Entrainment by periodic perturbations in the center manifold at Ginzburg-Landau critical regimes

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We consider the effect of periodic perturbations on open reactive systems far from the linear thermodynamics domain. The systems present a center-manifold contraction of the phase space achieved by sustaining a hard-mode instability. The critical regime is governed by a Ginzburg-Landau potential defined on the locally attractive, locally invariant center manifold. Within the framework of this reduction scheme, scaling properties of the Green's function sensitivity matrix are obtained. It is demonstrated that the entrainment is produced by a projection of the perturbing time-dependent field on the center manifold. Thus, the reduced equations for entrainment in the order parameter space are derived. It is demonstrated that two inherent properties of the system favor the entrainment: (a) a small sensitivity of the amplitude of the bifurcating limit cycle with respect to changes in the control parameter, (b) the departure of the system from the region of marginal stability. The results are applied in two different contexts: When there exists a separation of relaxation-time scales (in a truncation of Hopf's model for hydrodynamic turbulence) and when there is only one time scale involved (Brusselator). Agreement with previous derivations of the entrainment regions is found. Finally, a realistic experiment coupling two oscillatory reactors is suggested in order to test the theoretical findings. In this case, the frequency of the perturbation is a function of the bifurcation control parameter (the residence time) which measures the departure of the entrained system from marginal stability. The results are applicable in the case of convection in a rotating layer and convection driven by the Soret-Dufour effect since the oscillatory convection corresponds to a center manifold contraction of the phase space. This manifold contains the dominant velocity modes when the frequency of oscillation is small.

I. INTRODUCTION

The order parameters constitute a quantitative measure for the loss of symmetry in phase transitions, their value is zero in the symmetric phase. In this paper we shall describe the order-parameter space for open reactive systems sustaining a hard-mode instability in a far from equilibrium regime. The center manifold coordinates are the order parameters corresponding to the excited relaxation modes for the symmetry-breaking instability.

We shall be concerned with the situations in which a Ginzburg-Landau (GL) potential functional can be defined on the space of continuous slowly varying order parameters.[1,2] These variables are the slowly relaxing degrees of freedom which correspond to the excited modes at the bifurcation point. The GL phenomenological approach yields the same results as those determined from the Hopf-Birkoff theorem.[3,4] The general restrictions for the validity of GL mean-field equations in far from equilibrium kinetics are stronger conditions than Hopf's hypotheses for bifurcation.[1]

The contraction of the phase space represents the statistical enslaving of the fast-relaxing degrees of freedom to the order parameters.[5] It can be shown that this contraction corresponds asymptotically to the center manifold (c.m.) in the phase space.[6–10] This hypersurface is tangent at the steady state to the order-parameter space.

The dynamics corresponding to the center-manifold reduced equations remain qualitatively valid even for parameter values substantially far from those required for double degeneracy. The results presented in this work are thus applicable to doubly diffusive convection problems displaying oscillatory convection, for example, convection driven by the Soret-Dufour effect and convection in a rotating layer. *The order parameters are the amplitudes of the dominant velocity modes when the frequency of oscillation is small* (cf. Ref. 6). The evolution equations for the amplitudes of these modes are the center-manifold reduced equations. A linear local transformation of variables reduces the system to a normal canonical form near a codimension-2 bifurcation.

The problem of the entrainment of an open system sustaining a Hopf's instability by means of external periodic perturbations has been treated in the mathematical literature.[11] The cases considered do not present, however, a separation of relaxation-time scales indicating the enslaving of fast variables. The results were more recently specialized for the Brusselator.[12,13]

The aim of this work is to extend the theory to the case in which there is a separation of time scales at criticality and to provide a c.m. reduction of the perturbative vector field. We shall prove that the projection of the perturbative vector field on the c.m. is responsible for the entrainment in a GL regime.

The inherent properties of the Poincaré map associated to the GL potential are worked out making use of sensitivity analysis techniques. The first step is to find the fundamental Green's function sensitivity matrix which determines the propagation of perturbations along the c.m. The calculations are considerably simplified by observing that the c.m. reduction determines scaling relations among the matrix coefficients. An independent set of coefficients determines the GL potential functional and the Poincaré map.

The sensitivity information obtained in Secs. II and III plays a crucial role in the calculation of the entrainment regions in the amplitude-frequency space for the perturbation as shown in Secs. IV and V.

These regions are obtained by first projecting the perturbation on the c.m., finding the reduced equations for the slowly varying order parameters incorporating the contribution of the perturbation term and finally making a stability analysis of the periodic solutions. The results are then applied to a system which presents a separation of time scales and also, for the sake of testing the results, to the Brusselator which does not present a statistical enslaving of fast variables (since it involves only one time scale at criticality).

In the case of a single time scale, the results of the c.m. reduction are tested *vis-a-vis* previous treatments exhibiting very good agreement. It is worth emphasizing that, regardless of the external frequency, no entrainment will occur if the projection on the c.m. is zero. This statement will be illustrated in Sec. V.

In Sec. VI, a realistic experiment is suggested. We consider a continuous stirred-flow tank reactor in which two open reactive systems displaying, respectively, a subcritical and a supercritical Hopf bifurcation are coupled. It has been established that one oscillator entrains the other under suitable input concentrations. As demonstrated in this work, the inequalities determining the entrainment region can be verified directly. Each oscillator has to be considered separately, subject to the input conditions which yield the entrainment when both systems are coupled.

The frequency of the entraining oscillator is dependent on the scaling parameter which measures the departure of the entrained oscillator from marginal stability. The critical inherent frequencies for each oscillator should be determined separately and, in addition, the dependence of the frequency for the entraining oscillator on the flow rate must be found experimentally.

II. POINCARÉ MAP AT GL REGIMES

The reduction of a dynamical system to the Poincaré normal form determines the fast (subordinated) degrees of freedom and the order parameters.[8,9] In what follows we shall assume that such a reduction has already been performed and therefore, that the Jacobian matrix at criticality, denoted $\underline{J}_c$ is in Jordan (block diagonal and irreducible) normal form. Thus the type of unfolding of a dissipative structure can be inferred by direct inspection of $\underline{J}_c$ (cf. Refs. 1 and 9). In the case of a critical GL regime, the general evolution equations corresponding to a Hopf bifurcation unfolding are

$$\dot{Z}=\lambda(b)Z+F(Z,\bar{Z},\mathbf{Y},b),\quad Z\in\mathbb{C} \tag{2.1}$$

$$\dot{\mathbf{Y}}=\underline{J}_c\mathbf{Y}+\mathbf{G}(Z,\bar{Z},\mathbf{Y},b)\,, \tag{2.2}$$

where λ and $\bar{\lambda}$ are the complex conjugated eigenvalues which cross the imaginary axis at criticality, that is, for $b=b_c$. The other terms in Eqs. (2.1) and (2.2) are defined as follows. F is a complex valued function and $\mathbf{G}$ is a real valued vector field; they are given, respectively, by

$$F(Z,\bar{Z},\mathbf{Y},b)=\sum_j e_jN_j[\mathbf{Y}+2\,\mathrm{Re}(Z\mathbf{f}),b]\,, \tag{2.3}$$

$$\mathbf{G}(Z,\bar{Z},\mathbf{Y},b)=\mathbf{N}[\mathbf{Y}+2\,\mathrm{Re}(Z\mathbf{f}),b]-2\,\mathrm{Re}(F\mathbf{f})\,. \tag{2.4}$$

The vectors $\mathbf{e}$ and $\mathbf{f}$ are, respectively, the eigenvectors of $\underline{J}_c$ and its transpose given by the relations

$$\underline{J}_c\mathbf{f}=\lambda_c\mathbf{f},\quad \underline{J}_c\mathbf{e}=\bar{\lambda}_c\mathbf{e},\quad \lambda(b_c)=\lambda_c\,. \tag{2.5}$$

The vector field $\mathbf{N}(\mathbf{X},b)$ represents the nonlinear part of the original dynamical system: $\nabla_{\mathbf{X}}N_j\,|_{ss}=0$ for every component of $\mathbf{N}$. Therefore,

$$\dot{\mathbf{X}}=\tilde{\underline{J}}\mathbf{X}+\mathbf{N}(\mathbf{X},b)\,. \tag{2.6}$$

As we switch to the $(Z,\mathbf{Y})$ representation, the Jacobian matrix $\tilde{\underline{J}}_c$ transforms into $\underline{J}_c$. The phase-space vector $\mathbf{X}$ admits a unique decomposition

$$\mathbf{X}=\mathbf{Y}+2\,\mathrm{Re}(Z\mathbf{f})\,. \tag{2.7}$$

The complex variable Z is the coordinate giving the projection onto the slowly relaxing eigenmodes and the components of the vector $\mathbf{Y}$ are the fast relaxing degrees of freedom. The subordination of the evolution of $\mathbf{Y}$ [given in Eq. (2.2)] to the motion in the order-parameter space as given by Eq. (2.1) is determined by the analytic c.m. expansion

$$Y_j=\tilde{Y}_j(Z,\bar{Z},b)=\sum_{i+r+k=2}^{\infty}y_{i,r,k,j}\frac{Z^i\bar{Z}^rb^k}{i!r!k!}\,. \tag{2.8}$$

At criticality, we can obtain recursively the c.m. coefficients from the relations (the reader interested in such a computation may find details in Ref. 5)

$$\partial_Z\tilde{Y}_j^0=\partial_{\bar{Z}}\tilde{Y}_j^0=0\,. \tag{2.9}$$

The superindex 0 denotes the fact that the derivatives are evaluated at $Z=0$. The operators ∂_Z and $\partial_{\bar{Z}}$ are defined by

$$\partial_Z=\frac{1}{2}\left[\frac{\partial}{\partial v}-i\frac{\partial}{\partial w}\right],\quad \partial_{\bar{Z}}=\frac{1}{2}\left[\frac{\partial}{\partial v}+i\frac{\partial}{\partial w}\right],\quad Z=v+iw\,. \tag{2.10}$$

Finally, we have

$$\begin{aligned}\dot{\tilde{Y}}_j&=(\partial_z\tilde{Y}_j)[\lambda Z+F(Z,\bar{Z},\tilde{\mathbf{Y}},b)]\\&\quad+(\partial_{\bar{Z}}\tilde{Y}_j)[\bar{\lambda}\bar{Z}+\bar{F}(Z,\bar{Z},\tilde{\mathbf{Y}},b)]\\&=(\underline{J}_c\tilde{\mathbf{Y}})_j+\mathbf{G}_j(Z,\bar{Z},\tilde{\mathbf{Y}},b)\,.\end{aligned} \tag{2.11}$$

The system admits a potential functional[1,3] given by the function

$$P(Z)=-\frac{1}{2}\left|\frac{b-b_c}{b_c}\right|\,|Z|^2+(u/4)\,|Z|^4\,, \tag{2.12}$$

$$|Z|=(v^2+w^2)^{1/2}\ . \tag{2.13}$$

We denote by A the observable corresponding to the amplitude of the bifurcating solution which is assumed to emerge for $b>b_c$. The following sensitivity expansion holds (cf. Refs. 1 and 2):

$$b=b_c+\sum_{j=2}^{\infty} s_j \frac{A^j}{j!}\ . \tag{2.14}$$

The expansion does not contain a linear term, this fact follows from Hopf's theory.[2,3]

The s_j's are elementary sensitivity coefficients.[14–16] s_r, $r=2,3,4,\ldots$, can be interpreted as the rth derivative of the control parameter with respect to the observable evaluated at criticality:

$$s_r=\left[\frac{\partial^r b}{\partial A^r}\right]^0\ . \tag{2.15}$$

Therefore, we can conclude that within the mean-field approximation, the GL coefficient u can be written in terms of the sensitivity coefficient s as

$$\frac{\partial P(Z)}{\partial |Z|}(Z=A)=0 \rightarrow A=\left[\frac{b-b_c}{b_c u}\right]^{1/2}, \tag{2.16}$$

$$u=\tfrac{1}{2}b_c^{-1}s_2\ . \tag{2.17}$$

We shall now examine the Poincaré map using the fact that the limit cycle lies in the c.m. Consider a fixed point $\mathbf{p}_0$ belonging to the limit cycle $\mathbf{p}_A(t)$:

$$\mathbf{p}_0=\mathbf{p}_A(t_0)\ . \tag{2.18}$$

Let $\mathbf{n}$ denote a vector in phase space localized at $\mathbf{p}_0$ and perpendicular to the direction $\dot{\mathbf{p}}(t_0)$. This vector will be chosen in the following way. Let

$$P_0=\left[\mathbf{p}_0+\sum_j x_j \mathbf{g}_j\right] \tag{2.19}$$

denote the plane containing $\mathbf{p}_0$ and normal to $\dot{\mathbf{p}}(t_0)$, then we can consider the Jacobian matrix of the Poincaré map L defined on P_0 with respect to the variables x_j's. We shall denote this operator by $\underline{\partial}L$. We now choose $\mathbf{n}$ as the eigenvector of $\underline{\partial}L$ tangent at $\mathbf{p}_0$ to the intersection of P_0 with the c.m. (cf. Refs. 6 and 7):

$$\underline{\partial}L\,\mathbf{n}=(\exp[q(A)T(A)])\mathbf{n}\ . \tag{2.20}$$

Here $T(A)$ is the period of $\mathbf{p}_A(t)$ and $q(A)$ is the Floquet exponent associated to $\mathbf{n}$. This Floquet exponent can be expressed in terms of u since

$$q(A)=\tfrac{1}{2}M(b_c)A^2 s_2\ , \tag{2.21}$$

$$M(b_c)=-2\frac{d}{db}(\mathrm{Re}\lambda)(b=b_c)\neq 0\ . \tag{2.22}$$

The statement contained in expression (2.22) is one of Hopf's hypotheses. For a fixed small amplitude we can write, combining Eqs. (2.17) and (2.20)—(2.22),

$$\underline{\partial}L\,\mathbf{n}=\left[1+\frac{2\pi u b_c M(b_c)A^2}{|\mathrm{Im}\lambda_c|}\right]\mathbf{n}\ . \tag{2.23}$$

(The exponential has been expanded in terms of powers of A.) The Poincaré map is a contraction and the limit cycle, asymptotically stable when the bifurcation is supercritical, that is, when

$$\mathrm{sgn}[M(b_c)]\neq \mathrm{sgn}(u)\ . \tag{2.24}$$

This relation was obtained from Eq. (2.23) and it is in accord with previous findings.[6,7]

The aim of the following section is to make use of relations (2.17) and (2.21) to obtain the coefficient u from sensitivity information.

III. GREEN'S-FUNCTION SENSITIVITY ANALYSIS UNDER GL REGIMES

The fundamental sensitivity propagators along the c.m. are the Green's-function coefficients[14–16]

$$G_{ij}(t,t')=\frac{\partial Y_i(t)}{\partial Y_j(t')},\quad i,j=1,2,\ldots\ . \tag{3.1}$$

The generic coefficient G_{ij} determines the instantaneous change at time t on the subordinated variable Y_i caused by a perturbation at time t' in Y_j. However, in the asymptotic c.m. description, the functional dependence of the subordinated modes on the order parameters, as given by Eq. (2.8), determines the scaling relations among the G_{ij}'s. The asymptotic equations follow:

$$G_{ij}(t,t')=[\partial_z \tilde{Y}_i(t)][\partial_z \tilde{Y}_j(t')]^{-1}G_{zz}(t,t') + [\partial_{\bar z} \tilde{Y}_i(t)][\partial_{\bar z} \tilde{Y}_j(t')]^{-1}G_{\bar z\bar z}(t,t')\ . \tag{3.2}$$

The time scales for which that relation is valid are

$$t\geq t'\geq \operatorname*{Supreme}_j(|\mathrm{Re}\lambda_j|^{-1})\ . \tag{3.3}$$

The λ_j's are the eigenvalues of $\underline{J}_c$ different from λ and $\bar\lambda$. The order-parameter sensitivities are defined as

$$G_{zz}(t,t')=\frac{\partial Z(t)}{\partial Z(t')}=\frac{[\lambda Z+F(Z,\bar Z,\tilde{\mathbf{Y}},b_c)](t)}{[\lambda Z+F(Z,\bar Z,\tilde{\mathbf{Y}},b_c)](t')}\ , \tag{3.4}$$

$$G_{\bar z\bar z}(t,t')=\frac{[\bar\lambda \bar Z+\bar F(Z,\bar Z,\tilde{\mathbf{Y}},b_c)](t)}{[\bar\lambda \bar Z+\bar F(Z,\bar Z,\tilde{\mathbf{Y}},b_c)](t')}\ . \tag{3.5}$$

In order to apply the results obtained in the preceding section to derive u from sensitivity information involving only the observables, we shall introduce a representation of the phase space vector $\mathbf{X}$ in hypercyclindrical coordinates:

$$v=r\cos\theta,\quad w=r\sin\theta,\quad Y_j=Y_j\ . \tag{3.6}$$

The eigenvalue of the Jacobian matrix in the hypercylindrical framework calculated at the limit cycle corresponding to the coordinate r is $q=q(A)$ and the one corresponding to the phase coordinate is 1.

Thus, the subordination of the Y_j's to r can be obtained in the adiabatic elimination limit by setting $\dot{\mathbf{Y}}=\mathbf{0}$ (see for example, Ref. 3), we get

$$G_{ij}(t,t')=\left[\frac{\partial}{\partial r}\tilde{Y}_i(t)\right]\left[\frac{\partial}{\partial r}\tilde{Y}_j(t')\right]^{-1}H(t-t')e^{q(A)(t-t')}\,, \tag{3.7}$$

where H is the Heaviside unit-step function.

The following alternative formula indicates that the c.m. expansion and the GL potential determine the sensitivity coefficients

$$G_{ij}(t,t')=\left[\frac{\partial}{\partial r}\tilde{Y}_i(t)(r=A)\right]\left[\frac{\partial}{\partial r}\tilde{Y}_j(t')(r=A)\right]^{-1}\times H(t-t')e^{[M(b_c)ub_cA^2(t-t')]}\,. \tag{3.8}$$

This relation gives the sensitivity at a particular value of the observable. Thus, in the limiting case, when the system becomes insensitive to changes in the control parameter ($u\to\infty$), the autocorrelations $\langle Y_i(t)Y_j(t')\rangle$ become δ correlated and the corresponding power spectrum is a line spectrum (cf. Refs. 17 and 18).

The following system, already in Poincaré normal form, displays a Hopf bifurcation which can be adequately treated making use of the mean-field GL approach (this fact is proven in Ref. 1):

$$\begin{aligned}\dot{X}_1&=(2b-1)X_1-X_2+X_1X_3\,,\\ \dot{X}_2&=X_1+(2b-1)X_2+X_2X_3\,,\\ \dot{X}_3&=-bX_3-(X_1^2+X_2^2+X_3^2)\,.\end{aligned} \tag{3.9}$$

This system is obtained by truncating a model originally proposed by Hopf to describe the early onset of hydrodynamic turbulence.[1] The motion becomes quasiperiodic only when a torus bifurcates from the first limit cycle thus yielding a "Hopf upon Hopf" bifurcation. However, the second bifurcation does not follow a mean-field GL regime. We shall therefore concentrate in a neighborhood of the first critical point ($b_c=\frac{1}{2}$), the steady state being $r=X_3=0$. To a first approximation the c.m. can be obtained from the adiabatic elimination, that is from the implicit relation: $bX_3+(r^2+X_3^2)=0$.

The eigenvalue of the linearized Poincaré map along the $\mathbf{n}$ direction is given by the relation

$$\partial\underline{L}\mathbf{n}=(\exp\{[(3b-2)+(25b^2-36b+12)^{1/2}]\pi\})\mathbf{n}\,. \tag{3.10}$$

For a fixed amplitude, the contraction is given by

$$\partial\underline{L}\mathbf{n}=(\exp\{2\pi[-4A^2+O(A^2)]\})\mathbf{n}\,. \tag{3.11}$$

Since,

$$M(b_2)=-2\frac{d}{db}(2b-1)(b=b_c)=-4 \tag{3.12}$$

we get from relations (2.17) and (2.21), the sensitivity and the GL coefficients

$$s_2=u=2\,. \tag{3.13}$$

These equations show how the rate of contraction of the Poincaré map increases depending on the sensitivity of the observables to changes in the control parameter as derived from the GL potential.

The accuracy of the adiabatic elimination approximation to the c.m. reduction can be estimated making use of the sensitivity analysis. This procedure is based on the inspection of the singularities of the sensitivity derivatives

$$\left.\frac{\partial X_3}{\partial b}\right|_{\text{c.m.}} \text{ and } \left.\frac{\partial X_3}{\partial b}\right|_{\text{a.e.}}\,. \tag{3.14}$$

The latter derivative is evaluated by means of the adiabatic elimination. The example studied above will serve as an illustration. From relation (2.16) we obtain

$$\frac{\partial r}{\partial b}=\frac{A^{-1}}{2b_cu}\,. \tag{3.15}$$

The singularity occurs at the bifurcation point ($b=\frac{1}{2}$).

The adiabatic elimination (a.e.) yields

$$\begin{aligned}\left.\frac{\partial X_3}{\partial b}\right|_{\text{a.e.}}&=\frac{1}{2}\frac{\partial}{\partial b}\left\{-b+\left[b^2-4\left[\frac{b-b_c}{b_cu}\right]\right]^{1/2}\right\}\\ &=-\frac{1}{2}+\frac{1}{4}\frac{2b-4b_c^{-1}u^{-1}}{\left[b^2-4\left[\dfrac{b-b_c}{b_cu}\right]\right]^{1/2}}\,.\end{aligned} \tag{3.16}$$

Thus the singularity occurs at

$$b_0=\frac{4}{u}\left[1-\left[1-\frac{u}{4}\right]^{1/2}\right]>\frac{1}{2}\,. \tag{3.17}$$

However, substituting $u/4$ by x we get

$$\lim_{x\to 0}\frac{1}{x}[1-(1-X)^{1/2}]=\tfrac{1}{2} \tag{3.18}$$

or

$$\lim_{u\to 0}b_0=\tfrac{1}{2}\,. \tag{3.19}$$

We have that the adiabatic elimination becomes a better approximation as the GL coefficient u becomes smaller, that is, as the amplitude becomes more sensitive to changes in the control parameter [cf. Eq. (2.16)].

IV. PROJECTION OF PERIODIC PERTURBATIONS ON THE CENTER MANIFOLD

It has been established that a necessary condition for entrainment is the existence of a limit cycle sufficiently far from the marginal stability.[11–14] This condition is not sufficient as it will be demonstrated in this section; we also require that the projection of the perturbative vector field on the c.m. must be nonzero.

Thus, it is crucial to calculate the contribution of the perturbation to the right-hand side (rhs) of Eq. (2.1). The inherent frequency of the system defined by Eqs. (2.1)–(2.7) is

$$w_0=\mathrm{Im}\lambda_c\,. \tag{4.1}$$

We now add the perturbation $\mathbf{f}(t)$ to the rhs of Eq. (2.6).[11] The complex order parameter will be written as

$$Z=\epsilon z e^{iw_0 t},\quad \epsilon=\left[\frac{b-b_c}{b_c}\right]^{1/2}. \tag{4.2}$$

Then, the slowly varying complex variable z is subject to the restriction [cf. Eq. (2.16)]

$$\lim_{t\to\infty}\lim_{\|\mathbf{f}(t)\|\to 0}|z|=\frac{A}{\epsilon}=\frac{1}{\sqrt{u}}. \tag{4.3}$$

In order to determine the contribution of the perturbation to the order-parameter equation, we introduce first the scaling $\tau=\epsilon^2 t$ to account for the fact that z is a slowly varying parameter (cf. Ref. 8). The contribution will be denoted by $s(\tau)$. The projection of $\mathbf{f}(t)$ on the c.m. is given, according to relations (2.3) and (2.5) by the scalar product

$$[\mathbf{e},\mathbf{f}(t)]=Z(\mathbf{f}(t)). \tag{4.4}$$

This is the Z coordinate of $\mathbf{f}(t)$. By means of a c.m.-asymptotic perturbation analysis[1,5,6] we can find to which order in ϵ does $s(\tau)$ appear. Such an analysis requires the initial substitution

$$\mathbf{X}=\epsilon e^{iw_0 t}[2\,\mathrm{Re}z\,\mathbf{f}]+O(\epsilon^2) \tag{4.5}$$

in which the c.m. complex coordinate has $O(\epsilon)$ and all other coordinates are higher-order infinitesimals. To $O(\epsilon^3)$, we obtain the order-parameter equation with the contribution from the perturbation term. This reduced equation has the form (cf. Ref. 11)

$$\frac{dz}{d\tau}=\frac{b_c}{2}z-\left[\alpha+i\frac{(b_c-1)^2+7(b_c-2)^2+1}{12|b_c-1|^{3/2}}\right]\times|z|^2 z+s(\tau). \tag{4.6}$$

It follows from Eq. (4.3) that

$$\alpha=\frac{b_c u}{2}. \tag{4.7}$$

Given that Eq. (4.6) was obtained to $O(\epsilon^3)$ we can find the z coordinate of $\mathbf{f}(t)$:

$$z(\mathbf{f}(t))=\frac{Z(\mathbf{f}(t))}{\epsilon^3}e^{iw_0 t}. \tag{4.8}$$

In order to display the τ dependence of the perturbation term, we average over the inherent period T of the system ($T=2\pi/w_0$)

$$s(\tau)=\frac{1}{T}\int_{\tau/\epsilon^2}^{(\tau/\epsilon^2)+T}z(\mathbf{f}(t))dt=s_0 e^{i[(w-w_0)\epsilon^{-2}\tau+\phi_0]}, \tag{4.9}$$

where w is the external frequency ($w\sim w_0$) and ϕ_0 gives the phase displacement. Thus, if we represent the order parameter Z in terms of a radial and a phase component (denoted as, respectively, ρ and ϕ)

$$Z=\epsilon\rho e^{i(wt+\phi_0-\phi)}, \tag{4.10}$$

then the entrainment region in the (s_0,w) plane can be obtained by eliminating ϕ from the steady-state equations for ρ and ϕ:

$$\frac{b_c\rho}{2}-\left[\frac{b_c}{2}\right]u\rho^3+s_0\cos\phi=M_1(\rho,\phi)=0, \tag{4.11}$$

$$(w-w_0)\epsilon^{-2}+\frac{(b_c-1)^2+7(b_c-2)^2+1}{12|b_c-1|^{3/2}}\rho^2-\frac{s_0}{\rho}\sin\phi=M_2(\rho,\phi)=0. \tag{4.12}$$

From Eqs. (4.11) and (4.12) we obtain

$$\rho_s^2\left[\left[\frac{b_c}{2}\right]u\rho_s^2-\frac{b_c}{2}\right]^2+\rho_s^2\left[\left[\frac{(b_c-1)^2+7(b_c-2)^2+1}{12|b_c-1|^{3/2}}\right]\rho_s^2+\frac{w-w_0}{\epsilon^2}\right]^2-s_0^2=0. \tag{4.13}$$

From the stability analysis, it follows that s_0 and w should be chosen so that the root of (4.13) obeying

$$\frac{\partial}{\partial\rho}M_1+\frac{\partial}{\partial\phi}M_2=b_c[1-2u\rho_s^2]\le 0, \tag{4.14}$$

or, alternatively (without loss of generality, we assume $b_c>0$)

$$\rho_s\ge\frac{1}{\sqrt{2u}} \tag{4.15}$$

is stable. That is,

$$\det\begin{bmatrix}\dfrac{\partial M_1}{\partial\rho} & \dfrac{\partial M_1}{\partial\phi}\\[2ex] \dfrac{\partial M_2}{\partial\rho} & \dfrac{\partial M_2}{\partial\phi}\end{bmatrix}\ge 0. \tag{4.16}$$

Thus, for small s_0 we obtain

$$\left| \frac{w-w_0}{\epsilon^2} + \frac{(b_2-1)^2+7(b_c-2)^2+1}{12\,|\,b_c-1\,|^{3/2}} \frac{1}{u} \right| \le \left\{ u \left[1 + \left[\frac{\dfrac{(b_c-1)^2+7(b_c-2)^2+1}{12\,|\,b_c-1\,|^{3/2}}}{\dfrac{b_c}{2}u} \right]^2 \right] \right\}^{1/2} s_0 \ . \tag{4.17}$$

Thus, a system such that the amplitude A of the bifurcating limit cycle is relatively insensitive to changes in the control parameter b (i.e., u is very large) is more easily entrained [the region (s_0,w) giving a stable root ρ_s for Eq. (4.13) is larger] than if the amplitude is very sensitive ($u \approx 0$). This conclusion follows from two facts. (a) For large u we can write approximately,

$$\left| \frac{w-w_0}{\epsilon^2} \right| \le \sqrt{u}\, s_0 \ . \tag{4.18}$$

Considering again, Eq. (4.17) for $u \ll 1$, the term equal to u can be neglected in the expression under the radical and in the resulting equation. (b) The term in u^{-1} dominates over the term in $u^{-1/2}$.

V. EXAMPLES

We will now present the following examples.

(a) Any perturbation of the form

$$\mathbf{f}(t) = \begin{bmatrix} 0 \\ 0 \\ a\cos(wt) \end{bmatrix} \tag{5.1}$$

applied on the system given by Eqs. (3.9) ($b \sim \frac{1}{2}$) *will not* produce entrainment *regardless* of the values of a and w since the projection on the c.m. is zero.

(b) The entrainment region for the system given by Eqs. (3.9), ($b \sim \frac{1}{2}$), and $w \sim w_0 = 1$ is given by the inequality

$$\left| \frac{w-1}{2b-1} + \frac{17}{24} \left[\frac{1}{2} \right]^{-3/2} \right| \le \left[2 + \frac{2^6 \times 17^2}{12^2} \right]^{1/2} s_0 \ . \tag{5.2}$$

Observation. Notice that as the system departs from marginal stability, that is, the difference $(b-b_c)$ grows, then the range of frequencies in the entraining region also increases. This general principle can be observed by direct inspection of Eq. (4.7).

(c) The entrainment problem for the Brusselator has been studied by means of the reductive perturbation method.[12,13] This system presents only 2 degrees of freedom and therefore, in its GL regime, there are no enslaved fast-relaxing modes. The rate equations in reduced variables are given by (cf. Refs. 12 and 13)

$$\begin{aligned} \frac{d}{dt}X_1 &= A_0^2 X_2 + \epsilon^2 b_c X_1 + h(X_1,X_2) \ , \\ \frac{d}{dt}X_2 &= -X_1 \ , \end{aligned} \tag{5.3}$$

$$\begin{aligned} h(X_1,X_2) &= \left[\frac{1-A_0^2}{A_0} \right] X_1^2 + 2AX_1X_2 \\ &\quad + X_1^2(X_2-X_1) + \frac{\epsilon^2 b_c}{A_0} X_1^2 \ , \end{aligned} \tag{5.4}$$

$$b_c = 1 + A_0^2, \quad \lambda_c = iA_0 \ ,$$

$$\text{steady state: } X_1 = X_2 = 0 \ , \tag{5.5}$$

$$w_0 = A_0, T = \frac{2\pi}{A_0} \ .$$

The last term in the rhs of Eq. (5.4) has been omitted in Ref. 12. This term can only be neglected if the concentration of the external species A_0 is such that $|\,A_0 - 1\,| \gg 0$.

The perturbation analysis yields (cf. Ref. 12)

$$\frac{d}{d\tau}z = \frac{1+A_0^2}{2} z - \left[\frac{A_0^2+2}{2A_0^2} + i\frac{4A_0^2-7A_0^2+4}{6A_0^3} \right] |\,z\,|^2 z \ . \tag{5.6}$$

Thus, from Eq. (4.6) we can obtain the GL coefficient u for the Brusselator:

$$u = \frac{1}{(b_c-1)} \left[1 + \frac{1}{b_c} \right] = \frac{1}{A_0^2} \left[1 + \frac{1}{1+A_0^2} \right] \ . \tag{5.7}$$

Thus, combining Eqs. (4.5), (5.5), and (5.7) we obtain Eq. (5.6).

The entrainment region is obtained from Eqs. (4.17), (5.5), and (5.7), and it reads as

$$\left| \frac{w-A_0}{\epsilon^2} + \frac{4A_0^4-7A_0^2+4}{6A_0^3} \frac{1}{\dfrac{1}{A_0^2}\left[1 + \dfrac{1}{1+A_0^2} \right]} \right| \le \left[\frac{1}{A_0^2} \left[1 + \frac{1}{1+A_0^2} \right] \left[1 + \frac{(4A_0^4-7A_0^2+4)^2}{6A_0^3 \left[\dfrac{1+A_0^2}{2} \right]^2} \right] \right]^{1/2} S_0 \ . \tag{5.8}$$

This last relation is exactly the same as that obtained in Ref. 12.

VI. ENTRAINED OSCILLATIONS IN A CONTINUOUS STIRRED FLOW TANK REACTOR OPERATING FAR FROM THERMODYNAMIC EQUILIBRIUM

In recent years, there has been considerable interest in the dynamics arising when two oscillatory open reactive systems are linked through an internal species which corresponds to an internal degree of freedom for both systems. In particular (see, for example, Refs. 19 and 20), an open oscillatory system (I) with input concentrations $[ClO_2^-]$, $[I^-]$, $[H^+]$ has been coupled to another system (II) with input concentrations $[BrO_3^-]$, $[I^-]$, $[H^+]$ in a continuous stirred-flow tank reactor (CSTR).

Each of the two coupled systems is a relatively simple example of an uncatalyzed inorganic oscillator. We shall regard the flow rate

$$\tau_r^{-1}=b, \quad \tau_r=\text{residence time of the CSTR} \tag{6.1}$$

as the bifurcation control parameter. The other control parameters are the input concentrations for the external species (in large excess with respect to the internal species I_2 and IO_3^-), $[ClO_2^-]_0$, $[I^-]_0$, $[BrO_3^-]_0$, $[H^+]_0$, and the temperature.

The internal degrees of freedom are the concentrations of stable intermediates: $[I_2]$, $[IO_3^-]$ (cf. Ref. 21). It has been demonstrated[21] that there exists a parameter region for system (I) in which a Hopf bifurcation occurs. The specification of this region is irrelevant for our present purposes. The frequency of oscillations increases as the flow rate grows past its critical value $[\tau_r^c(\mathrm{I})]^{-1}=b_{c\mathrm{I}}$.

The kinetics for system (II) also displays a Hopf bifurcation but the frequency of the oscillations decreases as the flow rate increases $b>[\tau_r^c(\mathrm{II})]^{-1}$ [see Ref. 22].

In both cases, of course, the critical value for the flow rate depends on the input concentrations. It has been experimentally confirmed[20] that for the following selection of control parameters,

$$T=25\,^\circ\mathrm{C},\ [I^-]_0=4\times10^{-4}M\ [BrO_3^-]_0=2.5\times10^{-3}M\ , \\ [ClO_2^-]_0=10^{-4}M\ [H^+]_0=1.5M\ , \tag{6.2}$$

and for a flow rate in the range 16.4×10^{-3}–18.3×10^{-3} sec^{-1}, the CSTR coupling both systems displays compound oscillations for the internal species with a range of frequencies very close to the oscillator (II). It is also observed that the frequency of the compound oscillations decreases as τ_r^{-1} increases, also in accord with the dynamics for oscillator (II).

Systems (I) and (II) are linked through the internal variable $[I_2]$, however, the oscillations in (II) have large amplitudes and those of (I) small amplitudes. A direct inspection of the inequality (4.17) indicates that the larger amplitude oscillator will have a wider range of frequencies for entraining than the smaller amplitude oscillator. Thus, the effect of coupling both oscillators in the same CSTR is equivalent to introducing a periodic perturbation flow term in the rate equations for species I_2 in system (I). This perturbation is responsible for the observed entrainment of system (I) by system (II). It is worth noticing at this point that the external entraining frequency ω_{II}, with respect to oscillator (I), cannot be varied independently of the reduced scaling parameter

$$\epsilon=\left|\frac{\tau_r^{-1}-[\tau_r^c(\mathrm{I})]^{-1}}{[\tau_r^c(\mathrm{I})]^{-1}}\right| ,$$

which measures the departure of system (I) from marginal stability regime occurring at $\tau_r=\tau_r^c(\mathrm{I})$. That is so since ω_{II} is a function of the flow rate. (Numerical values for this dependence are given in Ref. 22.)

We can propose the following entraining inequality for the CSTR [cf. Eq. (4.17)]:

$$\left|\frac{w_{\mathrm{II}}-w_{0_{\mathrm{I}}}}{\left|\dfrac{\tau_r^{-1}-[\tau_r^c(\mathrm{I})]^{-1}}{[\tau_r^c(\mathrm{I})]^{-1}}\right|}+\frac{M[\tau_r^c(\mathrm{I})]}{u_{\mathrm{I}}}\right|\le\left[u_{\mathrm{I}}\left\{1+\frac{2\tau_r^c(\mathrm{I})M[\tau_r^c(\mathrm{I})]}{u_{\mathrm{I}}}\right\}\right]^{1/2}S_{0_{\mathrm{II}}}\ , \tag{6.3}$$

$$M(\tau_r^c(\mathrm{I}))=\frac{\{[\tau_r^c(\mathrm{I})]^{-1}-1\}^2+7\{[\tau_r^c(\mathrm{I})]^{-1}-2\}^2+1}{12\,|\,[\tau_r^c(\mathrm{I})]^{-1}-1\,|^{3/2}}\ . \tag{6.4}$$

In order to confirm this formula, one needs to examine separately each oscillator at the input concentrations that correspond to entrainment when the oscillators are coupled, that is, at the selection of control parameters given by Eq. (6.2) varying the flow rate.

Thus, one finds the critical values $\tau_r^c(\mathrm{I})$, $\tau_r^c(\mathrm{II})$, and $w_{0_{\mathrm{I}}}$. The experiments hitherto performed consider bifurcation points *outside* the entrainment domain and different for each oscillator. The calculation of u_{I} involves measuring the second variation of the flow rate with respect to the amplitude of the oscillations as given by relations (2.14)–(2.17).

The final step would be to vary the τ_r and calculate w_{II} versus τ_r [always under the external constraint given by Eq. (6.2)]. These values can be used to confirm the validity of relations (6.3) and (6.4).

VII. CONCLUSION

The entrainment of a dissipative reactive system in a Ginzburg-Landau critical regime by an external periodic perturbation is determined by the projection of this perturbation on the center manifold of the system. The center manifold coordinates are the order parameters. The ranges for the amplitude and frequency of the perturbations depend on three properties inherent to the system.

These properties are obtained from the Ginzburg-Landau potential functional and from the center manifold equation, and are the following.

(i) The elimination of the fast-relaxing degrees of freedom leading to scaling relations in the sensitivity coefficients.

(ii) The departure of the system from marginal stability measured by the scaling parameter ϵ.

(iii) The rates of contraction of the Poincaré iterative map along the eigendirections tangent to the center manifold and perpendicular to the limit cycle. These rates yield the Ginzburg-Landau coefficient u which defines the amplitude sensitivity to changes in the control parameter for bifurcation.

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