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Free boundary problem and its applications to reaction-diffusion systems of activator-inhibitor type

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Abstract

We present a brief overview of applications of free boundary problem to reaction-diffusion systems of activator-inhibitor type in the case when the inhibitor is long-ranged. We first discuss the physical origin of the considered class of reaction-diffusion systems and give a few examples. We then introduce the free boundary problem and its reduction that is relevant to the dynamics of interfacial patterns in the systems under consideration. Using the reduced free boundary problem, several cases are treated: self-replication of a single domain undergoing a morphological instability, breathing motion of a single domain on a disk, and slowly modulated waves of domain oscillations. In the first case the inhibitor is fast, and in the other two it is slow. In all cases, detailed information about the pattern dynamics can be obtained.

1 Introduction

In this paper, I will give a short overview of some of the applications of free boundary problem to interfacial patterns in reaction-diffusion systems. These systems play a fundamental role in understanding a variety of nonlinear phenomena observed in systems far from equilibrium [1, 6, 13, 18, 29, 34, 49]. Systems of reaction-diffusion equations serve as a general class of models with applications in physics, chemistry, and biology. Their pattern-forming capability was recognized half a century ago, starting with the pioneering work of Turing [62], and since then attracted an enormous amount of attention both in the modeling and mathematical community (for reviews, see, for example, [29, 35, 46, 64]).

An important class of reaction-diffusion models consists of systems of reaction-diffusion equations of activator-inhibitor type. This class of models in fact serves almost as a paradigm for pattern-forming systems of different nature. On one hand, these models are not overly complicated, which is inevitable for more realistic models of pattern-forming systems, and are therefore amenable to analysis. On the other hand, in many cases they can be systematically derived from the underlying physics of real pattern-forming systems and therefore be used for (at least) semi-quantitative explanations of patterns observed in experiments (see, for example, [27–29]).

In the simplest case, reaction-diffusion systems of activator-inhibitor type take on the following form:

$$\alpha u_t = \epsilon^2 \Delta u + f(u, v, A), \tag{1.1}$$

$$v_t = \Delta v + g(u, v, A). \tag{1.2}$$

Here $u = u(x, t)$ and $v = v(x, t)$ are scalar functions of time t and space $x \in \Omega$, where $\Omega \subseteq \mathbb{R}^n$ is a domain, both time and space are assumed to be suitably scaled; Δ is the n-dimensional Laplacian; f and g are nonlinear functions; ϵ is the ratio of the spatial scales and α is the ratio of the time scales of u and v, respectively, and A is a control parameter. If Ω is bounded, Eqs. (1.1) and (1.2) are also supplemented by no-flux boundary conditions. Note that in general nonlinearities can arise naturally in the diffusion terms as well [27, 29]. Here we will only consider the case of simple linear diffusion and concentrate on the effect of the nonlinearities in the reaction terms.

A pair of reaction-diffusion equations above is an activator-inhibitor system, with u and v being the activator and the inhibitor variables, respectively, if there exists a positive feedback with respect to u and

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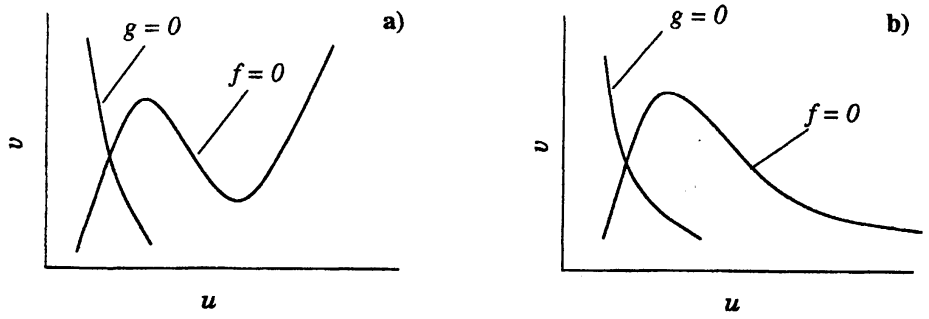

Figure 1: Two major types of the nullclines: N-systems (a) and Λ-systems (b).

a negative feedback between u and v. Mathematically, this can be expressed via the following inequalities obeyed by the nonlinearities f and g [26,29]:

$$f_u > 0 \quad \text{for some} \quad u, v, \tag{1.3}$$

$$g_v < 0 \quad \text{for all} \quad u, v, \tag{1.4}$$

$$f_v > 0,\ g_u < 0 \quad \text{or} \quad f_v < 0,\ g_u > 0 \quad \text{for all} \quad u, v. \tag{1.5}$$

The inequality in Eq. (1.3) means that small homogeneous increase in u may result in an increase in the production of u, the one in Eq. (1.4) implies that small homogeneous increase in v will result in a decrease in the production of v, and the ones in Eq. (1.5) imply that small homogeneous increase in u will result in such a change in the production of v that will result in the decrease in the production of u.

One can distinguish two major classes of nonlinearities f and g satisfying Eqs. (1.3) – (1.5). In the first class, for a given value of v the condition in Eq. (1.3) is satisfied only on single bounded interval of values of u. In the second class, the condition in Eq. (1.3) is satisfied on a semi-infinite interval of the values of u. It is not difficult to see that this implies that the nullcline of Eq. (1.1) (that is, the solution of equation $f(u, v, A) = 0$ in the u, v-plane) may be N- or И-shaped, while in the second case this nullcline may be V- or Λ-shaped (Fig. 1). Therefore, depending on the shape of the activator nullcline one can classify reaction-diffusion systems of activator-inhibitor type into N- or Λ-systems (the И- and V-systems are equivalent to the former, up to a change of variables). This classification was introduced by Kerner and Osipov in [25, 26, 28, 29]. It is not difficult to verify that the classical Brusselator model [48] is a Λ-system, the Gierer-Meinhardt model [14] is a V-systems, and the FitzHugh-Nagumo model [12,47] is an И-system. It turns out that the properties of Λ- and V-systems are fundamentally different from those of N- and И- systems [29].

In the following we will be considering only N-systems, since these are the systems that can generate interfacial patterns [29,43,44]. Note that N-systems correspond to the first case in Eq. (1.5). Furthermore, we will restrict ourselves to monostable systems. In other words, we will assume that the homogeneous state $u = u_h$, $v = v_h$, satisfying

$$f(u_h, v_h, A) = 0, \qquad g(u_h, v_h, A) = 0, \tag{1.6}$$

is unique for each value of A, and that furthermore the point $(u_h(A), v_h(A))$ in the (u, v) plane traces continuously the three monotonic branches of the activator nullcline. Physically, this means that the control parameter A measures the degree of nonequilibium in the system.

We will give an example of a derivation of a model of the considered type in a realistic physical context in the next section. Now, however, consider the following canonical example [29, 44, 64]:

$$f(u, v, A) = u - u^3 + v, \tag{1.7}$$

$$g(u, v, A) = A - u - v. \tag{1.8}$$

It is easy to see that in this case $f_u = 1 - 3u^2 > 0$ for $|u| < \frac{1}{\sqrt{3}}$ and negative otherwise, $f_v = 1$, $g_u = -1$, $g_v = -1$, so all the above conditions are satisfied. Furthermore the parameter A has the required property, since

$$u_h = A^{1/3}, \qquad v_h = -A^{1/3}(1 - A^{2/3}). \tag{1.9}$$

This homogeneous state is stable for all values of α and ϵ when $|A| > \frac{1}{3\sqrt{3}}$.

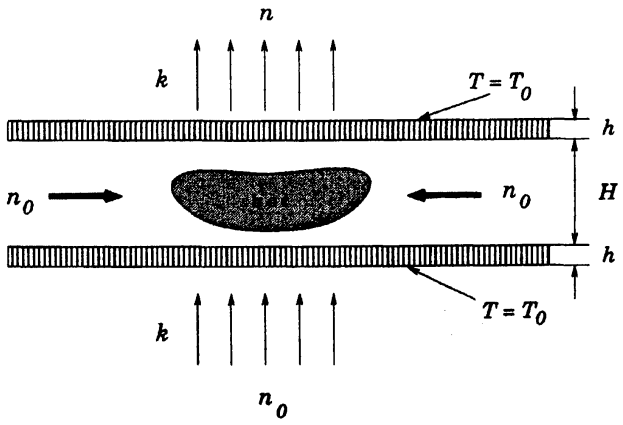

Figure 2: Combustion in a continuous flow reactor.

2 Example: system with uniformly generated combustion material

As a prototypical reaction-diffusion systems of activator-inhibitor type, consider a continuous flow reactor with porous walls inside which an exothermic reaction (e.g., combustion) takes place (Fig. 2). The reactor is placed between two porous walls of thickness h and is maintained at the ambient temperature T_0 on the outside. Gaseous fuel-oxidizer mixture is pumped with rate k (with the dimension of speed) through one of the walls into the reactor, and the products are removed from the other wall with the same rate. The reaction releases heat which is then absorbed by the reactor walls.

If one neglects the hydrodynamic effects, one can write down a system of reaction-diffusion equations describing the reaction inside the reactor (in three-dimensions) with appropriate boundary conditions on the reactor walls [65]. The situation may be simplified if the distance H between the reactor walls is small. In this case both the temperature and fuel concentration will vary little between the reactor walls. Then this system of equations can be averaged over the reactor thickness to obtain an effective two-dimensional reaction-diffusion system for the average concentration of fuel n and temperature T (in energy units) across the reactor:

$$\frac{\partial n}{\partial t} = D\Delta n + \frac{k}{H}(n_0 - n) - \nu n e^{-E_a/T}, \tag{2.1}$$

$$c\rho\frac{\partial T}{\partial t} = \kappa\Delta T + \nu n E e^{-E_a/T} - \frac{2\kappa_{wall}}{hH}(T - T_0). \tag{2.2}$$

Here, D is the diffusion coefficient of the fuel, κ is the thermal conductivity of the mixture and κ_{wall} is that of the walls (assumed to be small), h is the wall thickness; c is specific heat and ρ is the density of the mixture. The second term in the right-hand side of Eq. (2.1) is the supply and removal of the fuel with the incoming fuel concentration n_0, the last term in the right-hand side of this equation is the rate at which the fuel is consumed, this rate is characterized by the rate constant ν (for simplicity taken to be temperature-independent), the Arrhenius factor $e^{-E_a/T}$, where E_a is the activation energy, and we assumed a first-order reaction. Similarly, the second term in the right-hand side of Eq. (2.2) is the rate of heat release by the reaction, with E being the heat released in a single reaction, and the last term is the cooling rate. We assumed that the oxidizer is in excess and neglected the dependence of the transport coefficients on temperature. Note that this kind of model was introduced phenomenologically in [24,27,29,36].

Let us introduce the dimensionless variables $u = T/E_a$ and $v = n/n_0$ and scale time and length with H/k and $(DH/k)^{1/2}$, respectively. Then, Eqs. (2.1) and (2.2) can be reduced to the form of Eqs. (1.1)

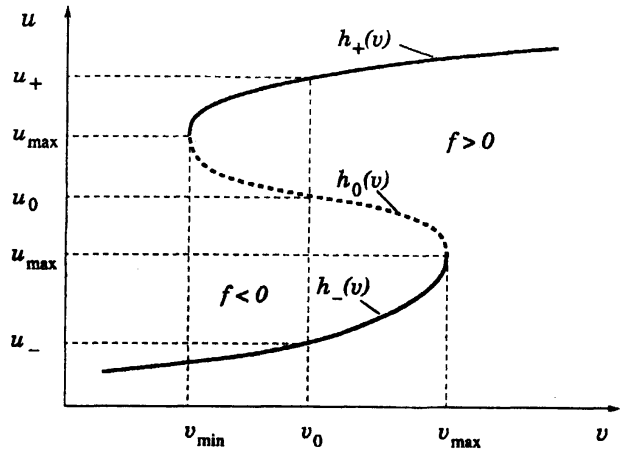

Figure 3: The activator nullcline (closeup).

and (1.2) with

$$f = Ave^{-1/u} + u_0 - u, \qquad g = 1 - v - \gamma v e^{-1/u}, \tag{2.3}$$

$$A = \frac{n_0 \nu h H E}{2\kappa_{wall} E_a}, \quad \gamma = \frac{\nu H}{k}, \quad u_0 = \frac{T_0}{E_a}, \tag{2.4}$$

$$\alpha = \frac{c\rho hk}{2\kappa_{wall}}, \qquad \epsilon = \sqrt{\frac{\kappa hk}{2\kappa_{wall} D}}. \tag{2.5}$$

It is not difficult to verify that the obtained nonlinearities are those of an N-system. The role of activator is played by the scaled temperature, and the role of inhibitor by the scaled fuel density. This is easy to understand: the thermo-activated character of the reaction results in the positive feedback with respect to temperature, while the consumption of fuel by this reaction gives a negative feedback. Furthermore, the parameter A represents the degree of deviation of the system from equilibrium, since it is proportional to the concentration n_0 of fuel being supplied; the more fuel is supplied the further away the system is from equilibrium. Then, under appropriate conditions this system can sustain spatiotemporal patterns. Physically, the simplest pattern in the form of a hot spot can be maintained by the balance of fuel consumption and supply through diffusion from the regions away from the spot (Fig. 2).

3 Free boundary problem

A free boundary problem arises in the analysis of Eqs. (1.1) and (1.2) in the asymptotic limits $\epsilon \to 0$ and/or $\alpha \to 0$ when the nullcline of Eq. (1.1) is N-shaped. Let us illustrate this for the case when both $\epsilon \to 0$ and $\alpha \to 0$ which we will concentrate upon in this paper. Clearly, in this limit the diffusion term in Eq. (1.1) can be set to zero on the time scale $O(1)$. Furthermore, for $\alpha \to 0$ and smooth initial conditions the value of u at each point x will immediately approach $h_\pm(v)$, where $h_+(v)$ and $h_-(v)$ are the two stable (in the sense of the ODE obtained from Eq. (1.1) with $\epsilon = 0$) branches of the activator nullcline (see Fig. 3). The choice between these two branches will be dictated by whether the initial condition for u at each point x lies above or below the unstable branch $h_0(v)$ of the nullcline; those points that lie below it will end up on $h_-(v)$, while those above on $h_+(v)$. Thus, the systems will instantly break up into regions $\Omega_\pm$ in which u is close to the branch $h_\pm(v)$, respectively, separated by the interface Γ, possibly disconnected (hence, the characterization of the state of the system as a *domain* pattern). Let us emphasize that on these time scales the location of Γ will be fixed by the initial conditions.

Motion of the interface will occur on the longer time scale following the interface formation after the initial transient. In order to understand it, we need to zoom in on the interface. Suppose that at time t_0 we have Γ passing through point x_0. Introducing

$$\tau = \frac{t - t_0}{\alpha}, \qquad \xi = \frac{x - x_0}{\epsilon}, \tag{3.1}$$

in the limit $\epsilon \to 0$ and $\alpha \to 0$ we will get

$$u_\tau = \Delta_\xi u + f(u, v(x_0, t_0), A), \tag{3.2}$$

where Δ_ξ is the Laplacian in the stretched variables, and we assumed that v does not significantly vary on the scales of τ and ξ. The problem becomes tricky, however, since the initial conditions for u in Eq. (3.2) dependent on α and ϵ. Indeed, these must be approaching $h_\pm(v(x_0, t_0))$ on the respective sides of Γ and have nearly flat level sets to be consistent with the discussion above. Now, it is known that at times $\tau \gg 1$ flat initial conditions of this type develop into traveling wave solutions $u(\xi, \tau) = \bar{u}(\hat{n} \cdot \xi - c\tau)$ moving with speed c in the direction of $\hat{n}$ (the unit normal vector to Γ pointing in the direction of Ω_-) [11]. Therefore, during interfacial motion the profiles of u in the neighborhood of Γ should be close to the solutions of

$$\bar{u}'' - c\bar{u}' + f(\bar{u}, v, A) = 0, \qquad \bar{u}(\pm\infty) = h_\pm(v), \tag{3.3}$$

in which v is treated as a constant. Small curvature introduces a correction to the propagation speed, equal to $-K$, where K is $(n-1)\times$ mean curvature of the interface, positive if Γ is convex when looked from Ω_+ (see, for example, [10]).

The free boundary problem is obtained from all this via a self-consistency argument. That is, we demand that at time t each point r on Γ move along the normal $\hat{n}(r,t)$ to Γ in the direction of Ω_- with the above-mentioned speed, which is now a function of the instantaneous value of the inhibitor $v(r,t)$ at point r on Γ and its curvature $K(r)$. The distribution of v, in turn, satisfies Eq. (1.2) in which $u = h_\pm(v)$ in $\Omega_\pm$, respectively. Going back to the original variables, we finally obtain

$$r_t = \alpha^{-1}\epsilon\hat{n}(r)[c(v(r,t), A) - \epsilon K(r)], \tag{3.4}$$

$$v_t = \Delta v + g(h_\pm(v), v, A), \quad f(h_\pm(v), v, A) = 0. \tag{3.5}$$

Note that this type of reduction of a system of reaction-diffusion equations to a free boundary problem goes back to Fife [9]. For reaction-diffusion systems of activator-inhibitor type in one dimension this reduction was used by Nishiura and Mimura [51] and by Ohta, Ito, and Tetsuka [54] to study the onset of oscillatory instabilities. Also, in higher dimensions this approach was first used by Ohta, Mimura, and Kobayashi for the stability analysis of localized patterns in a particular activator-inhibitor model [55].

Before concluding this section, let us mention that an important class of reaction-diffusion systems with the considered nonlinearities is characterized by small value of α and $\epsilon \to \infty$. By a simple rescaling, these so-called excitable systems are described by Eqs. (1.1) and (1.2), with the diffusion term being absent in the second equation. These systems are capable of supporting various types of wave patterns, including solitary waves, wave trains, spiral waves, and more complex autowave patterns (see, for example, [18, 35, 63, 64]). Free boundary methods have been successfully used for the analysis of a number of problems in these systems. In particular, in weakly excitable systems the free boundary problem reduces to the analogue of a single Eq. (3.4) (since v is always close to v_h in this situation); in this case a nice characterization of motion of the interface in the plane was introduced by Brazhnik, Davydov, and Mikhailov [4, 35]. Using this approach, Brazhnik constructed exact solutions of the free boundary problem for V-shaped waves [3]. Also, Karma worked out the asymptotics of the spiral wave solutions for N-systems both in the weakly and strongly excitable case [19, 20].

4 Reduced free boundary problem

The free boundary problem in Eqs. (3.4) and (3.5) represents a significant reduction of complexity by decreasing the effective dimensionality of the problem. Nevertheless, it is still a fundamentally nonlinear problem. In addition to the basic nonlinearity associated with the coupling between the shape of Γ and the inhibitor field v, which determines the evolution of Γ, two other sources of nonlinearity exist. The first is the dependence $c(v, A)$ in Eq. (3.4). This dependence is obtained by solving the nonlinear eigenvalue problem in Eq. (3.3). Phase plane analysis shows that this equation gives a unique value of c for each given $v_{\min} < v < v_{\max}$ (see, for example, [11]). Furthermore, for smooth nonlinearities the speed c is bounded and changes continuously from a negative value for $v \to v_{\min}$, since $f(u, v_{\min}, A) \le 0$ for all values of $u \ge h_-(v_{\min})$ (recall that in N-systems $f_v > 0$), to a positive value of c for $v \to v_{\max}$, since now $f(u, v_{\max}, A) \ge 0$ for all $u \le h_+(v_{\max})$, see also Fig. 3. Moreover, there is a unique value of

$v = v_0(A)$ at which the speed c is zero. The value of v_0 can be found from the algebraic equation [9] (see also [21,22,29,37])

$$\int_{u_-}^{u_+} f(u, v_0, A)du = 0, \tag{4.1}$$

where $u_\pm = h_\pm(v_0)$. Uniqueness of v_0 follows from the monotonic dependence of the integral in Eq. (4.1) on v. For example, in the exactly solvable case of the nonlinearity in Eq. (1.7) the speed c satisfies implicitly $c - \frac{2}{9}c^3 = \frac{3}{\sqrt{2}}v$; in this case $v_0 = 0$, $u_\pm = \pm 1$, $v_{\min} = -\frac{2}{3\sqrt{3}}$ and $v_{\max} = \frac{2}{3\sqrt{3}}$ [38]. The corresponding front solution has the form of a domain wall connecting u_- with u_+ [9,21,22,29,37] .

The second source of nonlinearity mentioned above is the functions $h_\pm(v)$. Note that these functions are only defined for $v \geq v_{\min}$ and $v \leq v_{\max}$, respectively. Therefore, if during the evolution of Γ the value of v reaches these critical values, a jump of the value of u from one branch of the nullcline to the other must occur. This phenomenon was termed *local breakdown* by Kerner and Osipov [23,28,29] and in the language of the free boundary problem amounts to the creation of new interface.

The two nonlinear aspects mentioned above can be eliminated if $v(x,t)$ deviates little from v_0 for all x. Clearly, this should be the case for $v(r,t)$ in a stationary pattern, see Eq. (3.4). In fact, this is a generic situation for stable stationary patterns in dimensions $n \geq 2$ [39–44,53]. Introducing

$$\tilde{v} = v - v_0, \tag{4.2}$$

we can linearize both the dependence $c(v,A)$ and $h_\pm(v)$ around v_0, if $|\tilde{v}| \ll 1$. As a result, to the leading order we obtain [38,40]:

$$c = b\tilde{v}, \tag{4.3}$$

where

$$b = \frac{\int_{u_-}^{u_+} f_v(u, v_0, A)du}{\int_{u_-}^{u_+} \sqrt{-2U(u, v_0, A)}du}, \qquad U(u, v_0, A) = \int_{u_-}^{u} f(s, v_0, A)ds. \tag{4.4}$$

Note that for N-systems we have $b > 0$. Similarly, Eq. (3.5) simplifies to [38,40]

$$\tilde{v}_t = \Delta\tilde{v} - (c_+\chi_+ + c_-\chi_-)\tilde{v} + g(u_-, v_0, A) - a\chi_+ \tag{4.5}$$

where $\chi_\pm$ are the characteristic functions of $\Omega_\pm$, and

$$a = g(u_-, v_0, A) - g(u_+, v_0, A), \tag{4.6}$$

$$c_\pm = -g_v(u_\pm, v_0, A) + \frac{g_u(u_\pm, v_0, A)f_v(u_\pm, v_0, A)}{f_u(u_\pm, v_0, A)}, \tag{4.7}$$

Observe that by Eqs. (1.3) - (1.5), and the fact that $f_u(u_\pm, v_0, A) < 0$ (recall that $u_\pm$ lie on the stable branches of the nullcline) for N-systems we have $a > 0$ and $c_\pm > 0$. For example, in the case of the nonlinearities in Eqs. (1.7) and (1.8) it is not difficult to check that $a = 2$, $b = \frac{3}{\sqrt{2}}$, $c_\pm = \frac{3}{2}$.

Thus, the reduced free boundary problem is obtained from Eq. (3.4) together with Eqs. (4.3) and (4.5). To proceed, we need to specify the relationship between the parameters ϵ, α, and A, as well as the type of pattern. Before we consider several particular cases, let us investigate the balance of different terms in this free boundary problem. The crucial feature of reaction-diffusion systems of activator-inhibitor type is that they are capable to support *autosolitons* — self-sustained inhomogeneous localized patterns [29]. In N-systems in dimensions $n \geq 2$, an autosoliton is a single radially-symmetric domain Ω_+ (or Ω_-) embedded into the whole of $\mathbb{R}^n$. If R is the radius of Ω_+ and T is the time scale of its variation, we obtain that different terms in Eqs. (3.4) and (4.5) together with Eq. (4.3) balance each other if

$$\frac{R}{T} \sim \frac{\epsilon\tilde{v}}{\alpha} \sim \frac{\epsilon^2}{\alpha R}, \qquad \frac{\tilde{v}}{T} \sim \frac{\tilde{v}}{R^2} \sim 1. \tag{4.8}$$

where here and below "$\sim$" denotes an order of magnitude, and we took into account that $|\tilde{v}| \ll 1$. Note that this requires that $g(u_-, v_0, A)$ be small, which implies that u_- and v_0 are close to the homogeneous state u_h, v_h. Let us define the values of $A_b^\pm$ to be the solutions of

$$g(u_\pm(A_b^\pm), v_0(A_b^\pm), A_b^\pm) = 0, \tag{4.9}$$

where we recalled that v_0 and $u_\pm$ are in general all functions of A. We will assume that $A_b^\pm$ exist and are also unique. The values of $A_b^\pm$ have the meaning of the critical values of A at which the formation of localized $\Omega_\pm$ patterns is possible [29]. Therefore, by continuity the value of A must be close to the value of A_b^- for a small Ω_+ domain to be able to form (the same argument applies to localized Ω_- patterns, with A_b^+ instead). For example, in the case of the nonlinearities in Eqs. (1.7) and (1.8) we have $A_b^\pm = \pm 1$.

One can see that the relations in Eq. (4.8) can be satisfied only if

$$R \sim \epsilon^{1/3}, \quad T \sim \alpha\epsilon^{-4/3}, \quad \alpha \sim \epsilon^2. \tag{4.10}$$

The consistency of these arguments is verified by checking that indeed $\tilde{v} \sim R^2 \sim \epsilon^{2/3} \ll 1$. The estimate in Eq. (4.10), although rather crude, allows to make a number of conclusions about the free boundary problem in Eqs. (3.4), (4.3), and (4.5). First, it identifies the precise scaling for the length scale of the domain patterns. In fact, this scaling was first obtained from the stability considerations of the localized patterns in N-systems in dimensions $n \geq 2$ [43,44], and from the energetic considerations in the case of extended domain patterns [39,42]. Second, it gives the scaling for α at which the delay in the response of the inhibitor becomes significant; this scaling has also been obtained from the stability considerations for the localized pattern [43], however, the situation is more complicated for extended patterns (see below). Also note that the situation is qualitatively different in one dimension, in which generically $R \sim 1$ and $\alpha \sim \epsilon$ for the effect of delay to appear [24,29,30,50,51,56].

5 Applications

We are now going to consider a few applications of the reduced free boundary problem which allow to get major insights into the nonlinear dynamics of interfacial patterns in reaction-diffusion systems of activator-inhibitor type. For simplicity, we will restrict ourselves to two-dimensional patterns, the results remain qualitatively the same for all $n > 2$. Throughout the entire discussion below, we assume that $\epsilon \ll 1$.

5.1 Self-replication

We first consider the situation in which A is close to A_b^- and the inhibitor is fast, that is $\alpha \gg \epsilon^2$ (compare with Eq. (4.10)). This means that during interfacial motion the inhibitor will quickly equilibrate to reach a quasi steady-state, so to the leading order the time derivative in Eq. (4.5) can be neglected. Furthermore, if the size of Ω_+ is small, we can neglect the term $c_+\chi_+$ in this equation as well. Then Eq. (4.5) can be easily solved with the help of Green's function:

$$\tilde{v}(x) = \frac{g(u_-, v_0, A)}{c_-} - \frac{a}{2\pi}\int_{\Omega_+} K_0(\sqrt{c_-}|x - x'|)dx', \tag{5.1}$$

where $K_0(x)$ is the modified Bessel function of the second kind, and we assumed that $\Omega = \mathbb{R}^2$. This integral can be further reduced to a line integral over Γ. In the present context this reduction was first performed by Goldstein, Muraki, and Petrich in the case of a reaction-diffusion system with weak activator-inhibitor coupling (which are necessarily bistable) [15,57]. A simple calculation shows that if r is a point on Γ, we have

$$\tilde{v}(r) = \frac{g(u_-, v_0, A)}{c_-} + \frac{a}{2\pi\sqrt{c_-}}\oint_\Gamma \left(K_1(\sqrt{c_-}|r - r'|) - \frac{1}{\sqrt{c_-}|r - r'|}\right)\frac{(r' - r)\times dr'}{|r' - r|}, \tag{5.2}$$

where $K_1(x)$ is the modified Bessel function of the second kind, "$\times$" denotes the cross product, and Γ is assumed to be oriented counterclockwise with respect to Ω_+. Now, rescaling length and time according to Eq. (4.10):

$$x \to \frac{\epsilon^{1/3}}{a^{1/3}b^{1/3}}x, \qquad t \to \frac{\alpha}{\epsilon^{4/3}a^{2/3}b^{2/3}}t, \tag{5.3}$$

and expanding the Bessel function in $\epsilon^{1/3}$, to the leading order Eq. (3.4) becomes [38]

$$r_t = \hat{n}(r)\left(-K(r) + \delta + \frac{1}{4\pi}\oint_\Gamma (-\Lambda + \ln|r - r'|)(r' - r)\times dr'\right), \tag{5.4}$$

$$\delta = \frac{b^{2/3} g(u_-, v_0, A)}{\epsilon^{2/3} a^{1/3} c_-}, \qquad \Lambda = \frac{1}{3} \ln \epsilon^{-1} + \frac{1}{3} \ln ab - \frac{1}{2} \ln c_- + \ln 2 - \gamma + \frac{1}{2}, \tag{5.5}$$

and $\gamma \simeq 0.5772$ is the Euler constant; Λ is a large logarithm. Note that a related free boundary formulation was discussed by Nishiura and Onishi [52].

To understand the significance of different terms in Eq. (5.4), let us consider an ideally round domain Ω_+ first. Then, Eq. (5.4) reduces to a single ODE for the domain radius ρ [41]

$$\frac{d\rho}{dt} = -\frac{1}{\rho} + \delta + \frac{1}{4}\rho^2 (-2\Lambda + 2\ln\rho + 1). \tag{5.6}$$

This formula is valid when ρ is not very large, when Λ is the dominant term in the right-hand side of Eq. (5.6). The analysis of this equation then shows that if the value of δ is negative in this range of ρ all radially-symmetric domains will shrink to zero in finite time. On the other hand, when δ becomes sufficiently large positive, two equilibria with radii $\rho_u < \rho_0$ appear, with ρ_u being unstable and ρ_0 stable (see also [42]). Furthermore, the radius ρ_0 is uniquely determined by δ and is $O(1)$ for $\delta = O(\Lambda)$. Note that this is precisely the solution in the form of an autosoliton.

A fundamental feature of reaction-diffusion systems of activator-inhibitor type in dimensions $n \geq 2$ is that patterns can undergo morphological instabilities resulting in shape changes [15,29,43–45,55,58]. This can readily be seen, if one perturbs Eq. (5.4) around a radially stable disk-shaped domain of radius ρ_0. Introducing polar coordinates (ρ, φ) for Γ and substituting $\rho(\varphi, t) = \rho_0 + \rho_m e^{im\varphi - \gamma_m t}$ into Eq. (5.4), we then take a limit of $\rho_m \to 0$ to obtain [41–43]

$$\gamma_m = \frac{m^2 - 1}{\rho_0^2} - \frac{\rho_0}{2}\left(1 - \frac{1}{m}\right), \tag{5.7}$$

which is valid for $m > 0$. From this equation follows that the radially-symmetric pattern undergoes the distortion instability corresponding to $m = 2$ when $\rho_0 > \rho_{c2} = (12)^{1/3}$.

Qualitatively, one can understand this morphological instability as follows. When $\rho \sim 1$, both δ and Λ in Eq. (5.4) are large. Then, since Λ multiplies the integral which gives the area of Ω_+, the effect of these two terms is basically to preserve the pattern's overall area. In contrast, morphological instabilities preserve the area and are therefore driven by the $\ln|r - r'|$ term in Eq. (5.4). Observe that this term can be positive if $|r - r'|$ is large and negative when $|r - r'|$ is small. Therefore, its effect will be to pull apart distant pieces of the interface. If the size of the pattern is large, then the stabilizing effect of curvature would not be able to compensate this tendency, and a pattern with complex morphology will develop [38,43,44].

The next question here is whether the pattern growing as a result of this instability will preserve its topology, namely, whether it will remain connected during the course of its evolution. Numerical simulations of reaction-diffusion systems with weak activator-inhibitor coupling [16], as well as simulations of the free boundary problem for these systems [8,15,57] showed that a connected labyrinthine pattern formed as a result of the morphological instability of the disk-shaped domain.

To test this question in the systems under consideration, we performed a numerical solution of Eq. (5.4). The interface was discretized as a piecewise-linear curve, integration in space was done using midpoint rule, curvature at a given point was obtained by drawing a circle through that point and its two neighbors to obtain an approximation for the curvature radius. The number of discretization points was controlled adaptively to maintain the distance between the neighboring points within a factor of two. In our numerical method, we also allowed Γ to reconnect when the interfaces came sufficiently close together. The result of a typical simulation is shown in Fig. 4. The initial condition is taken as a slightly perturbed disk-shaped domain of radius $\rho_0 > \rho_{c2}$. One can see that the domain undergoing instability pinches off and divides, the daughter domains keep undergoing divisions. Thus, in N-systems with fast inhibitor self-replication can be observed as a result of the morphological instability. This is also confirmed in the numerical simulations of the original reaction-diffusion equations with sufficiently small ϵ [41,42]. Let us point out that the numerical solution of the free boundary problem could run only until a certain time, after which the domains started to blow up, so only a few acts of self-replication could be observed. We attribute this to the fact that the replicating domains go so far apart that the expansion used to obtain

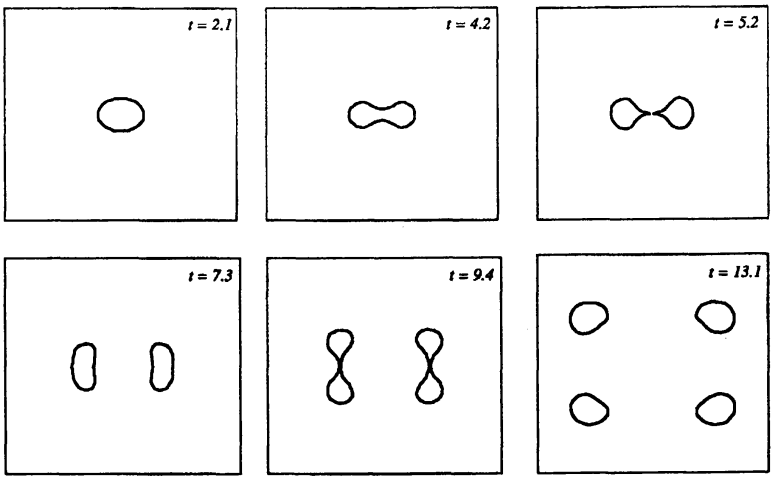

Figure 4: Self-replication of a spot. Result of the numerical solution of Eq. (5.4). The box is 50×50. Other parameters are: $\Lambda = 4$, $\delta = 18.3$, based on the model in Eq. (1.7) an (1.8) with $\epsilon = 10^{-4}$, $A - A_b^- = 0.0455$.

Eq. (5.4) from Eq. (5.2) breaks down. When the domains become separated by distances of order 1 (in the original variables), one needs to keep the full Bessel function (which decays exponentially at large distances) in the integral in Eq. (5.4). Note that both the phenomenon of self-replication and formation of labyrinthine patterns were observed experimentally in [31,32].

5.2 Breathing spot

We now turn to the study of the effect of the delay in the inhibitor response to the motion of the interface. To do that consider the following simple setup. Let Ω be a disk of diameter $L \ll 1$ (but, of course, $L \gg \epsilon$) and consider a radially-symmetric pattern Ω_+ with radius $\rho(t)$ in Ω. Since Ω is small, the spatial variation of v will be small across Ω. Therefore, for a stationary pattern we will once again have $|\tilde{v}| \ll 1$, so the reduced free boundary problem of Sec. 4 should work.

Equation (3.4) in the considered case becomes (we are back to the unscaled variables)

$$\frac{d\rho}{dt} = -\frac{\epsilon^2}{\alpha\rho} + \frac{\epsilon b\tilde{v}(\rho,t)}{\alpha}. \tag{5.8}$$

To proceed, let us break up $\tilde{v}$ as follows

$$\tilde{v} = \bar{v} + \hat{v}, \tag{5.9}$$

where $\bar{v}$ is the value of $\tilde{v}$ averaged over Ω. It turns out that for $\alpha \gg \epsilon^2$ the dynamics of $\bar{v}$ and $\tilde{v}$ are qualitatively different. Averaging Eq. (4.5) over x, we obtain

$$\frac{d\bar{v}}{dt} = -\left(\frac{(L^2-4\rho^2)c_-}{L^2} + \frac{4\rho^2 c_+}{L^2}\right)\bar{v} - \frac{4a(\rho^2-\rho_0^2)}{L^2}, \tag{5.10}$$

where

$$\rho_0 = \frac{L^2 g(u_-, v_0, A)}{4a}. \tag{5.11}$$

In writing Eq. (5.10), we neglected the term coming from $\hat{v}$, which is going to be small compared to the last term in the right-hand side of this equation in the considered regime (see below).

To close this system of equations, we need to find $\hat{v}(\rho,t)$. The equation for $\hat{v}$ is obtained by subtracting Eq. (5.10) from Eq. (4.5). If we also assume that $\alpha \gg \epsilon^2$, by the argument similar to the one in Eq. (4.10) we will see that the time derivative of $\hat{v}$ is small and can be dropped from this equation. Similarly, the

value of $\hat{v}$ should come out to be of order L^2 and therefore will be small compared to the last term in Eq. (4.5). With all these reductions, to the leading order the equation for $\hat{v}$ becomes

$$\frac{d^2\hat{v}}{dr^2} + \frac{1}{r}\frac{d\hat{v}}{dr} - a(\chi_+ - \langle\chi_+\rangle) = 0, \qquad \langle\hat{v}\rangle = 0, \tag{5.12}$$

where r is the radial coordinate and $\langle . \rangle$ denotes spatial average. This equation can be trivially solved to obtain

$$\hat{v}(\rho) = a\left(\frac{3\rho^2}{8} - \frac{3\rho^4}{2L^2} + \frac{\rho^2}{2}\ln\frac{2\rho}{L}\right). \tag{5.13}$$

Let us introduce the following quantities

$$\bar{\rho} = \frac{\rho}{L}, \quad \omega_0 = \sqrt{\frac{4\epsilon ab}{\alpha L}}, \quad \tau = \frac{\alpha}{\epsilon abL}, \quad \bar{\epsilon} = \frac{\epsilon}{abL^3}. \tag{5.14}$$

Assuming that $\omega_0 \gg 1$, while $\tau \sim 1$, we can further reduce Eqs. (5.8), (5.10), and (5.13) by eliminating $\bar{v}$ to [40]

$$\frac{d^2\bar{\rho}}{dt^2} + \beta(\bar{\rho})\frac{d\bar{\rho}}{dt} + \omega_0^2(\bar{\rho}^2 - \bar{\rho}_0^2) = 0, \tag{5.15}$$

where

$$\beta(\bar{\rho}) = c_- + 4(c_+ - c_-)\bar{\rho}^2 - \tau^{-1}\left(\frac{\bar{\epsilon}}{\bar{\rho}^2} + \frac{5\bar{\rho}}{4} - 6\bar{\rho}^3 + \bar{\rho}\ln 2\bar{\rho}\right). \tag{5.16}$$

We now demand that $\bar{\epsilon}$ and $\bar{\rho}_0$ are $O(1)$ quantities, which implies that

$$L \sim \epsilon^{1/3}, \qquad \alpha \sim \epsilon^{4/3}. \tag{5.17}$$

Equation (5.16) then describes an equation for a weakly damped nonlinear oscillator with nonlinear friction coefficient $\beta(\bar{\rho})$. A complete study of this dynamical system is possible (for a detailed analysis, see [40]). Importantly, in a wide range of $\bar{\rho}_0$ (implying a range of values of A between A_b^- and A_b^+) a Hopf bifurcation of the stationary solution with $\bar{\rho} = \bar{\rho}_0$ is realized at $\tau = \tau_c$, where to the leading order [40]

$$\tau_c = \frac{4\bar{\epsilon} + 5\bar{\rho}_0^3 - 24\bar{\rho}_0^5 + 4\bar{\rho}_0^3\ln 2\bar{\rho}_0}{4\bar{\rho}_0^2(c_- + 4(c_+ - c_-)\bar{\rho}_0^2)}, \tag{5.18}$$

which is obtained by simply equating $\beta(\bar{\rho}_0)$ to zero (recall that $\omega_0 \sim \epsilon^{-1/3} \gg 1$). We emphasize that these results are completely independent of the original nonlinearities f and g. Let us also point out that this behavior was experimentally observed in [17].

5.3 Phase waves

Lastly, we will discuss the problem of describing the interaction of different domains in a multidomain pattern. This is a fundamental problem in the theory of reaction-diffusion equations of activator-inhibitor type, since typically the patterns that form in these systems are extended [41,42,44]. The main obstacle in doing this is the fact that generally the patterns forming under these conditions are highly disordered [41,42]. Another problem is that different domains interact with each other on different length scales: the local arrangement of domains in a stable stationary pattern is due to the short-range interaction between nearest neighbors, while the long-range interactions at distances ~ 1 are responsible for the screening effects (see, for example, Eqs. (3.4) and (5.2)).

This problem can be simplified if one looks at perturbations (not necessarily small) of stationary periodic patterns. In the following we will consider a pattern in the form of (nearly-) circular domains arranged in a hexagonal lattice of period $\mathcal{L}_p$ as an example. Unless A is close to $A_b^{\pm}$, these patterns are stable only when $\mathcal{L}_p \sim \epsilon^{1/3}$ [39,41,42]. Numerical simulations show that for small enough α these patterns can support phase waves of oscillations of the domain radii (Fig. 5). It is observed that these

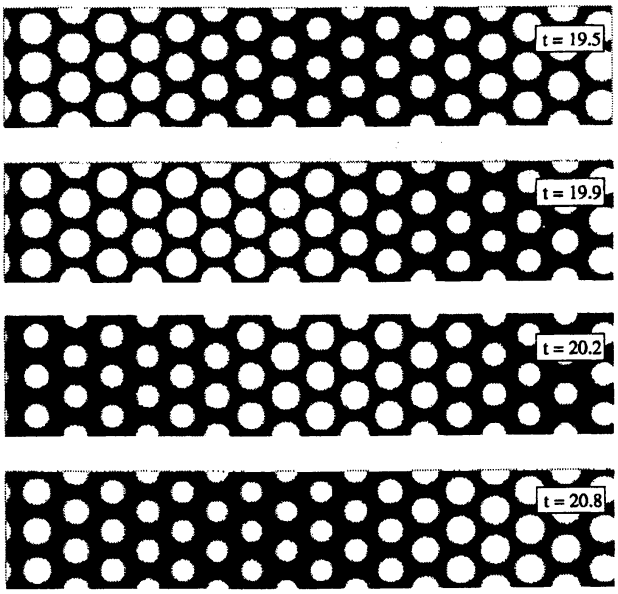

Figure 5: Propagation of the wave of collective oscillations of the domain radii obtained from numerical solution of Eqs. (1.1) and (1.2) with the nonlinearities given by Eq. (1.7) and (1.8) and $\epsilon = 0.05$, $\alpha = 0.02$, and $A = -0.4$. The domain is 20×4. The boundary conditions are periodic.

waves represent slow modulations in space of the periodic breathing motion of individual domains [40]. Note that these patterns are also realized in experiments [7].

Once again, since in a periodic pattern of small period the deviation of v from v_0 is small, the reduced free boundary problem of Sec. 4 can be applied. Moreover, for not very large domain radii compared to $\mathcal{L}_p/2$ (in practice, for virtually all radii) the deviations of the domain shape from that of an ideal disk can be neglected. Then, to obtain effective equations that describe the slowly varying modulations of the domain radii, we introduce the function $\rho(x,t)$, which is the coarse-grained (smoothed) version of the radius of an individual domain in a given hexagonal cell. We then separate $\tilde{v}$ as in Eq. (5.9), where now $\bar{v}$ is defined as the coarse-grained version of $\tilde{v}$, that is, we take $\bar{v}$ to be the average of $\tilde{v}$ over an area of the size which is much greater than $\mathcal{L}_p$ but smaller than the length scale of variation of ρ. To the leading order the equation for $\bar{v}$, now also slowly varying in space, becomes

$$\frac{\partial \bar{v}}{\partial t} = \Delta \bar{v} - \left(\frac{(L^2 - 4\rho^2)c_-}{L^2} + \frac{4\rho^2 c_+}{L^2} \right) \bar{v} - \frac{4a(\rho^2 - \rho_0^2)}{L^2}, \tag{5.19}$$

where ρ_0 is as in Eq. (5.11) and

$$L = 3^{1/4} 2^{1/2} \pi^{-1/2} \mathcal{L}_p, \tag{5.20}$$

so that $\pi L^2/4$ is equal to the area of a single hexagonal cell. Now, since $\hat{v}$ should be small, slow variations of $\hat{v}$ between different cells will be a higher-order correction and can be neglected. Therefore, to the leading order we can solve the problem for $\hat{v}$ in each hexagonal cell with no-flux boundary conditions. Assuming that $\alpha \gg \epsilon^2$ and following the argument of the section above, we obtain the following problem for $\hat{v}$ in the hexagonal cell Ω_0 for each ρ:

$$\Delta \hat{v} - a(\chi_+ - \langle \chi_+ \rangle) = 0, \qquad \langle \hat{v} \rangle = 0, \qquad \left. \frac{\partial \hat{v}}{\partial n} \right|_{\partial \Omega_0} = 0, \tag{5.21}$$

where now $\langle . \rangle$ denotes averaging over Ω_0. To finally close this system of equations, we use the average value of $\hat{v}$ over the domain's interface in Eq. (5.8):

$$\frac{\partial \rho}{\partial t} = -\frac{\epsilon^2}{\alpha \rho} + \frac{\epsilon b}{\alpha} \left(\bar{v} + \frac{1}{2\pi} \int_0^{2\pi} \hat{v}(\rho, \varphi) d\varphi \right), \tag{5.22}$$

where the variations of $\bar{v}$ across Ω_0 were neglected and $\hat{v}$ on Γ was written in terms of the polar coordinates (ρ, φ) of the interface. Note that it should also be possible to obtain these equations using homogenization techniques. Also note that the deviations from the ideal disk shape for each domain can be taken into account by writing the position of the interface as $r(\varphi, t) = \rho(t) + \sum_{m=1}^{\infty} \rho_{6m}(t) \cos(6m\varphi)$ in polar coordinates and modifying Eq. (5.22) accordingly, together with obtaining equations for each ρ_{6m}.

The main difficulty in analyzing the equations above must be the solution of Eq. (5.21), which has to be done in the hexagonal geometry. This problem, however, can be handled approximately by the Wigner-Seitz method from solid state physics (see, for example, [66]). Namely, instead of solving Eq (5.21) in Ω_0, we will solve it on a disk of the same area. This immediately allows us to use the results of the previous section. Once again, introducing the constants from Eq. (5.14) and following the same steps, we now arrive at the following effective PDE for $\bar{\rho}(x,t)$ [40][1]

$$\frac{\partial^2 \bar{\rho}}{\partial t^2} + \omega_0^2(\bar{\rho}^2 - \bar{\rho}_0^2) = -(\beta(\bar{\rho}) - \Delta)\frac{\partial \bar{\rho}}{\partial t}, \tag{5.23}$$

where $\beta(\rho)$ is still given by Eq. (5.16). Note that the spatially-independent solutions of this equation (which correspond to in-phase oscillations of all domains) are equivalent to the oscillations of a single domain considered in the previous section. Therefore, this means that hexagonal patterns also undergo a Hopf bifurcation leading to the onset of synchronous breathing motion [40]. One can further investigate the dynamic coupling between different domains. One striking observation here is that while locally the domain oscillations synchronize, globally they can exhibit chaotic behavior [40]. Let us also point out that similar treatment of this type of patterns is possible for a variety of geometries, including one dimension, where a rather complete study of the pattern's dynamics is possible [40].

6 Conclusion

To summarize, we have presented an overview of recent results on the applications of free boundary problem to reaction-diffusion equations of activator-inhibitor type. This overview was concentrated on the case of long-ranged and slow inhibitor ($\epsilon \ll 1$ and $\epsilon^2 \ll \alpha \ll 1$). The techniques we used for obtaining the free boundary reduction of the original reaction-diffusion equations were based on formal asymptotics and heuristic arguments. Let us point out that most of the results obtained in Sec. 5 can be systematically derived via formal asymptotic expansions of the original reaction-diffusion problem by assuming the appropriate scaling relationships between ϵ, α, and A, in the limit $\epsilon \to 0$. However, we chose to use a more intuitive approach, since it provides insights into the origins of these scaling relations. We also chose to present the cases which lead to interesting phenomenology; other regimes can be studied using similar methods.

To the best of our knowledge, rigorous work on the problems discussed above is quite recent. We first would like to mention the work of Soravia and Souganidis who obtained free boundary reductions for a subset of reaction-diffusion systems of activator-inhibitor type in $\mathbb{R}^n$ that are valid globally in time [61]. See also this work for the list of references on existence of solutions to the interfacial problem. More recently, Bonami, Hilhorst, and Logak analyzed a different scaling regime for a particular model [2, 33]. Their free boundary problem is essentially a bounded Ω version of the problem considered in Sec. 5.1 (except for the Dirichlet boundary conditions for $\tilde{v}$). A different scaling regime was investigated by Sakamoto, with the results along the lines of the discussion at the beginning of Sec. 3 [60].

Let us also point out that in the case of fast inhibitor the reduced free boundary problem possesses an energy functional [41, 42]. This allows to make a number of further conclusions about the dynamics of the interfaces and, in particular, about the stable stationary patterns, which are now local minimizers of the energy. For a certain scaling, these local minimizers were studied by Ren and Wei in the context of Γ-convergence [59]. Also, Choksi obtained precise scaling for the global energy minimizers [5]. Let us point out that these authors essentially consider bounded domains whose size shrinks to zero in the limit $\epsilon \to 0$. This is different from the situation of $\Omega = \mathbb{R}^n$ considered in Sec. 5, where two length scales: one of single domain size, $O(\epsilon^{1/3})$, and the other of screening effects, $O(1)$, exist (naturally, the third

[1] This equation improves slightly the result of [40], where L was chosen to be simply equal to $\mathcal{L}_p$ in reducing Eq. (5.21) to the radial problem.

length scale $O(\epsilon)$ which corresponds to the interfacial thickness, present in the full system of PDEs, does not enter the free boundary problem). This must lead to homogenization-type problems and is currently open. In particular, one problem of interest here is the dynamics of patterns with slowly modulated morphologies.

The case of slow inhibitor obeying the scaling from Eq. (5.17) and similar cases has not been analyzed rigorously yet. Here the cases of breathing patterns and perturbations of periodic patterns considered in Secs. 5.2 and 5.3 seem promising. More generally, a fundamental problem in this context is to understand similar phenomena of collective oscillations for disordered domain patterns. In this case it is not even clear what the starting point for the analysis of such patterns of large numbers of interacting domains should be. One step in that direction was made in [40], where a shadow limit of the reduced free boundary problem with the initial conditions in the form of a polydisperse mixture of radial droplets was considered; the dynamics of the pattern on a certain time scale could then be analyzed via a statistical description involving the distribution of the droplets' radii.

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