

Interplay between noise and boundary conditions in pattern formation in adsorbed substances

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We have studied the interplay between noise and boundary conditions on the possibility of noise induced pattern formation. With this aim, we have exploited a deterministic model for pattern formation in adsorbed substances—including the effect of lateral interactions—used to describe the phenomenon of adsorption in surfaces, where a multiplicative noise fulfilling a fluctuation-dissipation relation was added. We have found solutions for different boundary conditions, particularly corresponding to two stable and one unstable patterns, where one of the stable and the unstable one, are purely induced by the multiplicative noise. In the case of albedo boundary conditions we have found a transition from monostable to a noise induced bistable behavior as the albedo parameter is varied.

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I. INTRODUCTION

During the second half of the 20th century a wealth of research results on fluctuations or noise have lead us to the recognition that in many situations noise can actually play a constructive role, triggering new phenomena or new forms of order. To name just a few, consider the following examples: noise-induced transitions [1], noise-induced phase-transitions [2–4], stochastic resonance in zero-dimensional and extended systems [5–8], noise-induced transport [9,10], coupled Brownian motors [11,12], noise-sustained patterns [13–16].

Regarding the latter phenomenon, that is noise-sustained patterns, there have been numerous studies on many different situations. However, both the study of noise-sustained patterns in adsorbed substances and the study of the interplay between noise and boundary conditions are scarce. The problem of phase transitions and ordering phenomena in adsorbed films on crystal surfaces has attracted considerable interest for many years (see Ref. [17] for a recent review). Formation of ordered structures of particles adsorbed on surfaces due to mutual (lateral) interactions has been observed in many experiments, originating different types of adsorption isotherm [18,19]. Strong attractive (lateral) interactions lead to phase separation with different coverage, characteristic of first order phase transitions [18,19]. Recently, a simple extension of the indicated model was introduced in order to qualitatively describe multilayer adsorption and the formation of ordered structures [20]. This was motivated by that it is experimentally well known that adsorption on surfaces is not only restricted to the formation of monolayers, but rather a second layer can condensate on the first one, a third on the second, and so on [21]; with numerical simulations also showing similar results [22].

In this work, inspired by the experimental and theoretical results in Ref. [23], and in order to analyze on one hand the

possibility of generating noise-induced spatial structures in adsorbed systems, while on the other the interplay between noise and boundary conditions on pattern formation, we consider the same model discussed in Ref. [20], but restricted to the monolayer case as in Ref. [18] (where all the relevant deterministic contributions to the adsorption process, adsorption, desorption, lateral interaction, and diffusion, have been included), with the addition of a multiplicative noise source. The form we adopted here for the noise source is such that a fluctuation-dissipation relation is fulfilled [24] (hence avoiding the violation of the second law of thermodynamics). We exploit the same approach introduced in Refs. [25,26].

In the following we make a brief description of the deterministic model [18,20], and describe how the indicated multiplicative noise is included. Afterwards, we analyze the solutions for the different cases, that is for the different boundary conditions. Finally we draw some conclusions and discuss on the possibility of carrying out an experimental observation of the indicated phenomena.

II. DETERMINISTIC MODEL AND THE MULTIPLICATIVE NOISE

A. The deterministic model

As in Refs. [18–20], we adopt a continuous description for the surface, and characterize the adsorptive species through evolution equations for $\mathbf{C}(x, t)$, the local coverage at the surface. We consider a system that extends within the range $-L/2 < x < L/2$, and impose the chosen boundary conditions at $x = \pm L/2$. The adsorptive term is characterized by the constant k_a , and the adsorption is only possible at the $(1 - \mathbf{C})$ free sites. Hence, the adsorption rate is $k_a p (1 - \mathbf{C})$, where p is the partial pressure of the gaseous phase.

The desorption process has a rate k_d , that includes $k_{d,0}$, the desorption of noninteracting particles, and the corrections due to the lateral interactions. The strong local bond induced by the interaction $U(x)$, corrects the desorption rate as $k_d = k_{d,0} \exp[U(x)/kT]$, where k is Boltzmann constant and T is the temperature. According to the form we use to introduce

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such an interaction, we are assuming that it is a *substratum mediated* interaction [18–20].

The gradient of $U(x)$ produces a force $F = -\partial U(x)/\partial x$, that affects the adsorbed particles inducing a velocity $v = bF$, where b is the mobility (given by Einstein's relation $b = D/kT$, with D the diffusion coefficient). The associated particle flux is vC . Because the flux is only possible to the $(1 - C)$ free sites, its form results to be

$$j = -\left(\frac{D}{kT}\right)C(1 - C)\frac{\partial U(x)}{\partial x},$$

while the diffusive flux is given by

$$j_{\text{dif}} = -D\frac{\partial C}{\partial x}.$$

The evolution equation for the coverage is

$$\begin{aligned} \frac{\partial}{\partial t}C(x, t) &= k_{ad}p(1 - C) - k_{d,0}C \exp[U(x)/kT] \\ &+ \frac{\partial}{\partial x}\left(\frac{D}{kT}\frac{\partial U(x)}{\partial x}C(1 - C) + D\frac{\partial C}{\partial x}\right). \end{aligned} \quad (1)$$

For the functional form of $U(x)$ we assume an attractive (and as indicated earlier, substratum-mediated) potential among the particles separated a distance r , that we denote by $u(r)$. The potential acting on a particle located at r is

$$U(r) = -\int u(r - r')C(r')dr'. \quad (2)$$

The integration domain is the whole surface. The function $u(r)$ depends on the nature of the system. If the interaction radius is small compared with the diffusion length, and the coverage is not much affected by variations in this radius, we can approximate

$$U(r) = \int u(r - r')C(r')dr' \approx u_0C, \quad \text{where } u_0 = \int u(r)dr. \quad (3)$$

To simplify the notation we scale the variables as follows: $\xi = x/L_{\text{dif}}$ (and $\xi_M = L/2L_{\text{dif}}$), with the diffusion length $L_{\text{dif}} = (D/k_{d,0})^{1/2}$; $\tau = t/t_d$, with $t_d = 1/k_{d,0}$; and $\varepsilon = u_0/kT$, represents the lateral interaction. We also define $\alpha = k_{ad}p/k_{d,0}$, characterizing the coverage in equilibrium when $U(x)$ becomes zero. The evolution equation for $C(\xi, \tau)$ reduces to

$$\frac{\partial C}{\partial \tau} = \alpha(1 - C) - C \exp[-\varepsilon C] + \frac{\partial}{\partial \xi}\left([1 - C\varepsilon(1 - C)]\frac{\partial C}{\partial \xi}\right). \quad (4)$$

This equation may be written as

$$\frac{\partial C}{\partial \tau} = f(C) + \frac{\partial}{\partial \xi}\left(D_{\text{eff}}(C)\frac{\partial C}{\partial \xi}\right), \quad (5)$$

with

$$\begin{aligned} f(C) &= \alpha(1 - C) - C \exp[-\varepsilon C] \\ D_{\text{eff}}(C) &= 1 - C\varepsilon(1 - C). \end{aligned} \quad (6)$$

Hence, this problem may be visualized as a reaction-diffusion system being the reaction term $f(C)$ and the effective diffusion coefficient $D_{\text{eff}}(C)$. For some region of the parameters α and ε this system is bistable with two homogeneous solution indicating that the adsorbate may be present in two different coverage [18]. In this region the effective diffusion coefficient is negative. We worked with $\varepsilon < 0.4$, to assure that $D_{\text{eff}}(C) > 0$ always, and $\alpha > 0.14$ in order to be outside of the bistable region.

B. Multiplicative noise

Equation (5) may be written in a variational form as

$$\frac{\partial C}{\partial \tau} = -\frac{1}{D_{\text{eff}}(C)}\frac{\delta V[C]}{\delta C(\xi)}, \quad (7)$$

where the potential functional $V[C]$

$$\begin{aligned} V[C] &= \int_{-\xi_M}^{\xi_M} d\xi \left(-\int_0^C dC' D_{\text{eff}}(C')f(C') \right. \\ &\quad \left. + \frac{1}{2}[D_{\text{eff}}(C)\partial_\xi C]^2 \right), \end{aligned} \quad (8)$$

is a Lyapunov functional for the deterministic dynamics.

The starting point of our stochastic analysis will be Eq. (5) with the addition of a multiplicative noise. We introduce it here in an *ad hoc* form. However, a realistic analysis will require to indicate what parameter is the one that fluctuates, and the resulting associated form for the noise term. We assume that, in the Stratonovich interpretation, it is given by

$$\frac{\partial C}{\partial \tau} = -\frac{1}{D_{\text{eff}}(C)}\frac{\delta V[C]}{\delta C(\xi)} + g(C)\eta(\xi, \tau), \quad (9)$$

where η is a Gaussian white noise, that is with zero mean and correlation $\langle \eta(\xi, \tau)\eta(\xi', \tau') \rangle = 2\sigma^2\delta(\xi - \xi')\delta(\tau - \tau')$. For the coefficient of the noise term, $g(C)$ we adopt

$$g(C) = \frac{1}{\sqrt{D_{\text{eff}}(C)}}, \quad (10)$$

in order to guarantee that the fluctuation-dissipation relation is fulfilled [24]. As we are considering the Stratonovich interpretation, the stationary solution of the associated Fokker-Planck equation may be written as [25]

$$P_{st}[C] \sim \exp(-V_{\text{eff}}[C]/\sigma^2), \quad (11)$$

where the effective potential $V_{\text{eff}}[C]$ is given by

$$V_{\text{eff}}[C] = V[C] - \lambda \int_{-\xi_M}^{\xi_M} d\xi \ln D_{\text{eff}}(C). \quad (12)$$

Here λ is a renormalized parameter related to σ through $\lambda = \sigma^2/(2\Delta\xi)$ in a lattice discretization, where $\Delta\xi$ is the lattice parameter [25].

The extreme of V_{eff} corresponds to the (stationary) noise-sustained structures C_{st} that can be computed from the equation that results making the first variation of $V_{\text{eff}}(C)$ respect to C equal to zero, that is

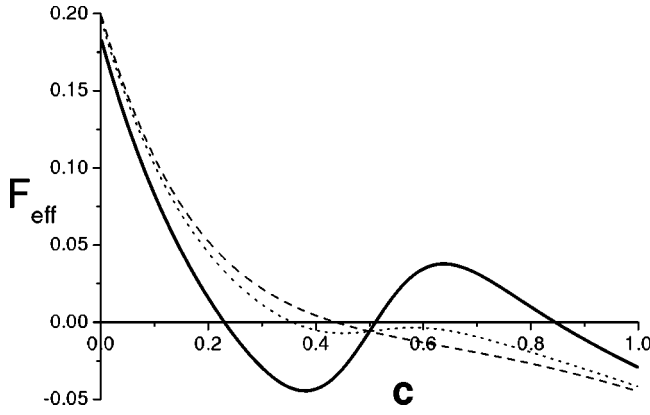


FIG. 1. (a) $F_{\text{eff}}(\mathbf{C})$ vs \mathbf{C} for $\alpha=0.2$ and $\varepsilon=3.1$. Dashed line for the reaction term with for $\lambda=0.0$ (monostable); dotted line for $\lambda=0.001$ (still monostable), and full line for $\lambda=0.005$ (bistable).

$$\delta V_{\text{eff}}[\mathbf{C}_{st}] = - \int_{-\xi_M}^{\xi_M} D_{\text{eff}}(\mathbf{C}) \{ \partial_{\xi} [D_{\text{eff}}(\mathbf{C}) \partial_{\xi} \mathbf{C}] + F_{\text{eff}}(\mathbf{C}) \} \delta \mathbf{C}(\xi) d\xi |_{\mathbf{C}=\mathbf{C}_{st}} = 0, \quad (13)$$

where

$$F_{\text{eff}}(\mathbf{C}) = f(\mathbf{C}) + \lambda \frac{1}{D_{\text{eff}}(\mathbf{C})^2} \frac{d}{d\mathbf{C}} D_{\text{eff}}(\mathbf{C}), \quad (14)$$

is the effective nonlinearity which drives the dynamics of the noise induced patterns.

As indicated before, we will consider a finite one dimensional system, i.e., limited to the region $-\xi_M \leq \xi \leq \xi_M$ ($-L/2 < x < L/2$); and consider parameter's values that assure a monostable dynamics with $D_{\text{eff}} > 0$ (for $\varepsilon < 4.0$ and $\alpha > 0.14$) in absence of noise [see Fig. 1 and Eq. (6)]. Then the effective spatial diffusivity $D_{\text{eff}}(\mathbf{C})$ induces an effective bistable dynamics when a multiplicative noise for which the fluctuation-dissipation relation is fulfilled [24] is added (see Fig. 1). However, for λ too small the bistability may even not appear. This means that a threshold exist beyond which the dynamics is bistable. The effective diffusion coefficient D_{eff} vs \mathbf{C} is always positive for $\varepsilon < 4$ and describes a parabola with a minimum in $\mathbf{C}=0.5$, implying that the effective diffusivity is larger when the coverage departs from this value. In this case it could be necessary to resort to an extended description where a multilayer system is considered [20]. However, such a case is beyond the scope of this work.

It is worth remarking here that for the deterministic problem the reaction term is monostable ($\lambda=0$) while, as we increase the noise intensity, and only due to the effect of noise, the effective nonlinear term F_{eff} becomes bistable (for λ larger than a threshold). However it is also necessary that the lateral interaction be present, otherwise the effective nonlinear term will not be bistable.

III. DIFFERENT BOUNDARY CONDITIONS

Here we analyze the patterns arising for three different boundary conditions. We will consider the case of perfect

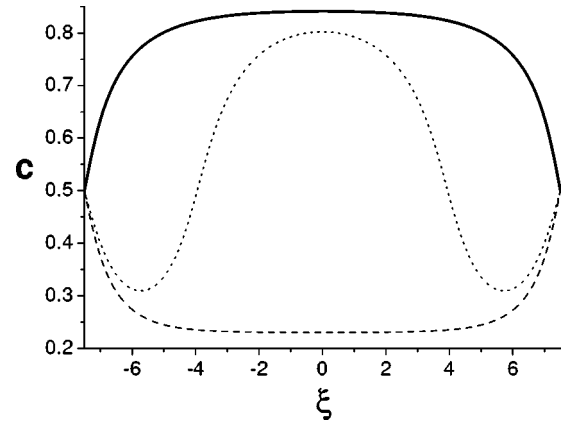


FIG. 2. Stationary patterns of the problem. We show \mathbf{C}_{up} (full line) and \mathbf{C}_{down} (dashed line), both stable patterns; and the unstable (saddle) \mathbf{C}_u (dotted line); all nonhomogeneous. Here we have $\lambda = \lambda_c \approx 0.005$, while the other parameters values are the same as in Fig. 1.

(and also imperfect) adsorption at the boundary (Dirichlet), perfect reflection (Neumann), and mixed or partially reflecting boundary conditions (albedo).

A. Dirichlet boundary conditions

The usual case of Dirichlet boundary conditions corresponds to adopting $\mathbf{C}(-\xi_M) = \mathbf{C}(\xi_M) = a$ (with a a constant), where $a=0$ or $a \neq 0$. As is well known, the former situation corresponds to having a perfect absorber at the boundary, while the latter corresponds to a case where some source is introduced at the boundary. We start here assuming the case with sources at the boundary with $\mathbf{C}(-\xi_M) = \mathbf{C}(\xi_M) = 0.5$, and postpone the analysis of the case of perfect absorption.

The patterns that we obtain as solutions, correspond to the extreme of the effective potential $V_{\text{eff}}[\mathbf{C}]$ (that is the maxima of the probability density). In order to obtain their form, we must (numerically) solve

$$\frac{d}{d\xi} \left(D_{\text{eff}}(\mathbf{C}_{st}) \frac{d}{d\xi} \mathbf{C}_{st} \right) + F_{\text{eff}}(\mathbf{C}_{st}) = 0, \quad (15)$$

for a stationary regimen profile $\mathbf{C}_{st}(\xi)$. This approach allows us to find both, the stable and unstable solutions. To analyze their stability we need to calculate $\delta^2 V_{\text{eff}}$, that defines a Sturm-Liouville problem, with orthogonality weight $D_{\text{eff}}(\mathbf{C}_{st})$. From the analysis it results that, for $\lambda=0$ (monostable dynamics) we have one stable (\mathbf{C}_{down}) nonhomogeneous symmetric pattern that presents a minimum in the unique root of the nonlinear term $[f(\mathbf{C})]$. Hence, for λ larger than a certain threshold we come to a bistability region where we found two stable nonhomogeneous symmetric patterns, \mathbf{C}_{up} with a maximum in the upper root of the nonlinear term $F_{\text{eff}}(\mathbf{C})$, and \mathbf{C}_{down} with a minimum in the lower root of the same nonlinear term; together with \mathbf{C}_u , the unstable solution (saddle). The typical form of these patterns is illustrated in Fig. 2.

It is convenient to clarify that the difference between \mathbf{C}_{down} with and without noise is due to the fact that the

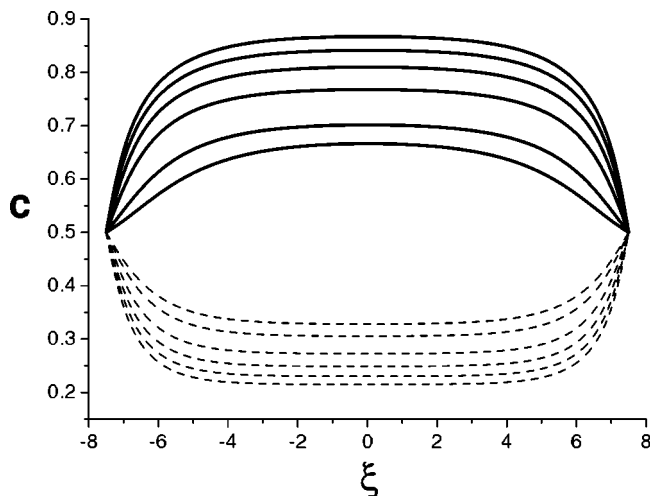


FIG. 3. Stationary patterns of the problem. We show C_{up} (full line) and C_{down} (dashed line), both nonhomogeneous stable patterns for different values of λ , increasing from bottom to top C_{up} and from top to bottom C_{down} ($\lambda=0.0015, 0.0020, 0.0030, 0.0040, 0.0050$, and 0.0060); while the other parameter values are the same as in Fig. 1.

unique root of $f(C)=0$ changes for effect of the noise. That is, it is the same pattern modified by the effect of the noise. However, C_{up} is a new pattern that is purely induced by the noise. Figure 3 depicts the stable patterns for different values of λ . We can observe those patterns have a strong dependence on the noise intensity λ (also the unstable one, not shown in the figure). For λ too large, the solution C_{up} is larger than one, and C_{down} is below zero, for the central ξ values. This result has no physical meaning, as the coverage should be normalized to one. In this case, the modified (stochastic) model does not represent a real physical situation anymore. It could maybe correspond to a transition from monolayer to multilayer adsorption as described in Ref. [20], that we will not consider here.

Figure 4 shows $V_{eff}[C_{st}]$ vs λ , evaluated on the different stationary patterns (we define λ_c as the value of λ where $V_{eff}[C_{up}] = V_{eff}[C_{down}]$). Here we see how the stable solution C_{up} is approached by the unstable solution C_u until both coalesce and only C_{down} survives as the noise intensity (λ) threshold is reached from above. These results induce to think on the possibility of stochastic resonance between the indicated noise-induced structures [26]. We have analyzed this possibility, introducing a weak external signal to rock the potential [26], and found that the typical maximum in the curve of the signal-to-noise ratio as a function of parameter λ results to be too small ($\sim 10^{-10}$) making irrelevant its study.

Let us now go back to the case $C(-\xi_M) = C(\xi_M) = 0$, corresponding to have a perfect absorber at the boundary. In this case, for $\lambda=0$ (monostable dynamics without noise), as before, we have a single pattern, but showing a maximum (at the unique root of the nonlinear term) instead of a minimum as in the previous case. Clearly this is due to the change of the boundary condition. Hence, in the region where the dynamics was previously bistable (by effect of the noise), even for larger values of λ there is also only one pattern, different from the one arising without noise, since now the maximum

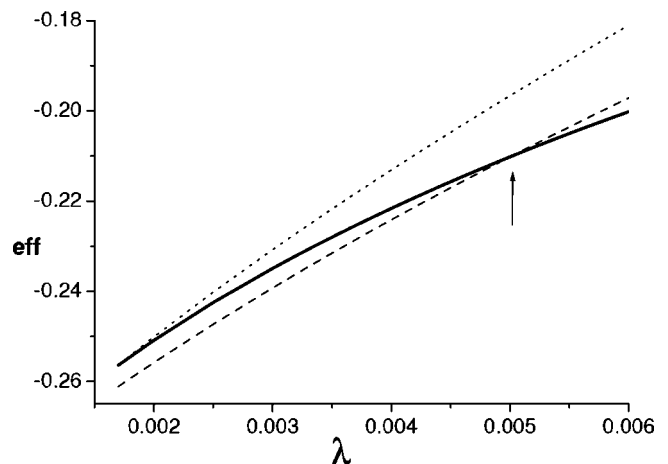


FIG. 4. Nonequilibrium potential $V_{eff}[C_{st}]$, as a function of λ , evaluated on the stationary patterns, full line corresponds to C_{up} , dashed line corresponds to C_{down} while the dotted line corresponds to C_u . The arrow indicates the point where $V_{eff}[C_{up}] = V_{eff}[C_{down}]$, corresponding to $\lambda = \lambda_c \approx 0.005$.

corresponds to the lower root of the nonlinear term with noise (it comes from the same previous root that has been modified by the noise). We observe that in this case there is no phase-transition-like phenomenon induced by noise. The other pattern that arises in the previous case, corresponding to the other root—“the upper one”—that would indicate a phase transition induced by noise, is not a solution.

B. Neumann boundary conditions

Neumann boundary conditions, defined by

$$\frac{\partial}{\partial \xi} C(\xi = -\xi_M) = \frac{\partial}{\partial \xi} C(\xi = \xi_M) = 0,$$

corresponds to the case where the current through the boundaries is zero, that is perfect reflection. In this case, without noise we have as solution a stable homogeneous pattern with a value given by the root of $f(C)=0$. When the multiplicative noise is included we find two new stable homogeneous solutions, now with values at two of the roots of $F_{eff}(C)=0$ [those where the slope of $F_{eff}(C)$ is negative]. There is also another root [where the slope of $F_{eff}(C)$ is positive], corresponding to a homogeneous saddle.

Again in this case, the homogeneous pattern corresponding to the lower root of $F_{eff}(C)$ is the same solution that for the case without noise, again modified by effect of the noise, while the one corresponding to the upper root only exists due to the presence of the noise.

C. Albedo boundary conditions

These conditions correspond to

$$\frac{\partial}{\partial \xi} C(\xi = -\xi_M) = KC(-\xi_M),$$

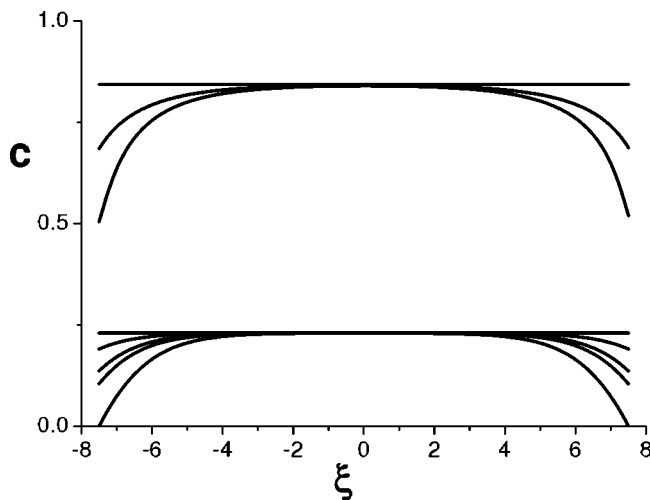


FIG. 5. Stationary stable patterns of the problem for different values of K . We show C_{up} (above $C=0.5$) with K increasing downwards ($K=0$, $K=0.01$, and $K=0.03$, respectively); and C_{down} (below $C=0.5$) with K increasing downwards too ($K=0$, $K=0.01$, $K=0.03$, $K=0.05$, and $K=\infty$, respectively). Here we have $\lambda=\lambda_c \approx 0.005$, while the other parameters values are the same as in Fig. 1.

$$\frac{\partial}{\partial \xi} C(\xi = \xi_M) = -KC(\xi_M),$$

where K is the albedo parameter, assumed positive. When $K=0$ we have Neumann's conditions; while for $K=\infty$, we recover Dirichlet's conditions [$C(-\xi_M)=C(\xi_M)=0$].

Here we explore the possible solutions varying K between both extremes, $0 < K < \infty$. The stable solutions are shown in Fig. 5. We see that for values of K smaller than a critical value ($K_c \approx 0.031$), there are two stable patterns, C_{down} and C_{up} , with a maximum that corresponds with each one of the two roots of $F_{\text{eff}}(C)=0$, those where the slope is negative. Hence, above K_c , the critical value of K , only one stable pattern is found (C_{down}), with a maximum corresponding to the smaller of the roots of $F_{\text{eff}}(C)=0$. As before, C_{up} is induced by the noise while C_{down} is only modified by it.

We observe that a phase transition induced by noise only exists for K larger than the critical value K_c . In Fig. 6 the stable solutions for $K=0.03$ with and without noise are shown. While for the first case two patterns exist, for the second there is only one. The same is shown for $K=0.05$ in Fig. 7 where, for both cases, with and without noise, only one pattern exists.

IV. CONCLUSIONS

We have studied a simple deterministic model able to describe qualitatively pattern formation in a monolayer of adsorbed particles on surfaces with lateral attractive interactions [19,20]. This model leads to the formation of interfaces separating phases with different local (homogeneous) coverage for certain region of parameters [19]. We have chosen the parameters's values outside this region, where the system has only a monostable solution (homogeneous coverage). In order to study the interplay between noise and boundary con-

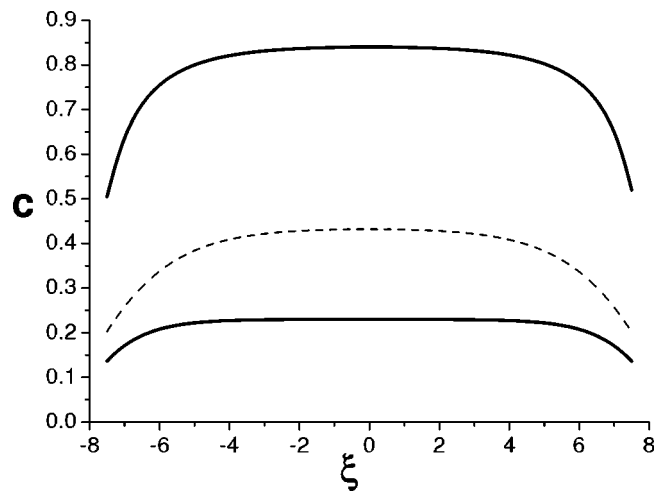


FIG. 6. Stationary stable patterns of the problem for $K=0.03$, with and without noise. We show C_{up} (above $C=0.5$) and C_{down} (below $C=0.5$) for $\lambda=\lambda_c \approx 0.005$ (full line) and C_{down} for $\lambda=0$. (dashed line). The other parameters values are the same as in Fig. 1.

ditions on the possibility of noise induced pattern formation, we add a multiplicative noise in such a way that a fluctuation-dissipation relation is fulfilled [24], and found the exact stationary solution of the Fokker-Planck equation corresponding to such a system.

We have explored different boundary conditions (Dirichlet, Neumann, and the more general albedo) for the modified model and have shown that when a source is introduced in the border, or more generally, when in addition we assume that the current is different from zero (albedo parameter below a critical value) a phase transition induced by noise occurs. In other words, when the multiplicative noise intensity does not exceed a certain critical value, the system has a unique stable pattern as solution. But, when the multiplicative noise intensity exceeds the critical value, in addition to the previous pattern (modified by the noise), the system has

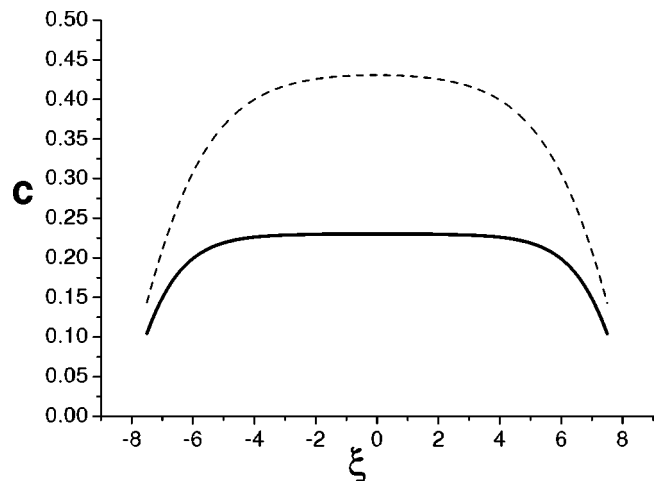


FIG. 7. Stationary stable patterns of the problem for $K=0.05$, with and without noise. We show C_{down} for $\lambda=\lambda_c \approx 0.005$ (full line) and C_{down} for $\lambda=0$. The other parameters values are the same as in Fig. 1.

another stable pattern as solution, with a nonhomogeneous (unstable) saddle separating both stable ones. It is worth remarking here that the second pattern arises abruptly, resembling a transition of the of first order type. Moreover, it is the multiplicative noise that induces this second pattern (as well as the unstable one). As indicated before, without multiplicative noise the solution corresponds to a unique pattern. We note that the necessary noise intensity is very small. In the other extreme, when the noise intensity is too large, the coverage escapes from the interval $(0, 1)$, and the results of the applied monolayer model loses its physical meaning. This case could correspond to a situation where it is necessary to resort to an extended description where a multilayer system is considered [20]. However, such a case was beyond the scope of this work.

To better visualize the indicated phase transition, we consider again Fig. 5 and in Fig. 8 we plot $H[C]$ vs K , where H is some measure of the pattern size. Here we choose

$$H[C] = \int_{-L/2}^{L/2} dx C(x).$$

It is apparent that for $K > K_c \sim 0.031$, the pattern indicated by C_{up} disappears abruptly, while the other stable pattern, C_{down} , remains. This indicates a kind of first order phase transition (or “subcritical-like bifurcation”).

An interesting point to be discussed is the possibility of carrying out an experimental observation of the above indicated phenomenon. It is known that up to a certain point it is possible both to control and vary the boundary conditions in chemical pattern forming experiments [27], as well as to introduce fluctuations in a controlled way [23,28]. However, if

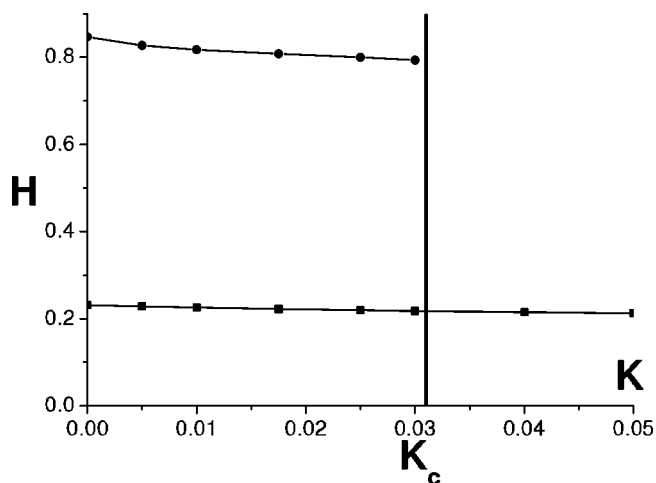


FIG. 8. H vs K for the same values of the parameters as in Fig. 5. We show $H(C_{up})$ (for $H > 0.5$) and $H(C_{down})$ (for $H < 0.5$).

a too fine tuning control is required by the above indicated interplay between noise and boundary conditions on noise induced pattern formation, it could be beyond the possibilities of such experimental systems. Another—maybe more—accessible possibility is to consider electronic experimental setups as has been used, for instance, for studying pattern formation and propagation, as well as some noise induced phenomena [29].

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