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Master equation approach to synchronization in diffusion-coupled nonlinear oscillators

William Vance and John Ross
Department of Chemistry, Stanford University, Stanford, California 94305

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We study the influence of internal fluctuations on phase synchronization in oscillatory reaction-diffusion systems through a master equation approach. In the limit of large system size, the probability density is analyzed by means of the eikonal approximation. This approximation yields a Hamilton-Jacobi equation for the stochastic potential, which may be reduced to coupled nonlinear diffusion equations for the phase of oscillation and (conjugate) ''momentum.'' We give explicit expressions for the coefficients of these equations in terms of averages over the deterministic periodic orbit. For one-dimensional systems, we obtain an explicit solution for the stationary stochastic potential: the width in phase, which is defined as the root mean square fluctuation in phase, characterizes the roughness of phase locking, and diverges with the system size L according to a power law $w \sim L^{\alpha}$, with $\alpha = 1/2$. To study higher-dimensional systems, we show that the eikonal approximations of the diffusion-coupled oscillator problem and the Kardar-Parisi-Zhang (KPZ) equation (in the limit of small noise intensity) are equivalent. The KPZ equation governs some forms of surface growth, and the height of a growing front corresponds to the phase (the 2π periodicity in phase is ignored) in the diffusion-coupled oscillator problem. From the equivalence, we obtain the result that spatially synchronized states may exist only in systems with a spatial dimension greater than or equal to 3; for dimensions 1 and 2, a ''rough'' state exists in which the width (in phase) diverges algebraically with the system size, $\alpha > 0$.

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I. INTRODUCTION

Much attention has been given to the problem of maintaining long-range order in systems governed by short-range interactions [1–4]. Of particular concern from chemical and biological points of view are large populations of self-oscillatory systems coupled through diffusion. These arise as models for oscillating chemical reactions [5,1,3], and for pacemakers in the description of cardiovascular, respiratory, and circadian rhythms [6,7]. Many ecological systems also exhibit population cycles, and are synchronized over large distances [8]. In the absence of noise, the evolution of these systems (for a slow spatial variation of the phase of the oscillators) may be described by an equation for the phase of oscillation—a nonlinear phase diffusion equation [5,1]. This equation has stable synchronous oscillatory solutions as well as a variety of other solutions, such as periodic wave trains, ''target patterns,'' and spiral waves [1,3,9]. It is of considerable interest to know whether such states are preserved in the presence of either internal or external noise (as the size of the system becomes arbitrarily large). For the case of a spatially synchronized state, this question was partially answered by Grinstein *et al.* [10], who argued that stable spatially coherent oscillations do not occur in d-dimensional, (external) noisy oscillatory systems with $d \leqslant 2$. They used the connection between the ''phase-only'' approximation of the noisy Ginzburg-Landau equation, which is used to model the onset of oscillations in spatially distributed systems, with the Kardar-Parisi-Zhang (KPZ) equation, which is a noise-driven nonlinear diffusion equation that governs some types of surface growth [11–13]. The phase of oscillators at a given spatial point corresponds to the height of a growing interface of the KPZ equation (ignoring the periodicity of the phase variable). Their simulations using (noiseless) cellular automata also suggest that the KPZ equation correctly describes the effects of internal fluctuations. Recent simulations [14] of the master equation for one-dimensional oscillatory reaction-diffusion systems near a Hopf bifurcation support these results. These simulations show that the homogeneous synchronized state, which is a stable solution for the deterministic system, is destroyed by internal fluctuations as the size of the system is increased.

We study the influence of internal fluctuations on synchronization in diffusively coupled oscillators at a mesoscopic level using a master equation approach. Our work makes explicit the connection between this description of oscillatory systems and the KPZ equation. Using a WKB approximation [15–19] of the probability density, and applying the method of multiple time scales, we derive a reduced set of (Hamiltonian) coupled nonlinear diffusion equations for the phase θ and the (conjugate) ''momentum.'' This procedure yields explicit expressions for the coefficients in terms of averages over the deterministic periodic orbit. Associated with these equations is a Hamilton-Jacobi equation for the leading term in the WKB approximation, which is the stochastic potential for the phase. For the stationary state of one-dimensional systems, we obtain an exact solution for the stochastic potential. For higher-dimensional systems we utilize the equivalence of the stochastic (phase) potential and the potential for the KPZ equation in the limit of weak noise. Using this correspondence we apply results from the extensive literature on kinetic roughening [13] (of growing interfaces) for the KPZ equation to the problem of the influence of internal fluctuations on spatial synchronization. (The influence of fluctuations on plane waves and other solutions to the nonlinear phase equation can also be studied using our reduced Hamilton-Jacobi equations with appropriate boundary conditions.)

The consequences for synchronization in reaction-diffusion systems follow from the above equivalence be-

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tween the eikonal approximations of the coupled oscillator and KPZ equations. The conditions under which coherence of a growing interface can be sustained were studied extensively for the KPZ equation. For dimensions less than or equal to a ''critical dimension'' $d_c = 2$, spatial synchronization cannot be maintained. A ''rough'' state exists in which fluctuations in the phase θ diverge algebraically with the system size L, $\langle [\theta(x) - \overline{\theta}]^2 \rangle^{1/2} \sim L^{\alpha}$, with $\alpha > 0$. That is, the difference in phase between two points separated by a distance L is typically of order L^{α} (the 2π periodicity of the phase is ignored). For a system with $d \geqslant 3$, a smooth spatially synchronized state exists ($\alpha < 0$ for a sufficiently small value of the coefficient of the nonlinear term of the phase-diffusion equation). In this case, the two-point correlations in phase decay asymptotically like $1/r^{d-2}$ in space and $1/t^{(d-2)/2}$ in time. In dimension $d = 3$, there is a phase transition in which a smooth state exists for values of the coupling strength g below a critical number g^* and a ''rough'' state exists for values greater than g^*. (The coupling strength g is a function of all the parameters in the phase-diffusion equation.)

In Sec. II we outline the master equation description of reaction-diffusion systems, and introduce the WKB approximation of the probability density. This leads to a classical Hamiltonian system, from which the leading order term in the WKB expansion, the stochastic potential or eikonal, may be obtained by integration of an associated Lagrangian along classical trajectories. In Sec. III we specialize to systems of oscillators weakly coupled through diffusion, and apply the method of multiple time scales to the above Hamiltonian system. The terms of leading order in this expansion produce a reduced Hamilton-Jacobi equation for two field variables: the phase and its conjugate ''momentum.'' In Sec. IV we derive solutions to this equation, and discuss the approximate solution to the full problem. Section V treats the equivalence of the reduced system to the eikonal approximation of the Kardar-Parisi-Zhang equation. We also quote some results for the KPZ equation that apply to the problem of phase synchronization of coupled oscillators. In Sec. VI we discuss our results, potential applications, and directions for further work.

II. MASTER EQUATION DESCRIPTION OF DIFFUSION-COUPLED OSCILLATORS

A. Reaction-diffusion master equation

Internal fluctuations in nonequilibrium systems are most simply treated by appealing to a mesoscopic level of description that regards reactions and diffusion as Markovian processes. Using jump processes to model reactions and a random walk to model diffusion, a reaction-diffusion master equation can be constructed [20,17]. In general, this equation is intractable; but with the introduction of a WKB approximation of the probability density [15–19], much progress can be made in obtaining solutions. We give a brief outline of this approach.

We divide the system into macroscopically small cells; each cell oscillates in time according to an homogeneous limit cycle solution, and exchanges matter with neighboring cells through diffusion. We assume a weak coupling, i.e., the diffusion terms are small compared to the reaction terms. This assumption may be interpreted as either small values for the diffusion coefficients or slow spatial variation of concentrations (with diffusion coefficients of ordinary magnitude). With this supposition, the structure of the limit cycle in individual cells is only slightly perturbed by neighboring cells. We take as variables the numbers of particles $\{X_{\mathbf{r}\alpha}\}$ within cells, and assume that they define a Markov process, where $\mathbf{r}$ labels the cell and α the species. These random variables change as a result of chemical reactions, which are modeled as jump Markov processes, and diffusion, which is modeled as a random walk between adjacent cells. This leads to a multivariate master equation for the probability distribution $P(\{X_{\mathbf{r}\alpha}\},t)$:

$$\begin{aligned}\frac{dP}{dt} = \sum_{\rho} \sum_{\mathbf{r}} & [W_{\rho}(\mathbf{X}_{\mathbf{r}} - \nu_{\rho} \to \mathbf{X}_{\mathbf{r}}) P(\{X_{\mathbf{r}\alpha} - \nu_{\rho\alpha}\}) \\ & - W_{\rho}(\mathbf{X}_{\mathbf{r}} \to \mathbf{X}_{\mathbf{r}} + \nu_{\rho}) P] + \epsilon \sum_{\alpha} \frac{\tilde{D}_{\alpha}}{2d} \sum_{\mathbf{r},\mathbf{a}} [(X_{\mathbf{r}\alpha} + 1) \\ & \times P(X_{\mathbf{r}\alpha} + 1, X_{(\mathbf{r}+\mathbf{a})\alpha} - 1) - X_{\mathbf{r}\alpha} P],\end{aligned} \tag{1}$$

where the transition probability per unit time for the ρth reaction is taken to be

$$W_{\rho}(\mathbf{X}_{\mathbf{r}} \to \mathbf{X}_{\mathbf{r}} + \nu_{\rho}) = k_{\rho} \Delta V^{1 - \Sigma_{\alpha} \overline{\nu}_{\rho\alpha}} \prod_{\alpha} \frac{(X_{\mathbf{r}\alpha})!}{(X_{\mathbf{r}\alpha} - \overline{\nu}_{\rho\alpha})!}. \tag{2}$$

The stoichiometric coefficient of X_{α} in the ρth chemical reaction is $\nu_{\rho\alpha}$; the order of the ρth reaction with respect to X_{α} is $\overline{\nu}_{\rho\alpha}$; the kinetic coefficient of the ρth reaction is k_{ρ}. The jump frequency $\epsilon \tilde{D}_{\alpha}$ of species α is related to Fick's diffusion coefficient ϵD_{α} by

$$\frac{\epsilon \tilde{D}_{\alpha}}{2d} l^2 \sim \epsilon D_{\alpha}, \tag{3}$$

where l is the characteristic length of a cell $l^d = \Delta V$. In Eq. (3) we assume that the diffusion term is a small perturbation, and scale the diffusion coefficient by a small parameter ϵ. Alternatively, for diffusion coefficients of ordinary magnitude, this assumption is equivalent to slow spatial variation of concentrations with a wavelength of order $\epsilon^{-1/2}$.

B. WKB approximation of the master equation

We consider ΔV as a large parameter, since a large number of particles are contained within a cell, and write P to leading order as an asymptotic WKB form

$$P(\{X_{\mathbf{r}\alpha}\},t) = K(x_{\mathbf{r}\alpha},t) e^{-\Delta V\, S(\{x_{\mathbf{r}\alpha}\},t)}, \tag{4}$$

where $x_{\mathbf{r}\alpha} = X_{\mathbf{r}\alpha}/\Delta V$ is the concentration of a species. In the following, we are concerned with S, the stochastic potential (also referred to as the nonequilibrium potential), and neglect the prefactor K. Substituting Eq. (4) into Eq. (1), to leading order we obtain the eikonal equation for the stochastic potential,

$$-\frac{\partial S}{\partial t}=\sum_{\rho}\sum_{\mathbf{r}} w_{\rho}(\mathbf{x}_{\mathbf{r}})\left[\exp\left(\sum_{\beta}\frac{\partial S}{\partial x_{\mathbf{r}\beta}}\nu_{\rho\beta}\right)-1\right]+\epsilon\sum_{\alpha}\frac{\tilde{D}_{\alpha}}{2}\sum_{\mathbf{r},\mathbf{a}} x_{\mathbf{r}\alpha}\left[\exp\left(-\frac{\partial S}{\partial x_{\mathbf{r}\alpha}}+\frac{\partial S}{\partial x_{(\mathbf{r}+\mathbf{a})\alpha}}\right)-1\right]\equiv H\left(\{x_{\mathbf{r}\alpha}\},\left\{\frac{\partial S}{\partial x_{\mathbf{r}\alpha}}\right\}\right), \tag{5}$$

where $w_{\rho}(\mathbf{x}_{\mathbf{r}})=W_{\rho}(\mathbf{X}_{\mathbf{r}})/\Delta V$. This is a Hamilton-Jacobi equation for the action S of an auxiliary system with coordinates $x_{r\alpha}$ and conjugate ''momenta'' $p_{r\alpha}=\partial S/\partial x_{r\alpha}$. Hamilton's equations of motion for these variables take the standard forms [21]

$$\frac{\partial x_{r\alpha}}{\partial t}=\frac{\partial H}{\partial p_{r\alpha}}=\sum_{\rho} w_{\rho}(\mathbf{x}_{r})\nu_{\rho\alpha}\exp\left(\sum_{\beta} p_{r\beta}\nu_{\rho\beta}\right)+\epsilon\frac{\tilde{D}_{\alpha}}{2}\{x_{r\alpha}[-\exp(-p_{r\alpha}+p_{(r-1)\alpha})-\exp(-p_{r\alpha}+p_{(r+1)\alpha})]+x_{(r-1)\alpha}\times\exp(-p_{(r-1)\alpha}+p_{r\alpha})+x_{(r+1)\alpha}\times\exp(-p_{(r+1)\alpha}+p_{r\alpha})\}, \tag{6}$$

$$\frac{\partial p_{r\alpha}}{\partial t}=-\frac{\partial H}{\partial x_{r\alpha}}=\sum_{\rho}\frac{\partial w_{\rho}(\mathbf{x}_{\mathbf{r}})}{\partial x_{r\alpha}}\left[\exp\left(\sum_{\beta} p_{r\beta}\nu_{\rho\beta}\right)-1\right]+\epsilon\frac{\tilde{D}_{\alpha}}{2}\{[\exp(-p_{r\alpha}+p_{(r-1)\alpha})-1]+[\exp(-p_{r\alpha}+p_{(r+1)\alpha})-1]\}. \tag{7}$$

The action S is given by the integral of the Lagrangian associated with the Hamiltonian H along classical trajectories, i.e. solutions to Eqs. (6) and (7). Boundary conditions for these equations are determined by the structure of the fluctuations. The quasistationary probability distribution far from the deterministic state is formed by occasional large fluctuations. These fluctuations follow with overwhelming probability trajectories given by the solutions to Hamilton's equations (on the unstable manifold) projected onto the x coordinates [18,19]. In the following we concentrate on fluctuations away from the synchronized solution of the deterministic system.

III. REDUCED SYSTEM: NONLINEAR PHASE AND MOMENTUM DIFFUSION EQUATIONS

A. Method of multiple time scales

In this section we apply the many variable version of the method of multiple time scales [22] to Hamilton's equations (6) and (7). We expand the time derivatives as well as the dependent variables in powers of the small parameter ϵ. This allows us to separate the fast time scale on which oscillations occur from slow time scales on which diffusion occurs, and to derive a reduced (Hamiltonian) system for the phase and conjugate momentum. In the Appendix we derive a corresponding reduced Hamilton-Jacobi equation for the reduced (action) stochastic potential. These expressions give the stochastic potential for any phase profile in terms of an integral of the Lagrangian along a deterministic path.

For small values of the diffusion terms, $\epsilon \ll 1$, a cell is only slightly perturbed by neighboring cells through diffusion, and the oscillation within the cell remains close to that of the unperturbed system. The evolution of concentrations within a cell may then be described in terms of its phase. We expect the phase to increase approximately linearly with time, and also to evolve on a slow time scale due to weak diffusive coupling. These considerations motivate the introduction of the method of multiple time scales in which the solution is assumed to depend upon infinitely many time scales $\tau_i=\epsilon^i t,\ i=1,2,\ldots;\ \tau_i$ defines a long time scale because it is not negligible when t is of order $1/\epsilon^i$. Even though t and τ_i are treated as independent variables, this is only an artifice that eliminates fast oscillatory motion and allows us to construct equations of motion on slow time scales. We use the time scales to keep the perturbation expansion uniformly valid (this is equivalent to the elimination of secular terms). First the variables $x_{r\alpha}$ and $p_{r\alpha}$ are expanded in ϵ,

$$x_{r\alpha}=x_{r\alpha}^{(0)}+\epsilon x_{r\alpha}^{(1)}+\cdots, \tag{8}$$

$$p_{r\alpha}=0+\epsilon p_{r\alpha}^{(1)}+\cdots, \tag{9}$$

and each term in the expansions is regarded as dependent on an infinite number of time scales τ_i. In the expansion for the momentum, we take the zeroth order term to be zero; this choice insures that the zeroth order term $x_{r\alpha}^{(0)}$ is a solution to the deterministic equation (which we will take to be the limit cycle solution). The derivative with respect to time becomes

$$\frac{d}{dt}=\frac{\partial}{\partial\tau_0}+\epsilon\frac{\partial}{\partial\tau_1}+\cdots. \tag{10}$$

Substituting these expansions into Eqs. (6) and (7), and collecting like powers of ϵ, we obtain a hierarchy of equations. At each order in the hierarchy we demand that the solution be periodic in the fast time τ_0 at each spatial position r; this condition will determine the evolution on slow time scales. At zeroth order we have

$$\frac{\partial x_{r\alpha}^{(0)}}{\partial\tau_0}=\sum_{\rho} w_{\rho}(x^{(0)})\nu_{\rho\alpha}. \tag{11}$$

This is the deterministic system without diffusion, and describes the evolution of the average numbers of species. We take the deterministic limit cycle as the solution for $x_{r\alpha}^{(0)}$; we assume from Eq. (9) that $p_{r\beta}^{(0)}=0$. The dependence of this solution on slow times $\tau_{i>0}$ and the spatial coordinate r is incorporated into a phase variable $\theta_r(\tau_1,\tau_2,\ldots)$:

$$x_{r\alpha}^{(0)}=x_{\alpha}^{(0)}(\tau_0+\theta_r). \tag{12}$$

At first order we have

$$\frac{\partial}{\partial \tau_0} x^{(1)}_{r\alpha} - \sum_{\rho,\beta} \frac{\partial w_\rho(x^{(0)}_{r\alpha})}{\partial x_\beta} \nu_{\rho\alpha} x^{(1)}_{r\beta} = -\frac{\partial}{\partial \tau_1} x^{(0)}_{r\alpha} + \sum_{\rho,\beta} w_\rho(x^{(0)}_{r\alpha}) \nu_{\rho\alpha} p^{(1)}_{r\beta} \nu_{\rho\beta} + \frac{\tilde{D}_\alpha}{2} \{ -2x^{(0)}_{r\alpha} + x^{(0)}_{(r-1)\alpha} + x^{(0)}_{(r+1)\alpha} \}, \tag{13}$$

$$\frac{\partial}{\partial \tau_0} p^{(1)}_{r\alpha} + \sum_{\rho,\beta} \frac{\partial w_\rho(x^{(0)}_{r\alpha})}{\partial x_\alpha} \nu_{\rho\beta} p^{(1)}_{r\beta} = 0. \tag{14}$$

The left hand side of Eq. (13), equated to zero, is the linearized deterministic system; it has a periodic solution, and is, to within a factor of the amplitude, the velocity $(\partial/\partial\tau_0) x^{(0)}_\alpha(\tau_0 + \theta_r)$. Equation (14) is the adjoint of the linearized deterministic equation; from Floquet theory it also has a periodic solution v_α. For a given spatial position r, the scalar product of these periodic solutions is a constant on the fast time scale τ_0: $\Sigma_\alpha v_\alpha(\tau_0 + \theta_r)(\partial/\partial\tau_0) x^{(0)}_\alpha(\tau_0 + \theta_r) = \text{const}$ (this may be shown by taking the time derivative of the product and using the governing equations). We take the constant to be unity. The general solution to Eq. (14) that is periodic in τ_0 may then be written as the product of v_α and an amplitude that is dependent on the slow times as well as on r:

$$p^{(1)}_{r\alpha} = a_r(\tau_1, \ldots) v_\alpha(\tau_0 + \theta_r). \tag{15}$$

Substituting this form of $p^{(1)}_{r\alpha}$ into Eq. (13) and requiring that solutions be periodic in τ_0, we obtain a solvability condition for $x^{(1)}_{r\alpha}$. This condition is the orthogonality of the right hand side of Eq. (13) to the solution v_α to the adjoint equation: $\int d\tau_0 R v = 0$. We refer the reader to Ref. [23] and to our previous work on the stochastic potential for time periodically forced limit cycles [24] for details.

The evolution of the amplitude a_r may similarly be obtained from the solvability condition for (a τ_0 periodic solution) $p^{(2)}_{r\alpha}$. From the second order term in the hierarchy of equations, the equation for $p^{(2)}_{r\alpha}$ is

$$\frac{\partial}{\partial \tau_0} p^{(2)}_{r\alpha} + \sum_{\rho,\beta} \frac{\partial w_\rho(x^{(0)}_r)}{\partial x_\alpha} \nu_{\rho\beta} p^{(2)}_{r\beta} = -\frac{\partial}{\partial \tau_1} p^{(1)}_{r\beta} - \frac{1}{2} \sum_\rho \frac{\partial w_\rho(x^{(0)}_r)}{\partial x_\alpha} \left(\sum_\beta \nu_{\rho\beta} p^{(1)}_{r\beta} \right)^2 - \sum_{\rho,\beta,\gamma} \frac{\partial^2 w_\rho(x^{(0)}_r)}{\partial x_\alpha \partial x_\beta} x^{(1)}_{r\beta} \nu_{\rho\gamma} p^{(1)}_{r\gamma} + \frac{\tilde{D}_\alpha}{2} \{ 2p^{(1)}_{r\alpha} - p^{(1)}_{(r-1)\alpha} - p^{(1)}_{(r+1)\alpha} \}. \tag{16}$$

At this order, the equation for $x^{(2)}$ is not needed for our approximation.

B. Nonlinear phase and momentum diffusion equations

After much simplification, the solvability conditions for Eqs. (13) and (16) may be reduced to coupled nonlinear phase and momentum diffusion equations:

$$\frac{\partial}{\partial \tau_1} \theta = c_1 a + c_2 \nabla^2 \theta + c_3 (\boldsymbol{\nabla} \theta)^2, \tag{17}$$

$$\frac{\partial}{\partial \tau_1} a = -c_2 \nabla^2 a + 2c_3 \boldsymbol{\nabla} \cdot (a \boldsymbol{\nabla} \theta). \tag{18}$$

In the above simplification, we have changed the discrete spatial coordinate, subscript r, to a continuous variable; accordingly, we write $\theta(r,\tau_1)$ and $a(r,\tau_1)$. The constants c_1, c_2, and c_3 have simple expressions as averages over the deterministic limit cycle:

$$c_1 = [v(\tau_0), B(x^{(0)}(\tau_0)) v(\tau_0)]_{T_0}, \tag{19}$$

$$c_2 = \left[v(\tau_0), D \frac{\partial x^{(0)}}{\partial \tau_0} \right]_{T_0} \tag{20}$$

$$c_3 = \left[v(\tau_0), D \frac{\partial^2 x^{(0)}}{\partial \tau_0^2} \right]_{T_0}, \tag{21}$$

where the matrix B is

$$B_{\alpha\beta}(x^{(0)}) = \sum_\rho w_\rho(x^{(0)}_{r\alpha}) \nu_{\rho\alpha} \nu_{\rho\beta}, \tag{22}$$

and the scalar product is defined as

$$[a,b]_{T_0} = \frac{1}{T_0} \int_0^{T_0} d\tau_0 a(\tau_0) \cdot b(\tau_0). \tag{23}$$

We note that the matrix B is positive definite, and therefore $c_1 > 0$. Also, if the matrix of diffusion coefficients D is, or is sufficiently close to, diagonal with equal elements, then $c_2 > 0$ (this follows from $v \cdot \partial x^{(0)}/\partial\tau_0 = 1$).

The deterministic system is recovered from Eqs. (17) and (18) for $a_r = 0$, and is a nonlinear phase diffusion equation, which is equivalent to the Burgers equation. Kuramoto [1] discussed how the coefficient c_2 may become negative, which causes a loss of stability of uniform oscillations. We see that this can only occur if the diffusion matrix is sufficiently far from a diagonal matrix with identical elements. In the following we assume that coherent oscillations in the deterministic equation are stable, i.e., $c_2 > 0$. We also note that the coefficient c_1 characterizes diffusion of the phase along the limit cycle in a homogeneous oscillatory system [25]. In this case the diffusion coefficient for the phase is proportional to c_1 and inversely proportional to the system size.

Using the solution, of Eqs. (8) and (9), and averaging the Hamilton-Jacobi equation (5) over the fast time τ_0, we derive the following reduced Hamilton-Jacobi equations in the Appendix:

$$\frac{\partial}{\partial \tau_1} S_1 = -\mathcal{H}(\theta,a) = -\int dr\, H_1(\theta,a), \tag{24}$$

$$\frac{\delta S_1[\theta(r)]}{\delta \theta(r)} = a(r), \tag{25}$$

where the reduced stochastic potential is defined through $S_1 = \Delta V\overline{S}/\epsilon$ (the overbar denotes averaging over τ_0) and

$$H_1 = a\left(\frac{1}{2}c_1 a + c_2 \nabla^2 \theta + c_3 (\boldsymbol{\nabla}\theta)^2\right). \tag{26}$$

Relation (25) expresses the momentum a as the functional derivative of S_1 with respect to θ. It is easily verified that Hamilton's equations that correspond to H_1 are Eq. (17) equal to $\delta H_1/\delta a$ and Eq. (18) equal to $-\delta H_1/\delta\theta$.

IV. SOLUTIONS FOR THE STOCHASTIC POTENTIAL

A. Solution to *d*-dimensional systems

The stochastic potential has the following convenient interpretation in terms of the auxiliary classical mechanical system. The total derivative with respect to τ_1 of the action S_1 (the reduced stochastic potential) along classical trajectories, i.e., solutions to Eqs. (17) and (18), is

$$\frac{dS_1}{d\tau_1} = \frac{\partial S_1}{\partial \tau_1} + \int dr \frac{\delta S_1}{\delta \theta(r)} \frac{\partial \theta(r)}{\partial \tau_1}, \tag{27}$$

which is the Lagrangian $\mathcal{L}$ corresponding to the reduced Hamiltonian:

$$\frac{dS_1}{d\tau_1} = \int dr\left[-H_1 + a\frac{\partial \theta}{\partial \tau_1}\right] = \frac{1}{2}c_1 \int dr\, a^2 \equiv \mathcal{L}. \tag{28}$$

That is, S_1 is the action evaluated along a classical dynamical trajectory. For general time-dependent distributions, the classical trajectories are not known, and the equations must be numerically integrated. For the stationary distribution, the projection of a trajectory onto the x coordinates is the path followed with overwhelming probability away from the deterministic solution; it is the most probable or optimal fluctuational path. In this case, the boundary conditions for Eqs. (17) and (18) follow from physical considerations: for times far in the past, $\tau_1 \to -\infty$, the system is near the spatially synchronized state $\theta(r) = \text{const}$. The paths followed away from this state lie on its unstable manifold (in the Hamiltonian system).

Referring to the definition of S_1 (see the Appendix) and relation (4), we may express the probability distribution in terms of S_1:

$$P[\theta(r),\tau_1] \sim \exp\{-\epsilon S_1[\theta(r),\tau_1]\}. \tag{29}$$

In summary, the main results of perturbation theory are the reduced Hamilton's equations and the relation for the stochastic potential in terms of a Lagrangian. These equations may be used to study probability densities for other states. We restrict our attention to the stationary distribution for synchronized states. From Eq. (28) and the appropriate boundary conditions, we write the stochastic potential for the stationary distribution as

$$S_1 \approx \frac{1}{2}c_1 \int_{-\infty}^{\tau_f} d\tau_1 \int dr\, a^2. \tag{30}$$

We note that the potential increases along classical mechanical trajectories away from the synchronized state.

An explicit expression for the unstable manifold $a(\theta(r))$ is obtained from the Hamiltonian-Jacobi equation (28). The action is independent of time τ_1, if $H=0$ for $a(\theta)=0$ (which gives the deterministic system), and for

$$a(\theta) = -\frac{2}{c_1}[c_2 \nabla^2 \theta + c_3 (\boldsymbol{\nabla}\theta)^2]. \tag{31}$$

Putting this expression into Eq. (17) for θ yields the macroscopic nonlinear phase diffusion equation with time reversed, i.e., $\tau_1 \to -\tau_1$. That is, most probable fluctuational trajectories that start from the vicinity of the synchronized state and arrive at a given $\theta(r)$ are time-reversed deterministic paths. This important property of coupled oscillator systems does not hold for typical n-dimensional systems (which lack detailed balance) [19]. This property of time reversibility also occurs in periodically forced (homogeneous) oscillatory systems [24].

Using the above result that the amplitude a is proportional to the velocity in the deterministic phase equation, we write the probability density in terms of deterministic paths:

$$P[\theta_0] \sim \exp\left(-\epsilon \int_{-\infty}^{t_f} d\tau_1 \int dr\, a^2(\theta)\right)$$
$$\sim \exp\left(\frac{4\epsilon}{c_1^2} \int_0^{\infty} d\tau_1 \int dr \left[\frac{\partial}{\partial \tau_1}\theta_{\text{det}}(r,\tau_1)\right]^2\right), \tag{32}$$

where θ_{det} is the solution to the deterministic equation:

$$\frac{\partial}{\partial \tau_1}\theta_{\text{det}} = c_2 \nabla^2 \theta_{\text{det}} + c_3 (\boldsymbol{\nabla}\theta_{\text{det}})^2. \tag{33}$$

The solution to this equation may be explicitly written as

$$\theta_{\text{det}}(r,\tau_1) = \frac{c_2}{c_3}\ln[w(r,\tau_1)],$$
$$w(r,\tau_1) = \int_{-\infty}^{\infty} \frac{d\zeta}{\sqrt{4\pi c_2 \tau_1}} \exp\left[-\frac{(r-\zeta)^2}{4c_2\tau_1} + \frac{c_3}{c_2}\theta_0(\zeta)\right]. \tag{34}$$

Except for one-dimensional systems, these equations must be evaluated numerically.

B. Explicit stationary solution for one-dimensional systems

For $d=1$ a stationary solution of Eq. (24) for the reduced stochastic potential is

$$S_1 = \frac{c_2}{c_1}\int dr (\boldsymbol{\nabla}\theta)^2, \tag{35}$$

which may be verified using Eqs. (28) and (25). From Eq. (29) we obtain a Gaussian stationary probability distribution

$$P[\theta]\sim \exp\left[-\epsilon\frac{c_2}{c_1}\int dr(\nabla\theta)^2\right]. \tag{36}$$

This solution is independent of c_3, the coefficient of the nonlinear term, and is a solution of the linear Hamilton-Jacobi equation [i.e., of Eq. (24), with the nonlinear term omitted]. It follows from the form of the above solution that $\nabla\theta$ is an independent random variable, and that the phase θ performs a random walk. (Corresponding to a random walk in one dimension is a roughness exponent of $\alpha=1/2$).

V. RELATION OF THE REDUCED EQUATION TO THE KPZ EQUATION

A. Kardar-Parisi-Zhang equation

The most general form of an equation describing the growth of a noise driven interface is the Kardar-Parisi-Zhang (KPZ) equation [11–13]

$$\frac{\partial h}{\partial t}=\nu\nabla^2 h+\frac{\lambda}{2}(\nabla h)^2+\eta, \tag{37}$$

where h is the height of a growing interface. The driving noise η, which may be thermal in origin, is considered to be Gaussian with $\langle\eta\rangle=0$, and δ function correlated in space and time:

$$\langle\eta(r,t)\eta(r',t')\rangle=C\delta(r-r')\delta(t-t'). \tag{38}$$

That is, the KPZ equation is the deterministic nonlinear phase diffusion equation [Eq. (17), with $a=0$] driven with white noise. The noise causes an initially flat front to roughen and display a self-affine structure. A measure of this roughening is the width

$$w^2(L,t)=\left\langle L^{-d}\int_0^L dr(h-\overline{h})^2\right\rangle, \tag{39}$$

where d is the dimension of the space, and the angular brackets denote averaging over samples.

Both transient and stationary scaling relations have been investigated for the KPZ equation. In the following, we recall some of these results [12,13]. An exact solution for the stationary probability density only exists for one-dimensional systems, and is

$$P[h]\sim \exp\left[-\frac{\nu}{C}\int dr(\nabla h)^2\right], \tag{40}$$

as may be verified using the Fokker-Planck equation [in Sec. B, Eq. (43)]. We note that this solution is of the same form as that for the density of the phase in one dimensional diffusion-coupled oscillators [Eq. (36)].

For long times, the width of a front saturates at a value that depends on the system size L,

$$w\sim L^{\alpha}\quad [t\gg t_{\times}], \tag{41}$$

where $t_{\times}$ is the crossover time at which the width changes from a time-dependent value to a time independent (saturation) value. The exponent α characterizes the roughness of a front. For one-dimensional systems, the exact solution [Eq. (40)] shows that the local slopes ∇h follow a Gaussian distribution, random and uncorrelated. The front is obtained by summing these local random slopes, which produces a random walk. Hence it follows that in one-dimensional systems $\alpha=1/2$, the random walk exponent. For short times the width increases as a power of time, and may be characterized by a growth exponent β:

$$w\sim t^{\beta}\quad [t\ll t_{\times}]. \tag{42}$$

With the use of scaling arguments, it can be shown that $\beta=1/3$. We refer the reader to the literature for other scaling relations [12,13].

Unlike the one-dimensional case, exact solutions do not exist for higher-dimensional systems, and numerical methods have been used to determine scaling exponents. For $d=2$, recent numerical solutions [13] of the Fokker-Planck equation give the following values: $\alpha=0.36$ and $\beta=0.22$. Therefore, as in one dimension, only a rough state exists. For $d>2$ the scaling exponents exhibit two different values depending on the coupling strength, which is defined through $g^2\equiv\lambda^2C/2\nu^3$. The weak coupling regime occurs for $g<g^*$; the exponents are described by linear theory: $\alpha=(2-d)/2$ and $\beta=(2-d)/4$. Since the exponents are less than zero, the front is flat. The strong coupling regime occurs for $g>g^*$; the exponents are positive, and hence the front is rough. For $d=3$ the critical value at which a phase transition occurs is $g^*=32$. The approximate values for the exponents are $\alpha=0.3$ and $\beta=0.18$.

B. Eikonal approximation of the KPZ equation

The probability density p for the KPZ equation obeys a Fokker-Planck equation [12,13]

$$\frac{\partial p}{\partial t}=-\int dr\frac{\delta}{\delta h}\left\{\left[\nu\nabla^2 h+\frac{\lambda}{2}(\nabla h)^2\right]p\right\}+\frac{C}{2}\int dr\frac{\delta^2}{\delta h^2}p. \tag{43}$$

For small values of the noise strength C, we assume an asymptotic WKB form for the probability density p:

$$p[h]\sim \exp\left[-\frac{s}{C}\right]. \tag{44}$$

Substituting this into the Fokker-Planck equation, and collecting leading order terms, we obtain the following eikonal approximation:

$$-\frac{\partial s}{\partial t}=\int dr\left\{\left[\nu\nabla^2 h+\frac{\lambda}{2}(\nabla h)^2\right]\xi+\frac{1}{2}\xi^2\right\}. \tag{45}$$

A comparison with Eq. (24) shows that the above eikonal approximation for the KPZ equation is equivalent to our reduced Hamilton-Jacobi description for diffusion-coupled oscillators. After changing the time variable $\tau=c_1\tau_1$, we have the following correspondence: $\nu=c_2/c_1$, $\lambda/2=c_3/c_1$, and $C=1/\epsilon$.

C. Results for synchronization of diffusion-coupled oscillators

From the above correspondence between Eqs. (24) and (45), the results outlined for the KPZ equation apply directly to the diffusion-coupled oscillator problem. In fewer than three spatial dimensions mesoscopic scale internal fluctuations have a profound effect on macroscopic dynamics: only a rough or spatially desynchronized state exists, even though homogeneous oscillations are stable for the deterministic system. The desynchronization of phase over large spatial scales may be characterized by a roughness exponent α; the root mean square deviation in phase scales with the length of the system as L^{α}. An exact solution exists for $d=1$, and gives a roughness exponent of $\alpha=1/2$. In $d=3$, weak and strong scaling regimes are separated by a critical value of the coupling strength g, where

$$g=\frac{2c_1c_3^2}{\epsilon c_2^2}. \tag{46}$$

From the results for the KPZ equation, we observe that desynchronization typically occurs in three-dimensional systems, in which the value of ϵ (diffusion coefficients or inverse spatial scale) is small (such that $g>g^*=32$). We note that small values of the coefficient c_1, which characterizes phase diffusion along the limit cycle in a homogeneous system, may partially cancel the effect of ϵ on the coupling strength. That is, for systems with small phase diffusion coefficients ($\propto c_1$), desynchronization of the phase occurs on longer spatial scales $l^2\sim\epsilon^{-1}\sim c_1^{-1}$. Larger values of ϵ increase coupling between oscillators, and inhibit phase desynchronization.

VI. DISCUSSION

We have described the influence of internal fluctuations on diffusion-coupled oscillatory systems. Using the eikonal approximation and the method of multiple time scales, we reduced a master equation description to a Hamilton-Jacobi equation for the stochastic potential of the phase. This simplification of the evolution equations allowed us to obtain an exact solution for the stochastic potential of one-dimensional systems and, in general, to relate the reduced Hamilton-Jacobi equation to the eikonal approximation of the KPZ equation (a noise driven nonlinear diffusion equation which describes some types of interface growth). Important results include the destruction of homogeneous synchronized solutions for $d=1$ and 2, which are stable in the deterministic system, and the presence of a critical system length below which oscillations are spatially synchronized for $d=3$.

An important area for investigation is the influence of internal fluctuations on periodically forced oscillatory reaction-diffusion systems. Work on homogeneously forced deterministic systems has shown mode-locked spatial patterns [26–30]. Such mode-locked patterns may serve as computational devices [31,32]. Whether these structures persist with internal fluctuations is an open question. Using the methods of the present paper along with those used to describe fluctuations in homogeneously forced (homogeneous) oscillatory systems [24] should help resolve this problem.

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APPENDIX

A reduced Hamilton-Jacobi (HJ) equation is associated with Eqs. (17) and (18). To derive this from the original HJ equation, [Eq. (5)], we substitute the expansion for the solution [Eq. (8)] into the Hamiltonian, and average over the fast time τ_0. First we write the potential in terms of the phase and time,

$$S(\{\theta_r\},t)=S(\{x^{(0)}(\tau_0+\theta_r)+\epsilon x^{(1)}+\cdots\},t), \tag{A1}$$

which gives the following partial derivative with respect to time:

$$\begin{aligned}\frac{\partial}{\partial t}S(\{\theta_r\},t)&=\frac{\partial}{\partial t}S(\{x^{(0)}+\cdots\},t)\\&\quad+\sum_{r,\alpha}\frac{\partial S}{\partial x_{r\alpha}}\frac{\partial}{\partial t}[x_{r\alpha}^{(0)}+\epsilon x_{r\alpha}^{(1)}+O(\epsilon^2)]\\&=-H+\epsilon\sum_r a_r\left(1+\epsilon v\frac{\partial}{\partial t}x^{(1)}+O(\epsilon^2)\right).\end{aligned} \tag{A2}$$

Then, expanding the time using multiple time scales and averaging over the fast time τ_0, we obtain, to second order in ϵ,

$$\begin{aligned}&\overline{\frac{\partial S(\{\theta_r\},t)}{\partial\tau_0}}+\epsilon\overline{\frac{\partial S(\{\theta_r\},t)}{\partial\tau_1}}+O(\epsilon^2)\\&\quad=-\overline{H}+\epsilon\sum_r a_r\left(1+\epsilon\left[v,\frac{\partial}{\partial\tau_0}x^{(1)}\right]_{T_0}\right),\end{aligned}$$

where the overbar denotes the average over $\tau_0:\overline{f}=(1/T_0)\int d\tau_0 f$. After simplification, we have the explicit form of the reduced Hamilton-Jacobi equation:

$$\frac{\partial}{\partial\tau_1}\overline{S}=-\epsilon\sum_r H_1(\{\theta_r\},\{a_r\}) \tag{A3}$$

$$H_1=a_r\left(\frac{1}{2}c_1a_r+c_2\nabla^2\theta_r+c_3(\boldsymbol{\nabla}\theta_r)^2\right). \tag{A4}$$

We note that θ_r and a_r are not conjugate variables since $\partial\overline{S}/\partial\theta_r=\epsilon a_r$. Also, the right hand side of Eq. (A3) has a sum over the discrete index r. [In Eq. (A4), we have taken the limit of small cell length l and introduced continuous derivatives.] These deficiencies may be remedied by scaling the potential: $S_1=\Delta V\overline{S}/\epsilon$. The Hamilton-Jacobi equation for S_1 is given by Eq. (24).

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