau systems far from equilibrium

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Using singular-perturbation techniques, we study the stability of modulated structures generated by driving Ginzburg-Landau systems far from equilibrium. We show that, far from equilibrium, the steady-state behavior is controlled by an effective Lagrangian which possesses the same functional form as the original free energy but with renormalized coefficients. We study both linear and nonlinear sources and determine their influence on the long-term stability of the bifurcating solutions.

I. INTRODUCTION

Condensed matter driven far from equilibrium can display a variety of behaviors which are not usually found in its equilibrium state. As the examples of stressed fluids and supercooled alloys show, far from equilibrium one can encounter regimes that can vary from a totally chaotic pattern to highly organized structures, characterized by temporal or spatial modulations of their properties. In particular, the possibility of generating modulated structures has led to the use of techniques such as spinodal decomposition to produce alloys with spatial composition variations that can be designed through rapid quenches.

In problems such as spinodal decomposition, a system is rendered unstable via a rapid change in the thermodynamic parameters, such as temperature or pressure.[1] The ensuring process then corresponds to a growth of fluctuations for a range of wave vectors that carry the system into the stable, equilibrium phase. This process is characterized by the initial appearance of a periodic modulation in composition which coarsens at later times.[2,3] The process of coarsening mathematically corresponds to the fact that the range of positive wave vectors for which fluctuations are unstable is bounded from above but not from below. Therefore, although slow in their growth, the unstable long-wavelength unstable modes lead to phase separation for very long times.

A different type of behavior is encountered in Ginzburg-Landau systems kept far from equilibrium by an external continuous source. Such can be the case in binary mixtures undergoing chemical reactions,[4] superconducting films under a microwave field or strong injection of quasiparticles,[5] Peierls insulators in the presence of electromagnetic radiation, or semiconducting ferroelectrics.[6] In these systems, although one also observes a bifurcation away from a spatially homogeneous state, the range of wave vectors for which fluctuations become unstable is bounded both from above and below. This constraint effectively quenches the growth of long-wavelength modes, leading to a sharp modulated structure which has been recently reported in superconductors.[7]

The analysis of all these phenomena usually proceeds in two stages. In a first attempt at solving the nonlinear equations describing the dynamics of the order parameter, a linear stability analysis is performed. This implies looking at the behavior of small fluctuations away from the initial state and studying their temporal evolution. For the case of unstable modes one then identifies the fastest growing one as determining the basic period, which can in general be computed from a knowledge of the eigenvalues for the linearized problem. The second stage, which is mathematically more involved, tries to ascertain the nonlinear interactions between various unstable modes at later stages and their role in determining the ultimate fate of the instability.

In this paper we present a general technique for dealing with the long-time behavior of Ginzburg-Landau systems far from equilibrium. Our theory, which relies on singular-perturbation techniques,[8,9] leads to surprisingly simple results, which determine not only the stability of the bifurcating solutions for long times, but also their spatial structures. We also show that the functional form of the source terms responsible for keeping the system far from equilibrium is crucial to the stability of the bifurcating solutions. Such a result is not evident in any linearized stability approach to the problem.

We have organized this paper as follows: In Sec. II we study some basic models and their bifurcations as a function of the intensity of the external sources, which we assume are time independent. In particular, we explicitly solve problems containing linear and nonlinear source terms, and calculate the wavelength of the fastest growing instabilities. Section III deals with the nonlinear dynamics of the bifurcating solutions. By using multiple-scale techniques in space and time, we obtain an asymptotic power-series expansion for the unstable modes which leads to a complicated functional relation for their space-time dependence. Using a Laplace method in the resulting integral we are then able to obtain the long-time behavior of the solutions, which is determined by an effective Lagrangian functional. This Lagrangian possesses the same functional form as the original Ginzburg-Landau free energy of the system, but its coefficients now contain information on both the source terms and kinetics of the system. In Sec. IV we look at the stability of the long-time amplitudes for specific source terms and show that linear sources do not lead to stationary periodic patterns, whereas nonlinear ones do stabilize the modulated structures. We also point out that the mode which ultimately gets selected strongly depends on the boundary conditions of the problem.

Throughout this paper we deal with a one-dimensional problem. Although there exist many physical systems for which our results are of relevance, a generalization of our results to higher dimensions may lead to different stability conditions, such as those encountered by Newell and Whitehead in their study of the Rayleigh-Bernard problem.[9] Nevertheless we believe that our basic results can be used as a general test of the behavior of condensed matter systems far from equilibrium.

II. MODELS AND BIFURCATIONS

A. Linear source terms

Consider a quasi-one-dimensional Peierls insulator driven far from equilibrium by dynamic photoproduction of electron-hole pairs. This state can be produced by laser pumping across the gap Δ, in which case an excess number of electrons can be produced. If the time scales involved are such that the electrons in the upper band thermalize down to the bottom of the band in times that are short compared with recombination times, we can assume the existence of a quasi-equilibrium situation. Under those circumstances, the rate equations for the excess electron density, Δn, can be written as

$$\frac{\partial \Delta n}{\partial t} = I[n] - \vec{\nabla} \cdot \vec{J} \tag{2.1}$$

with $I[n]$ the source term specifying the detailed dynamics of photoproduction and recombination, and J the excess electron current, which in terms of the effective chemical potential μ^* is given by

$$\vec{J} = -N(0) D \vec{\nabla} \mu^* \tag{2.2}$$

with D the diffusion coefficient and $N(0)$ the density of states per atom. We have previously shown[6] that as the number of excess electrons is increased, μ^* first increases, until a critical density, Δn_c, is reached for which the system becomes unstable, i.e., $(\partial \mu^* / \partial \Delta \nu)_c = 0$. Near the instability point, μ^* can be written as

$$\mu^*(\Delta n) = \mu^*(\Delta n_c) - \frac{C(\Delta n_c - \Delta n)^2}{N(0)\Delta_0} , \tag{2.3}$$

where C is a constant of order unity, $N(0)$ the density of states at the Fermi level, and Δ_0 the value of the gap at $T=0$.

The nonlocal contirbution to $\mu^*(\Delta n)$ arises from considering the extent to which spatial fluctuations in the gap order parameter modify the free energy. If the latter possesses a Ginzburg-Landau form the correlation energy can be written as $\int N(0)\xi_0^2 |\nabla \Delta|^2 dx$, with ξ_0 the zero-temperature coherence length, which is given by[6]

$$\xi_0^2 = \frac{7\zeta(3)\hbar^2 v_F^2}{16\pi^2 k_B^2 T_c} , \tag{2.4}$$

with v_F the Fermi velocity of the electrons, k_B the Boltzmann constant, and T_c the critical Peierls temperature. Assuming that the gap is renormalized by the excess electrons as $\Delta = \Delta_0(1 - \frac{1}{2}\Delta n)$ and since $\mu^*(r) = \delta F[\Delta n]/\delta \Delta n(r)$ we obtain, together with Eq. (2.3)

$$\mu^*(\Delta n) = \mu^*(\Delta n_c) - (\Delta n - \Delta n_c)^2 \frac{\xi_0^2}{2N(0)} \nabla^2(\Delta n) . \tag{2.5}$$

In order to completely specify the kinetics as given by Eq. (2.1) we need to write down the source term. If I is the rate at which electrons are pumped across the gap and τ their recombination time, Eq. (2.5), together with Eq. (2.2) gives

$$\frac{\partial \Delta n}{\partial t} = I - \frac{\Delta n}{\tau} - \frac{CD}{\Delta_0} \frac{\partial^2}{\partial x^2} (\Delta n - \Delta n_c)^2 - D\xi_0^2 \frac{\partial^4 \Delta n}{\partial x^4} , \tag{2.6}$$

which completely determines the excess electron population within the model.[10]

The stability of the solutions of Eq. (2.6), together with their possible bifurcations, can be studied using linear stability theory. We first notice that a possible steady state solution of Eq. (2.6) can be written as

$$\Delta n_0 = I\tau \tag{2.7}$$

with no spatial dependence. In order to study the stability of this solution against small fluctuations we write

$$\Delta n = I\tau + u(x,t) \tag{2.8}$$

with $u << I\tau$ and behaving as

$$u(x,t)=\sum_q e^{\alpha(q)t}\cos qr \quad . \tag{2.9}$$

Replacing Eqs. (2.8) and (2.9) into (2.6) and keeping only terms linear in u one obtains the following equation for $\alpha(q)$:

$$\alpha(q)=-\left[\frac{1}{\tau}=\frac{2CD}{\Delta_0}(\Delta n_c-I\tau)q^2+\frac{D\xi_0^2}{2}q^4\right] \quad , \tag{2.10}$$

whose behavior as a function of q, for several values of the laser pumping power I, is shown in Fig. 1. As can be seen, for values of I such that $I\tau < \Delta n_c$, $\alpha(q) < 0$ for all wave vectors, and fluctuations away from the uniform solution Δn_c die away exponentially fast. However, for values of $I > \Delta n_c/\tau$, $\alpha(q)$ can start increasing until a critical power is reached, I_c, beyond which α can become positive for a range of wave vectors. Under those conditions fluctuations of the form given by Eq. (2.9) will grow away from the steady state solution and a spatially modulated gap state will start to set in. The bifurcation point can be obtained from the condition that Eq. (2.10) has a double root. This leads to

$$I_c=\frac{1}{\tau}\left[\Delta n_c+\frac{\xi_0\Delta_0}{c\sqrt{2D\tau}}\right] \quad , \tag{2.11}$$

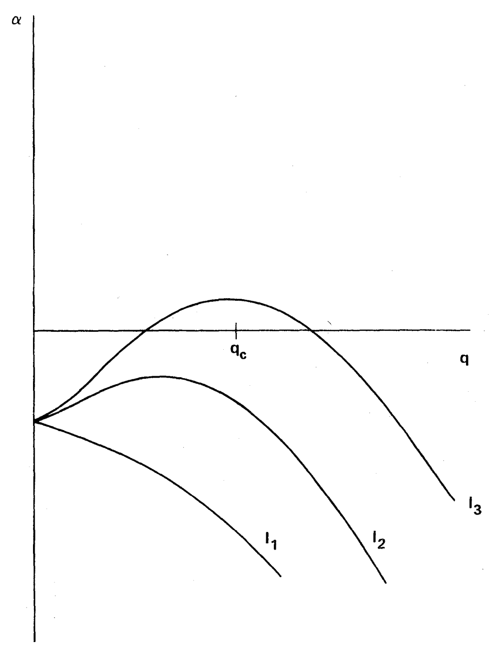

FIG. 1. The amplification factor as a function of wave vector for a set of pumping levels.

whereas the maximum of $\alpha(q)$ at I_c gives

$$q_c=\frac{1}{\xi_0}\left[\frac{1}{D\tau}\right]^{1/2} \quad . \tag{2.12}$$

Within the limits of linear stability Eqs. (2.11) and (2.12) specify the threshold power and the wavelength of the most unstable mode. It is clear however, that the linearized analysis is inadequate in two respects: (1) it ignores all the interactions between the modes within the range $q_1 \leq q \leq q_2$ that become unstable, and (2) for long times the exponential growth of the bifurcating solution will violate the assumption that $u << \Delta n_0$. These are the problems that the nonlinear analysis of Sec. III will deal with.

B. Nonlinear source terms

We now turn our attention to systems where the kinetic terms in the absence of mass currents are nonlinear. Such a situation can be encountered in Peierls insulators or ferroelectric semiconductors with high excess electron densities, nonequilibrium superconductors under microwave fields, or certain chemical autocatalytic reactions. In what follows we will deal explicitly with nonequilibrium superconductors because of their present experimental interest.[11]

Consider a thin strip of superconducting metal below its critical temperature in the presence of an external source of quasiparticles. Since the BCS gap equation is formally similar to the problem of a Peierls insulator, the analysis of the previous section allows us at once to write an equation for the excess quasiparticle current. Adding the Rothwarth-Taylor equations for the time rate of the quasiparticle density to the kinetic terms of our previous case, we obtain[5]

$$\frac{\partial N_{\rm qp}}{\partial t}=I_{\rm qp}-2rN_{\rm qp}^2+2\beta_0 N_{\rm ph}-\vec{\nabla}\cdot\vec{J} \quad , \tag{2.13a}$$

$$\frac{\partial N_{\rm ph}}{\partial t}=I_{\rm ph}+rN_{\rm qp}^2-\beta_0 N_{\rm ph}-\frac{N_{\rm ph}-N_{\rm ph}^e}{\tau_{es}} \quad , \tag{2.13b}$$

where $I_{\rm qp}$ represents a quasiparticle source, $(2RN_{\rm qp})^{-1}$ is the quasiparticle recombination time τ_R and β_0^{-1} is the pairbreaking lifetime, τ_B, of the phonons. $N_{\rm ph}$ is the nonequilibrium density of phonons with energies greater than 2Δ, $I_{\rm ph}$ the phonon source, $\tau_{\rm es}$ the phonon escape time to the thermal bath, and $N_{\rm ph}^e$ the bath equilibrium phonon density. The last term of Eq. (2.13a) is identical to the last two of Eq. (2.6). We can now study the bifurcation solutions away from the uniform steady state in a form similar to that of the previous section. Writing

$$N_{\rm qp}=N_{\rm qp}^0+\delta N_{\rm qp}\sum_q e^{\alpha(q)t}\cos qx \quad , \tag{2.14}$$

$$N_{\rm ph}=N_{\rm ph}^0+\delta N_{\rm ph}\sum_q e^{\alpha(q)t}\cos qx \quad , \tag{2.15}$$

where the uniform steady state solutions, $N_{\rm qp}^0$ and $N_{\rm ph}^0$, are given by

$$N_{\rm qp}^0 = \frac{1}{r^{1/2}}[\beta_0\tau_{\rm es}I_{\rm ph} + \tfrac{1}{2}I_{\rm qp}(1+\beta_0\tau_{\rm es}) + \beta_0 N_{\rm ph}^e]^{1/2} , \quad (2.16)$$

$$N_{\rm ph}^0 = (I_{\rm ph} + \tfrac{1}{2}I_{\rm qp})\tau_{\rm es} + N_{\rm ph}^e , \quad (2.17)$$

and keeping linear terms in $\delta N_{\rm qp}$ and $\delta N_{\rm ph}$ we obtain the following secular equation for $\alpha(q)$:

$$[\alpha(q) + 2\tau_r^{-1} + D(q)][\alpha(q) + \tau_B^{-1} + \tau_{\rm es}^{-1}] - 2\tau_r^{-1}\tau_B^{-1} = 0 \quad (2.18)$$

with

$$D(q) = Dq^2[2(N_c - N_{\rm qp}^0)/N(0)\Delta_0 + \tfrac{1}{2}\xi_0^2 q^2] . \quad (2.19)$$

Equations (2.18) and (2.19) imply an instability for fluctuations in a range of wave vectors, with the fastest growing one corresponding to the maximum of $\alpha(q)$. From the condition that Eq. (2.19) has a double root we can obtain the value of the critical wave vector q_c for which the instability sets in. We obtain

$$q_c^2 = \frac{2}{\xi_0}\left[\frac{2rN_{\rm qp}^0}{D(1+\beta_0\tau_{\rm es})}\right]^{1/2} , \quad (2.20)$$

where the critical density $N_{\rm qp}^c$ is given by

$$N_{\rm qp}^c = N_c + \mu^2 + (\mu^4 + 2N_c\mu^2)^{1/2} \quad (2.21)$$

and

$$\mu^2 \equiv \frac{N_0^2\Delta_0^2\xi_0^2 r}{D(1+\beta_0\tau_{\rm es})} . \quad (2.22)$$

This completes our linear stability analysis of Ginzburg-Landau systems far from equilibrium. In what follows we will study the nonlinear effects that become important at the bifurcation point.

III. NONLINEAR DYNAMICS

The results of the previous section allowed us to determine the critical pumping power for which fluctuations centered around a critical wave vector would become unstable. Regardless of the linear or nonlinear character of the source terms, the linearized stability analysis was able to predict that a modulated structure would start growing exponentially fast as the external driving source exceeded a certain threshold. It is clear however, that those results are of validity only for extremely short times. As the instability sets in, two effects conspire so as to render the predictions of the linear theory unreliable: (1) for long times the approximation of small fluctuations breaks down; and (2) instead of a single unstable mode, fluctuations with a range of different length scales will start to grow in time, with nonlinear interactions among themselves which will affect the ultimate fate of the bifurcated state.

In order to study the long-time evolution of the bifurcations we will first concentrate on the case of linear sources. Although perhaps not very realistic when compared to the high levels of excitation needed to drive a Peierls system of a ferroelectric semiconductor unstable, this simple case will illustrate the method of calculation in a clearer fashion than using a nonlinear source as an example. The details of the nonlinear analysis for the superconducting case will be worked out in Appendix A. Those results will be directly applicable to more complicated kinetics.

In order to set up a convenient scheme for a perturbation theory it is convenient to introduce new space and time scales. These scales are naturally suggested by the functional form of the amplification factor for pumping levels above the threshold value. If we denote by ϵ the dimensionless small parameter (see Fig. 2)

$$\epsilon = \frac{q_2 - q_1}{q_c} \quad (3.1)$$

we can introduce a new length $\tilde{x}$ and a rescaled time $\tilde{t}$ defined as

$$\tilde{x} = \epsilon x , \quad (3.2a)$$

$$\tilde{t} = \epsilon^2 t , \quad (3.2b)$$

so that the partial derivatives entering Eq. (2.4) be-

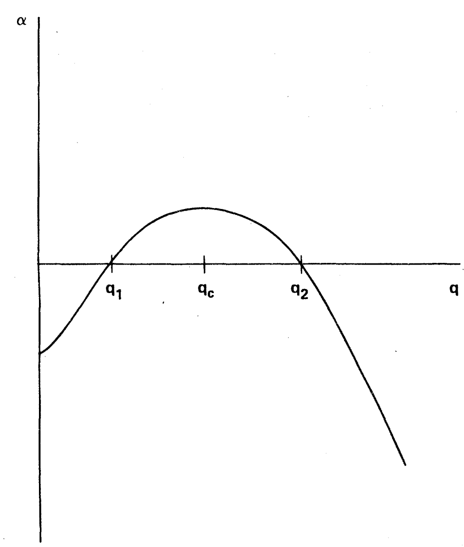

FIG. 2. The range of wave vectors that become unstable near threshold.

come

$$\partial_x \equiv \partial_x + \epsilon \partial_{\tilde{x}} \quad (3.3)$$

and

$$\partial_t \equiv \epsilon^2 \partial_{\tilde{t}} \ . \quad (3.4)$$

Writing the bifurcation control parameter as

$$I = I_c + s\epsilon^2 \quad (3.5)$$

with $s = \pm 1$ indicating whether one is below or above the critical value, we can express Eq. (2.7) in operator form as

$$L(\epsilon)u = N(u) \ , \quad (3.6)$$

where the linear operator $L(\epsilon)$ is given by

$$L(\epsilon) = \epsilon\partial_{\tilde{t}} + \frac{1}{\tau} + \tfrac{1}{2} D\xi_0^2(\partial_x + \epsilon\partial_{\tilde{x}})^4 + 2CD(I_c\tau - \Delta n_c + s\epsilon^2)(\partial_x + \epsilon\partial_{\tilde{x}})^2 \quad (3.7)$$

and the nonlinear operator $N(u)$ can be written as

$$N(u) = \frac{CD}{\Delta_0}(\partial_x + \epsilon\partial_{\tilde{x}})^2 u^2 \ . \quad (3.8)$$

In order to solve Eq. (3.6) we assume the existence of an asymptotic power-series expansion in ϵ for the bifurcating amplitude u and write

$$u = \epsilon u_0 + \epsilon^2 u_1 + \epsilon^3 u_2 + O(\epsilon^4) \quad (3.9)$$

so that in the limit $\epsilon \rightarrow 0$, $u \rightarrow 0$, in agreement with the linear stability analysis of the previous section. Replacing the expansion given by Eq (3.9) in Eq. (3.6) we obtain

$$L(\epsilon) = L_0 + \epsilon L_1 + \epsilon^2 L_2 + O(\epsilon^3) \quad (3.10)$$

with

$$L_0 = \frac{1}{\tau} + \frac{D\xi_0^2}{2}\partial_x^4 + \frac{2CD}{\Delta_0}(I_c\tau - \Delta n_c)\partial_x^2 \ , \quad (3.11a)$$

$$L_1 = 2D\xi_0^2\partial_{xxx\tilde{x}} + \frac{4CD}{\Delta_0}(I_c\tau - \Delta n_c)\partial_{x\tilde{x}} \ , \quad (3.11b)$$

and

$$L_2 = \left[\partial_{\tilde{t}} + \frac{2CD}{\Delta_0}(I_c\tau - \Delta n_c)\partial_{\tilde{x}\tilde{x}} + \frac{2SCD}{\Delta_0}\partial_{xx} + 3D\xi_0^2\partial_{xx\tilde{x}\tilde{x}} \right] , \quad (3.11c)$$

whereas Eqs (3.8) and (3.9) become

$$N(u) = \epsilon^2 N_0(u_0) + \epsilon^3 N_1(u_0,u_1) + O(\epsilon^4) \ , \quad (3.12)$$

with

$$N_0(u_0) = \frac{CD}{\Delta_0}\partial_{xx}u_0^2 \quad (3.13a)$$

and

$$N_1(u_0,u_1) = 2\partial_{x\tilde{x}}u_0^2 + 2\partial_{xx}u_0u_1 \ . \quad (3.13b)$$

Equating the coefficients of the power-series expansion in Eq. (3.6) we then obtain the following system of equations:

$$L_0u_0 = 0 \ , \quad (3.14)$$

$$L_0u_1 = -L_1u_0 + N_0u_0 \ , \quad (3.15)$$

and

$$L_0u_2 = -L_2u_0 - L_1u_1 + N_1(u_0,u_1) \quad (3.16)$$

which, once solved, will determine the space-time dependence of the modulation, u, to order ϵ^3 away from the bifurcation point.

First, we note that Eq. (3.14) is a restatement of our linear stability analysis, so that u_0 is given by

$$u_0 = \int_{-\infty}^{\infty} [A(\tilde{x},\tilde{t},q)\cos qx + B(\tilde{x},\tilde{t},q)\sin qx]e^{\alpha(q)t}dq \ . \quad (3.17)$$

This is the full solution, valid for short times as well, but it leads to expressions of unmanageable complexity. The picture simplifies somewhat if one considers the long-time behavior of the equation. Since $\alpha(q)$ has a maximum at q_c we can approximate the integral by Laplace's method and obtain

$$u_0 = A(\tilde{x},\tilde{t})\cos q_cx + B(\tilde{x},\tilde{t})\sin q_cx \quad (3.18)$$

with the functions $A(\tilde{x},\tilde{t})$ and $B(\tilde{x},\tilde{t})$ as yet undetermined and q_c given by Eq. (2.10). Moreover, since q_c is a double root at the bifurcation point it follows that $L_1u_0 = 0$. Equation (3.15) can be then solved for u_1 yielding

$$u_1 = -\frac{4\tau q_c^2}{9}\frac{CD}{\Delta_0}\left[\frac{A^2 - B^2}{2}\cos 2q_cx + AB\sin 2q_cx\right] . \quad (3.19)$$

In order to determine the long space-and-time dependences of the coefficients A and B we now need to consider Eq. (3.16). To $O(\epsilon^3)$ it reads

$$L_0u_2 = -L_2u_0 + \frac{2CD}{\Delta_0}\partial_{xx}(u_0u_1) \ . \quad (3.20)$$

Since Eqs. (3.18) and (3.19) imply that

$$u_0u_1 = -\frac{4\tau q_c^2}{18}\frac{CD}{\Delta_0}\left[\frac{A^2 + B^2}{2}\right]u_0 \quad (3.21)$$

and

$$\frac{2CD}{\Delta_0}\partial_{xx}(u_0u_1) = \left[\frac{4C^2D}{(q\xi_0^2\Delta_0^2)}\right](A^2 + B^2)u_0 \ .$$

Equation (3.11c) allows us to write Eq. (3.20) as

$$L_0 u_2 = \left[-\partial_{\tilde t} + \frac{2CD}{\Delta_0}(I_c\tau - \Delta n_c)\partial_{\tilde x\tilde x} + \frac{2SDC}{\Delta_0}\partial_{xx} + 3D\xi_0^2\partial_{xx\tilde x\tilde x} + \frac{4C^2D}{q\xi_0^2\Delta_0}(A^2+B^2)\right] u_0 \ , \quad (3.22)$$

which determines the behavior of u_2 in terms of u_0. We first notice that since the right-hand side of this equation acts as a forcing term at a resonance, it would cause u_2 to become unbounded, in contradiction to the assumption that there exists an asymptotic series expansion for the bifurcating solution u, as given by Eq (3.9). Therefore, in order to avoid the divergence in u_2 we choose A and B such that the resonant terms vanish. Setting the expression in large parenthesis in Eq. (3.22) equal to zero yields

$$\partial_{\tilde t} u_0 - \frac{4}{q_c^2\tau}\partial_{\tilde x\tilde x} u_0 = \frac{2CD\tau}{\Delta_0} q_c^2 u_0 + \frac{2}{q}\left[\frac{2C^2D}{\Delta_0^2\xi_0^2}\right](A^2+B^2)u_0 \quad (3.23)$$

or, using Eq. (3.18)

$$\partial_{\tilde t} A - \left[2\xi_0\left[\frac{2D}{\tau}\right]^{1/2}\right]\partial_{\tilde x\tilde x} A = \frac{2CS\sqrt{2D\tau}}{\xi_0\Delta_0}A + \frac{2}{q}\left[\frac{2C^2D}{\Delta_0^2\xi_0^2}\right](A^2+B^2)A \ , \quad (3.24a)$$

$$\partial_{\tilde t} B - \left[2\xi_0\left[\frac{2D}{\tau}\right]^{1/2}\right]\partial_{\tilde x\tilde x} B = \frac{2CS\sqrt{2D\tau}}{\xi_0\Delta_0}B + \frac{2}{q}\left[\frac{2C^2D}{\Delta_0^2\xi_0^2}\right](A^2+B^2)B \ . \quad (3.24b)$$

Equations (3.24), together with Eq. (3.18), determine the nonlinear behavior of the bifurcating solution away from the uniform state. Their meaning becomes more transparent when expressed in functional form. Introducing a complex parameter $\eta = (A+iB)$ they can be rewritten as

$$\frac{\partial\eta}{\partial\tilde t} = \frac{\delta L}{\delta\eta^*} \ , \quad (3.25)$$

where the new functional L is given by

$$L = \int\left[\frac{S\alpha}{2}|\eta|^2 + \frac{\beta}{4}|\eta|^4 + \frac{\gamma}{2}\left|\frac{\partial\eta}{\partial\tilde x}\right|^2\right]dx \ , \quad (3.26)$$

and

$$\alpha = \frac{2C\sqrt{2D\tau}}{\xi_0\Delta_0} \ , \quad (3.27)$$

$$\beta = \frac{2}{q}\left[\frac{2DC^2}{\Delta_0^2\xi_0^2}\right] \ , \quad (3.28)$$

$$\gamma = 2\xi_0\left[\frac{2D}{\tau}\right]^{1/2} \ . \quad (3.29)$$

We have therefore shown that the long-time behavior of the modulated state is governed by a new "Lagrangian" which has the same functional form as the original Ginzburg-Landau free energy but with renormalized coefficients. These coefficients contain the detailed kinetics of the source term.

IV. RESULTS

The lengthy calculations of Sec. III enabled us to derive an equation of motion for the slowly varying amplitude of the modulated structure as the system is driven far from equilibrium by a steady state source. Since formally this equation resembles the dynamics of a complex order parameter near a phase transition,[12] we can use the considerable amount of work that has been devoted in that field in order to obtain concrete analytic results.

We start by looking at the stability of the bifurcating solutions. Writing $\eta = Re^{i\phi} = A + iB$, the functional L given by Eq. (3.26) becomes

$$L = \int d\tilde x\left[\frac{S\alpha}{2}R^2 + \frac{\beta R^4}{r} - \frac{\gamma}{2}[(\partial_{\tilde x}R)^2 + (R\,\partial_{\tilde x}\phi)^2]\right] \quad (4.1)$$

so that Eq. (3.25) can be expressed as

$$\partial_{\tilde t}R = S\alpha R + \beta R^3 - \gamma R(\partial_{\tilde x}\phi)^2 - \gamma\partial_{\tilde x\tilde x}R \quad (4.2)$$

and

$$R^2\partial_{\tilde t}\phi = \gamma\partial_{\tilde x}(R^2\partial_{\tilde x}\phi) \ . \quad (4.3)$$

Demanding $\partial_t \equiv 0$ we can find all the possible steady-state structures of the bifurcating state. Setting $\phi_t = 0$ in Eq (4.3) implies that $\partial_{x\phi} = \nu R^{-2}$ with ν a constant. We then obtain for the real part of η the following equation:

$$\gamma\partial_{\tilde x\tilde x}R + S\alpha R + \beta R^3 - \frac{\nu\gamma}{R^3} = 0 \ , \quad (4.4)$$

whose stability depends on the relative signs of α and β, as we show below. For linear sources we found in the previous section [Eqs. (3.27) and (3.28)] that $\alpha > 0$ and $\beta > 0$. In this case we find that the system displays an inverted bifurcation. For values of the pumping power, I, smaller than the critical value I_c (which corresponds to $s=-1$) large enough fluctuations can make the uniform-state solution unstable and a modulated structure will grow in time. Its ultimate fate, however, cannot be determined within the limits $[O(\epsilon^3)]$ of our perturbation theory.

For nonlinear sources, such as nonequilibrium superconductors, it is shown in Appendix A that the coefficients entering the functional L are given by

$$\alpha=\frac{4r(\alpha_0-1)}{1+\beta_0\tau_{es}+rN^0_{qp}\beta_0\tau_{es}} , \tag{4.5}$$

$$\beta=\frac{4r(\alpha_0-1)(\alpha_0-\frac{19}{4})}{N^0_{qp}(1+\beta_0\tau_{es}+4rN^0_{qp}\beta_0\tau_{es})} , \tag{4.6}$$

$$\gamma=\frac{4\xi_0[(2rN^0_{qp}D)(1+\beta_0\tau_{es})]^{1/2}}{1+\beta_0\tau_{es}+4rN^0_{qp}\beta_0\tau_{es}} , \tag{4.7}$$

so that since $\alpha>0$ and $\gamma>0$, a negative β will lead to a stable nonuniform state to the right ($s=+1$) of the critical point I_c, while for $I<I_c(s=-1)$ the uniform state is stable.

We should point out that none of these results could have been anticipated from linear stability theory. As shown in Sec. II, both the systems with linear and nonlinear sources displayed an instability towards a modulated state characterized by an amplification factor of the same functional form. It is only for longer times that the presence or absence of nonlinearities in the source term act so as to stabilize or destabilize the nonuniform state.

We now study Eq. (4.4) in detail to determine the possible steady-state structures of the stable modulated state. Introducing rescaled variables such that $T=\alpha t$, $X=(\alpha/\gamma)^{1/2}x$, and $\hat{R}=(\beta/\alpha)^{1/2}R$, the steady state solutions of Eq. (4.4), for the case $s=+1$, $\beta<0$ become

$$\partial_{xx}\hat{R}=-\hat{R}+\hat{R}^3+\frac{\nu}{\hat{R}^3} \tag{4.8}$$

which corresponds to the motion of a particle of unit mass in an effective potential given by

$$V_{\text{eff}}(\hat{R})=\frac{\nu}{\hat{R}^2}+\hat{R}^2-\hat{R}^4 , \tag{4.9}$$

i.e., an anharmonic problem with a centrifugal term proportional to ν. This problem is analogous to the one encountered in fluctuating one-dimensional superconductors[13] and finite amplitude convection.[9] Equation (4.8) can be integrated to yield

$$(\partial_x\hat{R})^2=\frac{\hat{R}^4}{2}-\hat{R}^2-\frac{\nu^2}{\hat{R}^2}+\Gamma \tag{4.10}$$

with Γ a constant; so that if we set $R^2=\rho$, we obtain

$$X=\frac{1}{2}\int_{R_0^2}^{R^2}\frac{d\rho}{(\rho 3/2-\rho^2+\Gamma\rho-\nu^2)^{1/2}} . \tag{4.11}$$

Depending on the behavior of the function $f(\rho)=\rho_2^3-\rho^2+\Gamma\rho-\nu^2$ Eq. (4.11) will give rise to several possible patterns of the modulated state. Since which one is ultimately chosen will depend on the initial conditions, we will not list them all. It is worth noting however, that besides a uniform modulation Eq. (4.11) has a particular solution given by

$$\hat{R}=\tanh\frac{X}{\sqrt{2}} \tag{4.12}$$

which implies that the steady state full bifurcating solution, as given by Eq. (3.18), will behave as

$$u_0(x)=\tanh\frac{X}{\sqrt{2}}\cos q_c x , \tag{4.13}$$

which corresponds to the existence of two uniform domains of the same wave number q_c but out of phase by 180°, with their amplitude vanishing out smoothly at their interface (Fig. 3). It is easy to see that for all pairs of values of Γ and ν for which the minimum of the cubic in (4.11) is on the ρ axis, there exist steady-state solutions that are stable to one-dimensional perturbations (Fig. 4). At $x\rightarrow\pm\infty$ these solutions approach a uniform state with $\phi=\pm(1-R^2)^{1/2}$, $1/\sqrt{3}\leq R\leq 1$ (two domains out of phase but joined smoothly). A straightforward

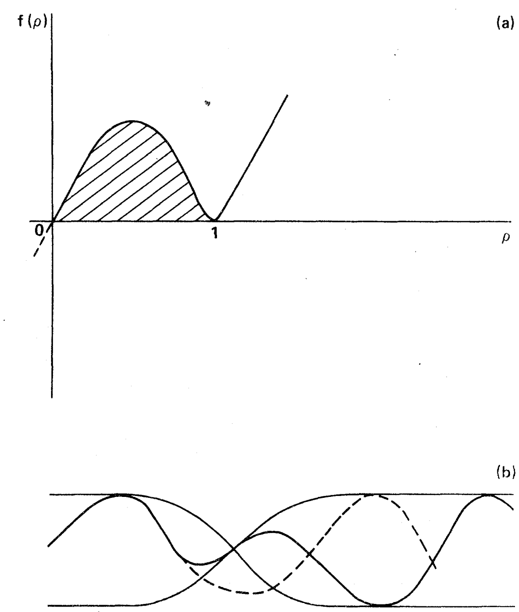

FIG. 3. The case of two uniform domains with the same characteristic wave vector, q_c, but out of phase. (a) The function $f(\rho)$. (b) The amplitude of the modulated state as a function of position. Solid line: full solution with continuous interphase. Dashed line: continuation of full solution illustrating the phase shift.

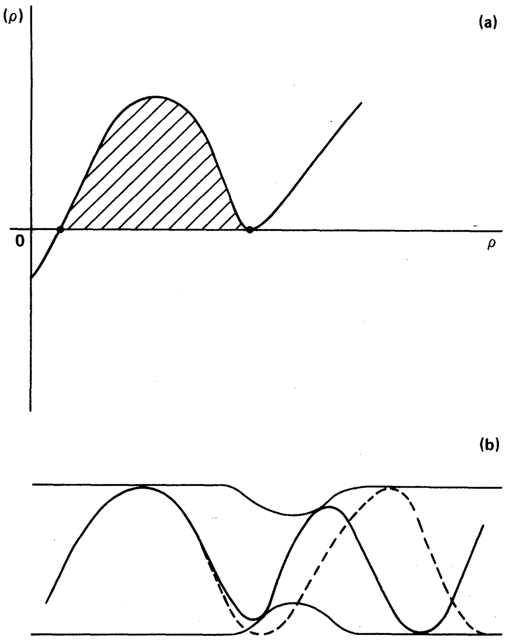

FIG. 4. Two uniform regions with $q+q_c$ joined by an interface. (a) The function $f(\rho)$ for that particular case. (b) The amplitude as a function of position. Solid line: full solution with different wavelengths in each region. Dashed line: the uniformly modulated pattern.

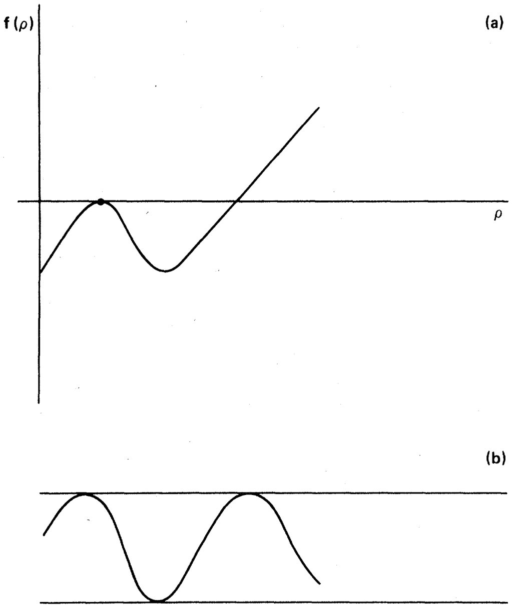

FIG. 5. Uniformly modulated state. (d) The function $f(\rho)$ for this case. (b) The uniform, but unstable, solution corresponding to the maximum of the cubic with $O \leq R \leq 1/\sqrt{3}$.

analysis shows that small perturbations in these uniform states obey a diffusion equation and therefore they decay eventually to a uniform state (but not necessarily the initially perturbed one, if the perturbation is not square integrable).

Uniform states with $O \leq R \leq 1/\sqrt{3}$ correspond to the maximum of the cubic and are unstable (Fig. 5). Solutions periodic in R are also easily seen to be unstable (Fig. 6).

Another interesting class of solutions is found when we look for traveling waves that satisfy Eqs. (4.2) and (4.3). Writing $R=R(x-ct)$ and setting $\phi=$constant we obtain

$$R''+cR'+R-R^3=0 \ , \tag{4.14}$$

which corresponds to a phase plane system with critical points at 0, ±1, with the trajectory joining the points 0 and $+1$ giving a bounded traveling solution. For $0\leq c\leq 2$ the origin is a stable spiral and the corresponding wave is unstable, whereas for $c\geq 2$, 0 is a stable node and the wave monotonic and stable (for $c<0$ the origin is unstable and the waves move to the left). Therefore for $|c|\geq 2$ one finds shocklike solutions propagating from regions where the uniform modulated state is fully developed to regions where the amplitude is zero (Fig. 7).

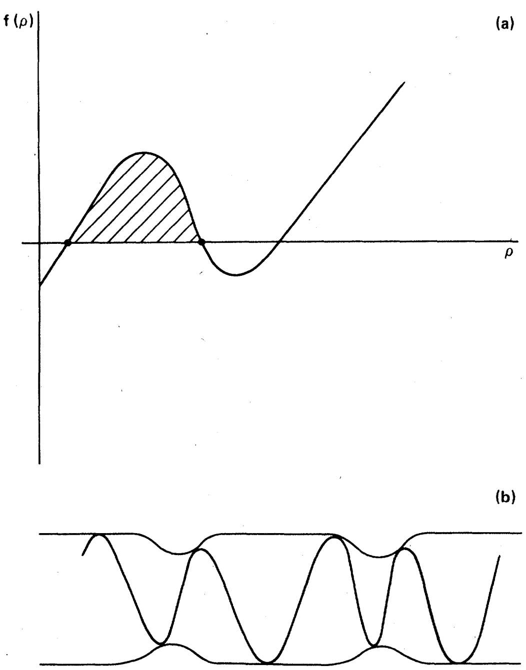

FIG. 6. Solution periodic in R. (a) $f(\rho)$. (b) The amplitude versus position. Linear stability analysis shows this case to be unstable.

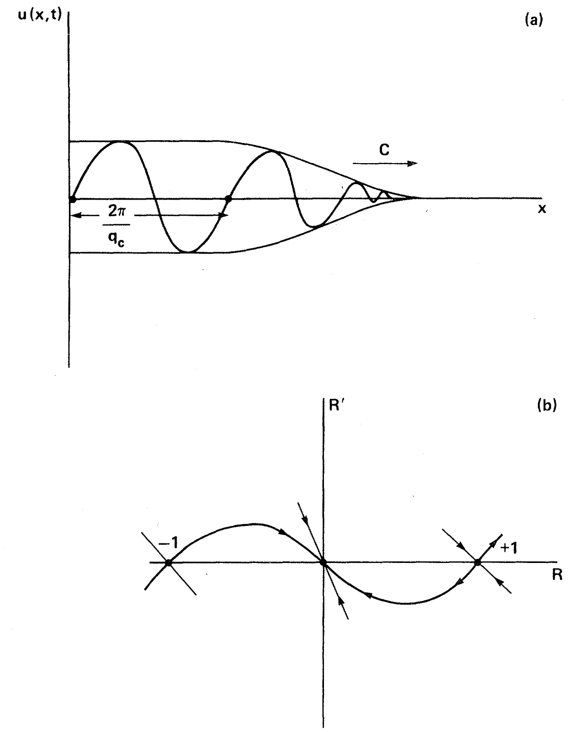

FIG. 7. (a) Nucleation front $\mu = R(x-ct)\cos(q_c x)$ converting the zero-amplitude state into a uniformly modulated one. (b) Phase plane trajectories corresponding to Eq. (4.14) for $2 < c < \infty$. The trajectories connecting ± 1 to the origin correspond to a front in R traveling to the right. If $c < 0$, the directions of all arrows are reversed and the front travels towards the left.

In closing we must stress the fact that all our results are one dimensional. For two-dimensional systems, the process of wave-vector selection involves other factors not included in our analysis, such as the ones considered by Pomeau and Manneville[14] in their study of the Rayleigh-Bernard problem or Langer[15] in the case of dendritic growth.

V. CONCLUSION

Using singular perturbation techniques, we have studied the long-time behavior of the bifurcating solutions of Ginzburg-Landau systems kept far from equilibrium by the action of external sources. This problem is of importance in a variety of situations where external constraints can keep a given system (such as a superconductor) always away from the absolute minimum of its thermodynamic free energy. What we have shown is that the steady-state behavior of such a system is governed by an effective Lagrangian with the same functional form as the original equilibrium free energy, but with coefficients which contain the detailed kinetics of the problem.

Since our methods are quite general, they should be applicable to a variety of stiuations in which condensed matter exhibits cooperative critical behavior which can be modeled by similar free-energy expansions. Also, they point to a possible formulation of a thermodynamics far from equilibrium in which generalized forces can be defined in the same fashion as in the equilibrium problem. On a more concrete level, our analysis indicates that modulated structures can indeed be produced in a stable fashion by driving a wide variety of solids far from equilibrium

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APPENDIX A

In this section we provide the details of the nonlinear analysis for the bifurcating solutions of the nonequilibrium superconductor.[16]

Equation (2.6), which determines the nonequilibrium dynamics of quasiparticles can be written as

$$\hat{L}(\epsilon)\begin{pmatrix}u\\v\end{pmatrix} = \hat{N}\begin{pmatrix}u\\0\end{pmatrix}, \tag{A1}$$

where the linear operator $\hat{L}(\epsilon)$ is given by

$$\begin{aligned}\hat{L}(\epsilon) = {} & \begin{pmatrix}1&0\\0&1\end{pmatrix}\epsilon^2\partial_{\tilde{t}} - \begin{pmatrix}-4rN^0_{\rm qp} & 2\beta_0\\ 2rN^0_{\rm qp} & -\beta_0+1/\tau_{\rm es}\end{pmatrix} - s\epsilon^2\begin{pmatrix}-4r&0\\2r&0\end{pmatrix}\\ & -\begin{pmatrix}1&0\\0&0\end{pmatrix}\left[\frac{2D}{N_0\Delta_0}(N_c-N^0_{\rm qp}-s\epsilon^2)(\partial_x+\epsilon\partial_{\tilde{x}})^2-\frac{D\xi_0^2}{2}(\partial_x+\epsilon\partial_{\tilde{x}})^4\right]\end{aligned} \tag{A2}$$

and the nonlinear operator $\hat{N}(u)$ can be written as

$$\hat{N}(u) = r\begin{pmatrix}-2\\1\end{pmatrix}u^2 - \frac{D}{N_0\Delta_0}\begin{pmatrix}1\\0\end{pmatrix}[(\partial_x+\epsilon\partial_{\tilde{x}})^2u^2]. \tag{A3}$$

In order to solve Eq. (A1) we follow the steps of Sec. III. Assuming the existence of an asymptotic power-series expansion in ϵ for u and v, we write

$$u=\epsilon u_0+\epsilon^2 u_1+\epsilon^3 u_2+O(\epsilon^4) \ , \tag{A3a}$$

$$v=\epsilon v_0+\epsilon^2 v_1+\epsilon^3 v_2+O(\epsilon^4) \ , \tag{A3b}$$

where v, the modulation of the uniform phonon concentration, is important insofar as it affects the concentration of quasiparticles. In what follows we will only use it as an auxiliary variable for determining u.

Expanding Eq. (A2) in powers of ϵ, we obtain

$$\hat{L}(\epsilon)=L_0+\epsilon L_1+\epsilon^2 L_2+O(\epsilon^3) \tag{A4}$$

with

$$L_0=\begin{bmatrix}-4rN_{\rm qp}^0 & 2\beta_0\\ 2rN_{\rm qp}^0 & -(\beta_0+1/\tau_{\rm es})\end{bmatrix} -\begin{bmatrix}1&0\\0&0\end{bmatrix}\left[\frac{2D}{N_0\Delta_0}(N_c-N_{\rm qp}^0)\partial_{xx}-\frac{D\xi_0^2}{2}\partial_{xxxx}\right] , \tag{A5a}$$

$$L_1=-\begin{bmatrix}1&0\\0&0\end{bmatrix}\left[\frac{4D}{N_0\Delta_0}(N_c-N_{\rm qp}^0)\partial_{x\tilde{x}}-\frac{D\xi_0^2}{2}\partial_{xxx\tilde{x}}\right] , \tag{A5b}$$

$$L_2=\begin{bmatrix}1&0\\0&1\end{bmatrix}\partial_{\tilde{t}}-s\begin{bmatrix}-4r&0\\2r&0\end{bmatrix}-\begin{bmatrix}1&0\\0&0\end{bmatrix} \left[\frac{2D}{N_0\Delta_0}(N_c-N_{\rm qp}^0)\partial_{\tilde{x}\tilde{x}}-\frac{2SD}{N_0\Delta_0}\partial_{xx}-3D\xi_0^2\partial_{xx\tilde{x}\tilde{x}}\right] , \tag{A5c}$$

whereas Eq. (A3) can be written as

$$\hat{N}(u)=-\epsilon^2 N_0(u_0)-\epsilon^3 N_1(u_0,u_1)+O(\epsilon^4) \tag{A6}$$

with

$$N_0(u_0)=r\begin{bmatrix}-2\\1\end{bmatrix}u_0^2-\frac{D}{N_0\Delta_0}\begin{bmatrix}1\\0\end{bmatrix}\partial_{xx}u_0^2 \tag{A7a}$$

and

$$N_1(u_0,u_1)=r\begin{bmatrix}-2\\1\end{bmatrix}(2u_0u_1) -\frac{D}{N_0\Delta_0}\begin{bmatrix}1\\0\end{bmatrix}[-2\partial_{x\tilde{x}}u_0^2+2\partial_{xx}(u_0u_1)] \ . \tag{A7b}$$

Proceeding as in Sec. III, we equate the coefficients of the power-series expansion in ϵ and obtain the following system of operator equations:

$$L_0\begin{bmatrix}u_0\\v_0\end{bmatrix}=0 \ , \tag{A8}$$

$$L_0\begin{bmatrix}u_1\\v_1\end{bmatrix}=-L_1\begin{bmatrix}u_0\\v_0\end{bmatrix}+N_0(u_0) \ , \tag{A9}$$

and

$$L_0\begin{bmatrix}u_2\\v_2\end{bmatrix}=-L_2\begin{bmatrix}u_0\\v_0\end{bmatrix}-L_1\begin{bmatrix}u_1\\v_1\end{bmatrix}+N_1(u_0,u_1) \ . \tag{A10}$$

Since Eq. (A8) is a restatement of the linear stability analysis, we can write for u_0 and v_0

$$u_0=A(\tilde{x},\tilde{t})\cos q_c x+B(\tilde{x},\tilde{t})\sin q_c x \ , \tag{A11}$$

$$v_0=\frac{2rN_{\rm qp}^0}{\beta_0+1/\tau_{\rm es}}u_0 \tag{A12}$$

with q_c given by Eq. (2.20). Furthermore, since q_c is a double root at the bifurcation point, it follows that

$$L_1\begin{bmatrix}u_0\\v_0\end{bmatrix}=0 \ .$$

To next order in ϵ [Eq. (A9)] we then have

$$L_0\begin{bmatrix}u_1\\v_1\end{bmatrix}=N_0(u_0) \ , \tag{A9a}$$

which gives for v_1 the following solution:

$$v_1=\frac{2rN_{\rm qp}^0}{\beta_0+1/\tau_{\rm es}}u_1+\frac{r}{\beta_0+1/\tau_{\rm es}}u_0^2 \ ; \tag{A13}$$

writing $\sigma\equiv(1+\beta_0\tau_{\rm es})/4RN_{\rm qp}^0$ we can now express Eq. (A9a) as

$$L_0u_1=\frac{1}{\sigma}\left[\frac{1}{q_c^4}\partial_{xxxx}u_1+\frac{2}{q_c^2}\partial_{xx}u_1+u_1\right] =\frac{-2r}{1+\beta_0\tau_{\rm es}}u_0^2-\frac{D}{N_0\Delta_0}\partial_{xx}u_0^2 \ , \tag{A14}$$

whose solution is

$$u_1=-\frac{1}{2N_{\rm qp}^0}(A^2+B^2)+\frac{1}{q}\left[\frac{4D}{N_0\Delta_0}q_c^2\sigma-\frac{1}{2N_{\rm qp}^0}\right] \times\left[\frac{A^2-B^2}{2}\cos 2q_cx+AB\sin 2q_cx\right] \ . \tag{A15}$$

In order to determine the space-time dependences of the coefficients A and B we need to consider Eq. (A10). After determining from the second equation in the pair, that v_2 is given by

$$v_2=\frac{2rN_{\rm qp}^0}{\beta_0+1/\tau_{\rm es}}u_2-\frac{2rN_{\rm qp}^0}{(\beta_0+1/\tau_{\rm es})^2}\partial_{\tilde{t}}u_0 +\frac{2ru_0}{\beta_0+1/\tau_{\rm es}}(s+u_1) \tag{A16}$$

we obtain for u_2

$$L_0 u_2 = \left\{ -\left[1+\frac{4rN_{qp}^0\beta_0}{(\beta_0+1/\tau_{es})^2}\right]\partial_{\tilde{t}} + 4\xi_0\left[\frac{2rN_{qp}^0 D}{1+\beta_0\tau_{es}}\right]^{1/2} + \frac{(\alpha_0-1)4r}{1+\beta_0\tau_{es}}\left[S+\frac{1}{N_{qp}^0}(\alpha_0-\tfrac{19}{4})(A^2+B^2)\right]\right\} \times (A\cos q_c x + B\sin q_c x) \;, \tag{A17}$$

where

$$\alpha_0 = \frac{1}{N_0\Delta_0\xi_0}\left[\frac{2D(1+\beta_0\tau_{es})}{r}N_{qp}^0\right]^{1/2} . \tag{A18}$$

Choosing the amplitudes A and B such that the resonant terms of Eq. (A17) vanish gives

$$\partial_{\tilde{t}}A - \gamma\partial_{\tilde{x}\tilde{x}}A = \alpha A + \beta(A^2+B^2)A \;, \tag{A19}$$

where

$$\alpha = \frac{4r(\alpha_0-1)}{1+\beta_0\tau_{es}+4rN_{qp}^0\beta_0\tau_{es}} \;, \tag{A20}$$

$$\beta = \frac{4r(\alpha_0-1)(\alpha_0-\frac{19}{4})}{N_{qp}^0(1+\beta_0\tau_{es}+4rN_{qp}^0\beta_0\tau_{es})} \;, \tag{A21}$$

$$\gamma = \frac{4\xi_0[(2rN_{qp}^0 D)(1+\beta_0\tau_{es})]^{1/2}}{1+\beta_0\tau_{es}+4rN_{qp}^0\beta_0\tau_{es}} . \tag{A22}$$

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[10] Notice that the nonlinearity of Eq. (2.6) is only due to the second term of the equation, the source terms being linear since we assume a recombination time τ which is independent of Δn. More complicated nonlinear source terms will be discussed in the following sections.

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