Stochastic Analysis of Limit Cycle Behavior in Spatially Extended Systems

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The statistical properties of a one-dimensional reaction-diffusion system undergoing a Hopf bifurcation are studied using the master equation approach. The analysis reveals nontrivial interferences between macroscopic dynamics and mesoscopic local fluctuations that eventually wipe out any trace of homogeneous oscillations, even though the latter are asymptotically stable solutions of the deterministic equations. [S0031-9007(96)00867-8]

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There is ample experimental evidence of sustained oscillations in reaction diffusion systems [1]. Most of the reported experimental observations, however, are restricted to well stirred systems. In the absence of stirring, small spatial desynchronizations start to appear that eventually invade the reaction volume as a whole, long before the reactants are exhausted. As we will show in this paper, the desyncronization mechanism is closely related to the anomalous behavior of local fluctuations, in conjunction with the spatial dimensionality of the system.

In the absence of fluctuations, the dynamics of spatially distributed systems with local diffusion coupling and evolving near a supercritical Hopf bifurcation can be cast into a complex Ginzburg-Landau equation (CGLE) which represents the associated normal form [2]. Ordinarily, the effect of fluctuations is accounted for at the level of Langevin equations, where local random white noise terms (in both time and space) are added heuristically to the macroscopic evolution equation [3]. The validity of such a Langevin formulation rests on the fundamental assumption that the nonlocal diffusional noise terms are negligible near a bifurcation point. From a theoretical foreground, this hypothesis can be justified only if the system behaves in a perfectly coherent manner [4], which is precisely what we want to check. For this reason, we resort here to a description based on the master equation (ME) [5], in which information on the elementary processes going on at the molecular level is introduced, without referring explicitly to the macroscopic behavior of the system. It has been shown in the past that in a variety of situations, including the case of a Hopf bifurcation in 0D [6], results based on ME are in excellent agreement with those obtained from the more fundamental molecular dynamic simulations of reactive fluids (for a review, see [7] and references therein). Furthermore, the ME can be easily simulated on digital computers, running several orders of magnitude faster than the corresponding molecular dynamic simulation [8].

As stated above, the ME approach requires the explicit knowledge of each elementary step. We choose the so-called "Brusselator" model which has been studied extensively in the past [9]. The normal form equation around the unstable fixed point assumes the explicit form [10]

$$\frac{\partial z}{\partial t} = \left(\frac{b - b_0}{2} + ia\right)z - \left(\frac{a^2 + 2}{2a^2} + \frac{i}{2}\,\frac{4a^4 - 7a^2 + 4}{3a^3}\right)|z|^2 z + \frac{1}{2}[(D_1 + D_2) + ia(D_2 - D_1)]\frac{\partial^2 z}{\partial r^2}, \quad (1)$$

where a, b denote the control parameters and D_1, D_2 are the diffusion coefficients of the two intermediate variables of the original model. The complex order parameter $z(\mathbf{r}, t)$ accounts for both the (local) amplitude and phase of the oscillation, and $b - b_0$ is the distance from criticality. The threshold b_m for the Hopf bifurcation is given by linear stability analysis [11]

$$b_m = (a^2 + 1) + q_m^2(D_1 + D_2), \quad (2a)$$

where q_m is the wave number of the inhomogeneous perturbation around the uniform steady state. In what follows, we shall limit ourselves to the study of a one-dimensional system of length L subjected to zero flux boundary conditions. In this case, the wave number q_m can only take discrete values given by

$$q_m = \frac{m\pi}{L}, \qquad m = 0, 1, 2, \ldots . \quad (2b)$$

In addition to a homogeneous limit cycle, corresponding to the uniform mode $m = 0$, Eq. (1) can lead to a variety of secondary and higher order instabilities culminating to spatiotemporal chaos [12]. Analysis of the numerical solutions of the CGLE has been carried out to characterize the various spatiotemporal chaotic regimes (phase and defect turbulence) [13–15]. The threshold at the onset of these phenomena is given by the Benjamin-Feir condition [16]. Using the explicit values of the normal form coefficients in Eq. (1), we easily verify that the homogeneous limit cycle is stable toward inhomogeneous perturbations as long as [12]

$$\frac{D_1 - D_2}{D_1 + D_2}\,\frac{4a^4 - 7a^2 + 4}{3(a^2 + 2)} < 1 . \quad (3)$$

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In this range the uniform mode $m = 0$ in Eq. (2) bifurcates as a stable (supercritical) solution, whereas the modes $m \neq 0$ lead to unstable branches as they bifurcate for values b_m of the control parameter b higher than b_0. In what follows, we choose $D_1 = D_2 = D, a = 2, b = 5.2$. Condition (3) is then always satisfied: The uniform limit cycle is stable and, as D/L^2 is decreased, an increasing number of linearly unstable spatial modes becomes excited.

In an unstirred reactor, the evolution of the system at the stochastic level can be described through the so-called "multivariate master equation," where, in addition to the local chemical contributions, treated as a birth and death process, the transport of the reactants is also included through a random walk process [11]. The theoretical analysis of this equation proves to be extremely involved for the case of interest, i.e., above the bifurcation point [17]. We thus resort to the numerical simulation of the ME, using the technique described in Ref. [8]. A uniform initial state, corresponding to the unstable reference state, is chosen in all reported simulation. The average number of particles per cell is about 500, leading to a relatively small noise amplitude of about 5% [18].

The space-time plots of Fig. 1 represent the result of such a stochastic simulation for two different values of the ratio D/L^2 in Eq. (2): (a) The system length L is small enough ($D/L^2 = 2.56 \times 10^{-2}$) so that only the mode $m = 0$ is excited, i.e. $b_0 < b < b_1$. (b) The system length is 8 times larger than in the previous case ($D/L^2 = 4 \times 10^{-4}$) so that, in addition to the uniform mode $m = 0$, five inhomogeneous modes are also excited, i.e., $b_5 < b < b_6$.

As can be seen, in case (a) the system oscillates synchronously in a spatially coherent fashion, whereas in case (b) different parts of the system oscillate with markedly different phases. The statistical properties of the system follow the above observations. For instance, the density of states in phase space in case (a) remains practically the same as in a 0D case (craterlike distribution), whereas in case (b) it takes the form of a broad one humped distribution centered on the unstable state. This can be further quantified through the analysis of the space-time autocorrelation function, $C(r,t) = \langle \delta c(r,t) \delta c(0,0) \rangle$, of a concentration variable. In Fig. 2 we show the real part of the (discrete) spatial Fourier transform $C_m(t)$ of this function, for both case (a) and (b). Besides the uniform mode ($m = 0$), two nonuniform modes ($m = 1$ and $m = 2$) are also depicted. For each mode, $C_m(t)$ is normalized to unity at $t = 0$. For the case (a), shown in Fig. 2(a), the inhomogeneous modes become rapidly uncorrelated, whereas the homogeneous one shows high persistence, indicating the robustness of the macroscopic limit cycle toward inhomogeneous fluctuations. The situation is completely different for the case (b), shown in Fig. 2(b). Not only does the persistence time of the uniform mode decrease dramatically, but this mode behaves essentially the same as the inhomogeneous ones.

These results clearly indicate that in a one-dimensional system the homogeneous limit cycle is wiped out through the interference with inhomogeneous fluctuations, when a sufficient number of inhomogeneous modes are excited [five in the case of Fig. 2(b)]. For a given system

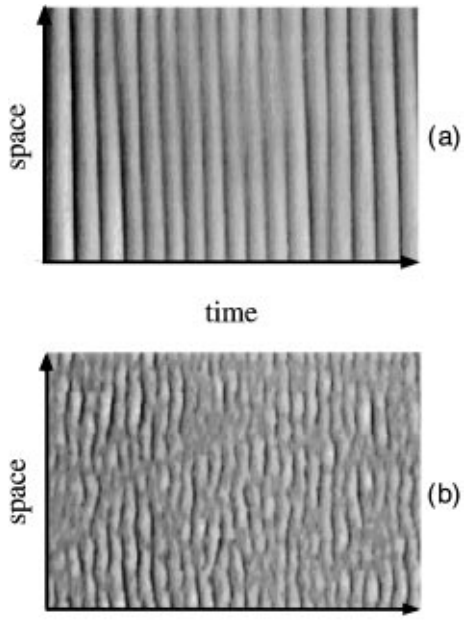

FIG. 1. Space-time plot of a concentration variable for the one-dimensional Brusselator. Dark and bright regions indicate high and low concentrations, respectively, with $a = 2, b = 5.2, D/L^2 = 256 \times 10^{-2}$ for the case (a) and $D/L^2 = 4 \times 10^{-4}$ for the case (b).

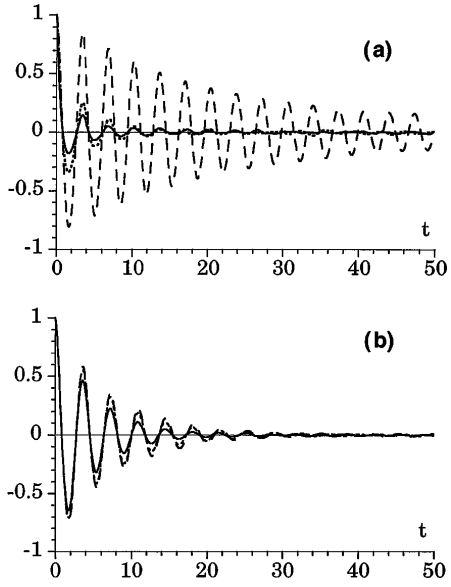

FIG. 2. Time dependence of the autocorrelation function of the homogeneous (dashed line) and the first two inhomogeneous modes in the one-dimensional Brusselator (dotted and full lines). Parameter values are as in Fig. 1.

size the onset of this phenomenon depends on the local noise amplitude as determined by the average number of particles per cell. Detailed numerical studies show that for the choice of 500 particles per cell, made above, this conclusion remains valid even if a single inhomogeneous mode is excited, except that, as the system length L is decreased, the spatial size of coherent oscillation is increased. For instance, in a situation where only the modes $m = 0$ and $m = 1$ are excited, we first observe that the opposite ends of the medium start to oscillate out of phase. This is then followed by the spontaneous emergence of large phase domain patterns. As times goes on, however, both the size and location of these domains change in an apparently chaotic manner so that, upon a scaling factor, the situation remains basically the same as in case (b).

One possible mechanism at the origin of this behavior is suggested by the observation that a well chosen strong local perturbation, larger than macroscopic composition variables, can destabilize the homogeneous limit cycle and drive the system into a triggering wave propagation behavior [19]. Such a condition, however, is impossible to realize in a stochastic dynamics based on the master equation since, in this case, the noise is acting continuously on the system, and its amplitude barely exceeds 5% of that of macroscopic variables. Another possibility is provided by the Benjamin-Feir instability for parameter values leading to a "defect chaos" regime, observed in detailed numerical studies of a CGLE [13] (see also [20]). Not only is the CGLE used in these studies equivalent to Eq. (1), but their space-time plot of the defect chaos regime (Fig. 1 of Ref. [13]) is surprisingly similar to our space-time plot (Fig. 1). The parameter values which we have chosen to simulate the master equation, however, manifestly preclude the above observations (recall that $D_1 = D_2$). Finally, the spontaneous breakdown of synchronization has also been noted in the context of the 1D CGLE with global coupling [21]. Again, in this case, the nature of the phenomenon is different since the global coupling indicates an instability of the phase-locked stationary solution.

In short, there seems to be no purely macroscopic analog of the behavior discovered in our simulations. The situation is all the more intriguing since results based on phenomenological Langevin equations, with strictly local additive noise terms, have always agreed with the predictions of purely macroscopic equations, at least in the presence of single attractors. Such a Langevin approach was first proposed by Dewel *et al.* [22,23] to emphasize the analogy between nonequilibrium cusp bifurcations and equilibrium critical phenomena. Later on, these authors extended the theory to the case of a Hopf bifurcation [3], and, since then, many others have used similar approaches to analyze specific features of nonequilibrium transitions [24].

Now, a Langevin equation can also be derived directly from the master equation [25], but here the noise term associated to diffusion is "nonlocal," i.e., it appears as the gradient of a white noise process [4]. A correct Langevin formation of reaction diffusion equations thus requires the presence of nonlocal diffusional noise terms, in addition to local noise terms associated to reactions. The thermodynamic formulation of the problem, based on Landau-Lifshitz fluctuations theory, leads precisely to the same result [26,27]. It is in such nonlocal diffusion terms that the origin of the behavior discovered in our simulations is to be sought.

The above results clearly imply that the validity of a Langevin CGLE, with strictly local additive noise terms, can only be guaranteed under conditions for which the diffusional noise terms can be neglected. On intuitive grounds, one expects that this will be the case if, on average, the gradient of fluctuations remains small, i.e., if nearby regions of the system fluctuate in a "coherent" fashion. In other words, diffusional noise terms become negligible if the system exhibits markedly coherent behavior, i.e., if the correlation functions are macroscopically long ranged. So far, such a result has been established only for systems evolving in the close vicinity of a cusp bifurcation point, where it has been shown that the stationary solution of the ME can be cast into the exponential of a "stochastic potential," identical to the Ginzburg-Landau functional [28]. The extension of the above analytic calculations to the case of a Hopf bifurcation seems extremely difficult. In fact, the very existence of a stochastic potential has been questioned in this case [29,30].

In conclusion, our simulations show that, as the number of linearly unstable spatial modes is increased, the correlation length gradually becomes smaller and the uniform limit cycle is wiped out by local fluctuations, even though it is macroscopically stable. This indicates that small wavelength processes remain strongly coupled to the large wavelength behavior of the system. We have here a clear indication of the intrusion of the microscopic dynamics into the macroscopic behavior, at least for the class of one-dimensional reaction-diffusion systems evolving in an ideal medium.

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