Domain stability, competition, growth, and selection in globally constrained bistable systems

Baruch Meerson
Racah Institute of Physics, Hebrew University of Jerusalem, Jerusalem 91904, Israel

Pavel V. Sasorov
Institute of Theoretical and Experimental Physics, Moscow 117259, Russia

(Received 8 May 1995; revised manuscript received 24 October 1995)

A general globally constrained reaction-diffusion equation is suggested that describes formation of equilibrium domains and competition between domains in bistable media. We study the stability properties of domains (strips and perfect circlular or spherical "drops") in one, two, and three dimensions. The dynamics of the distribution function (DF) of many drops with respect to their radii is mapped into a mean-field model of Ostwald ripening. A family of similarity solutions for the DF is found, and the long-standing problem of selection rule for the "correct" asymptotic DF is solved.

PACS number(s): 47.54.+r, 64.60.My, 64.60.Cn

Many pattern formation phenomena outside of equilibrium originate in the interplay between local and global dynamics and diffusion [1,2]. As an example, consider a bistable medium, describable by a dimensionless scalar reaction-diffusion equation (RDE):

$$\frac{\partial u}{\partial t}=\nabla^2 u+f(u,p), \tag{1}$$

where $u(\mathbf{r},t)$ is an order parameter and p is an additional parameter (inhibitor). We assume that the characteristic dimension of the system $L\gg 1$. At fixed p, the function $f(u,p)$ has three zeros, $u_1(p)<u_u(p)<u_2(p)$, such that $\partial f/\partial u<0$ for $u=u_{1,2}$ (stable phases 1 and 2) and >0 for $u=u_u$ (unstable phase). Start with the one-dimensional (1D) case. At fixed p this system does not support large-scale equilibrium domains unless the area rule,

$$\int_{u_1(p)}^{u_2(p)} f(u,p)du=0\,, \tag{2}$$

holds. In general, Eq. (2) can be satisfied only for a special choice (say, p_*) of the parameter p. For $p\neq p_*$ strips of the metastable phase shrink, and only one of the phases survives (e.g., Ref. [1], p. 23). Even for $p=p_*$ the length ratio of the two phases L_1/L_2 is arbitrary, as the phases are not recovered after a perturbation. The situation changes significantly, if some (possibly implicit) dynamics $p=p(t)$ of the inhibitor is allowed by imposing a global constraint on the system, so that the inhibitor $p=p(t)$ can adjust itself to the momentary positions of the interphase boundaries (domain walls) and arrest their motion. A large variety of such physical and physico-chemical systems have been studied in 1D. These include, first of all, many types of electrothermal domains in semiconductors, metals, and superconductors, reviewed in Ref. [3] (some of them have been known since the beginning of this century). Bistable heterogeneous chemical reactions [4], thermal contraction in weakly ionized plasmas [5], and radiative condensations in optically thin plasmas [6] provide further examples. Besides, there is a number of related systems, complicated by the presence of an additional *local* variable [7,8].

In each of these systems, the function $f(u,p)$ and the form of integral constraint are quite different (the latter can follow from such diverse conditions as constant voltage [3], constant average temperature [4], mass conservation [6], etc.). Therefore, the first question that we shall address concerns the general conditions, when the stable two-phase coexistence (STPC) (or domain stability) is possible in 1D. This question was addressed by Elmer, who derived a necessary condition for STPC for a particular system (voltage-controlled ballast resistor) [9]. We shall derive an important additional criterion which, combined with Elmer's criterion, provide the necessary and sufficient condition for STPC. In higher dimensions, the problem has received much less attention [10–12] (see also Ref. [13], p. 3137). In this case, the long-time multidimensional dynamics are strongly influenced by the domain interface curvature, and we shall present a number of results on the domain equilibrium, stability, and growth. In particular, we show that the dynamics of a large number of domains can be mapped into a classical mean-field model of Ostwald ripening. Finally, we resolve a long-standing controversy in the latter model.

We shall consider Eq. (1) in a finite region Ω. Assume for simplicity that none of the functions f, K, u_1, and u_2, nor their derivatives introduce any small or large parameters, and that the typical domain size L_d is much larger than the width of the interphase boundary (which is of order unity). The small parameter L_d^{-1} will be very important in our theory. We employ a general functional form of the integral constraint:

$$\int_\Omega K[u(\mathbf{r},t),p(t)]d\mathbf{r}=\text{const}. \tag{3}$$

Equations (1) and (3), complemented with the initial conditions [we usually assume the no-flux condition $(\nabla u)_n=0$ as the boundary condition] represent a closed set.

Consider the *large-scale* nonuniform equilibria. In 1D, these are alternating strips of phases 1 and 2. A necessary

stability condition can be obtained if we consider the short-time dynamics and therefore neglect the diffusion term in Eq. (1). We employ this equation separately for the phases 1 and 2, while Eq. (3) takes the simple form $L_1K(u_1(p),p)+L_2K(u_2(p),p)=\text{const}$. We require that steady state solutions $u_{1,2}=u_{1,2}(p)$ with some $p=p_0=\text{const}$ obey the global constraint for $L_{1,2}\neq 0$. Linearizing these three equations around the steady state, one arrives at a quadratic characteristic equation for the growth or damping rate of small perturbations, similar to that obtained by Elmer [9] [see his Eq. (3.11)]. This equation yields the required stability condition in terms of two straightforward but cumbersome inequalities containing the u and p derivatives of functions f and K, evaluated at each of the two steady states. We shall call these inequalities the Elmer inequalities and assume in the following that they hold. The characteristic time scale for relaxation is typically of order unity, the fast time scale of our theory.

Now we consider the long-time dynamics and take into account the diffusion term $\nabla^2 u=\partial^2 u/\partial x^2$. A collection of alternating equilibrium strips [14] now requires $p=p_*$. In the "pure" phases we have $u=U_1$ and $u=U_2$, where $U_{1,2}\equiv u_{1,2}(p_*)$. The profile of the (standing) transition front $U_f(x)$ between any two adjacent strips can be easily found analytically. In particular, the x derivative of this solution (the absolute value of this derivative will be denoted by v) can be expressed through the solution itself:

$$\frac{dU_f}{dx}\equiv \pm v(U_f)=\pm\left[2\int_{U_f}^{U_2} f(u,p_*)\,du\right]^{1/2}. \tag{4}$$

Now we find the stability condition for this nonuniform equilibrium. Let p deviate slightly from the equilibrium value p_*, so that $p(t)=p_*+\delta p(t)$, $\delta p\ll p_*$. Strips of one of the phases will start expanding (the other phase shrinking). We shall see *a posteriori* that the characteristic relaxation time is of the order of $L_d\gg 1$. This enables one to simplify the analysis considerably. First, in the regions of the pure phases $u(x,t)$ rapidly (on a time scale of order unity) adjusts to the slowly changing value of the inhibitor $p(t)$: $u_{1,2}=u_{1,2}(p)$. Second, one can look for a traveling wave solution in the regions of domain walls: $u_f(\xi)$, $\xi=x-\int^t c(t')dt'$, with a *small* and *slowly varying* front speed $c(t)$. Technically, we employ the well-known result for constant $\delta p\ll p_*$ and c (see, e.g., [1], p. 21), and then permit their slow time variations. The domain wall speed is

$$c(t)=-g\,\delta p(t),\quad g=\frac{\int_{U_1}^{U_2}[\partial f(u,p)/\partial p_*]\,du}{\int_{U1}^{U2} v(u)\,du}. \tag{5}$$

A positive value of $c(t)$ corresponds to the front moving toward phase 2. Here and in the following the p_* derivative means the p derivative, evaluated at $p=p_*$. Obviously, the front speed vanishes if $\delta p(t)\to 0$. We shall assume that $g>0$.

Now we differentiate Eq. (3) with respect to time and substitute $\partial u/\partial t$ separately in the regions of the pure phases and domain walls. We obtain

$$\frac{d\,\delta p}{dt}=-\frac{\delta p}{\tau},\quad \tau=\frac{L_1\left.\frac{dK}{dp_*}\right|_1+L_2\left.\frac{dK}{dp_*}\right|_2}{gN_f[K(U_2,p_*)-K(U_1,p_*)]}. \tag{6}$$

Here N_f is the total number of domain walls in the equilibrium. Also,

$$\left.\frac{dK}{dp_*}\right|_{1,2}=\left.\frac{\partial K}{\partial p_*}\right|_{u=U_{1,2}}+\left.\frac{\partial K}{\partial u}\right|_{u=U_{1,2}}\frac{du_{1,2}}{dp_*}. \tag{7}$$

Equation (6) shows that the strips are stable as long as $\tau>0$. In this case $p(t)\to p_*$, the e-fold relaxation time being τ. Correspondingly, the front speed goes to zero and the steady state is restored, as it was indeed observed in many systems, where τ happens to be positive (see an extensive list of experiments in Ref. [3]). The relaxation time τ is typically of order $L/N_f=L_d\gg 1$ as expected. Combination of the Elmer inequalities and inequality $\tau>0$ represents the necessary and sufficient condition for the domain stability. Notice that stability of any single domain remains marginal with respect to its translation within the region occupied by the other phase. The true positions of the domain walls are determined by the initial conditions.

Now we extend the theory to the 2D and 3D cases. Let the region Ω contain a number of large-scale "drops" of phase 1 on the background of phase 2, and that the distances between different drops, and between any drop and the system boundary is much larger than the transition layer width (that is, than unity). The motion of a domain interface will be affected, in addition to δp, by its (presumably small) local curvature. Looking for the front speed, we can employ the 2D and 3D results obtained for constant δp and $\mathscr{K}$ (e.g., [1], p. 28) and then permit their slow time variation:

$$c(t)=-g\,\delta p(t)-\mathscr{K}(t), \tag{8}$$

where $\mathscr{K}$ is the local curvature of the interface in 2D, or the sum of the two local principal curvatures in 3D. $\mathscr{K}$ is defined to be positive if the interface is convex towards phase 2 and negative otherwise. It is clear from Eq. (8) that an equilibrium implies $\delta p=-\mathscr{K}/g=\text{const}$. Therefore, only a drop with a constant $\mathscr{K}$ along its surface (that is, a perfect circle in 2D or ball in 3D) has an equilibrium shape. For an ensemble of drops to be in equilibrium, all the drop radii must be equal to each other. Such an equilibrium, however, proves to be unstable [10–12]. The instability proceeds as harsh competition (larger drops thrive at the expense of smaller ones) and is similar to Ostwald ripening (OR) .

Now we again differentiate Eq. (3) with respect to time and substitute $\partial u/\partial t$ separately in the regions of the pure phases and transition fronts. After some algebra we obtain

$$\frac{d\,\delta p}{dt}=-\left(\delta p+\frac{\langle\mathscr{K}\rangle}{g}\right)\frac{g\Lambda[K(U_2,p_*)-K(U_1,p_*)]}{\Omega_1\left.\frac{dK}{dp_*}\right|_1+\Omega_2\left.\frac{dK}{dp_*}\right|_2}, \tag{9}$$

where $\Omega_{1,2}$ are the total areas (in 2D) or volumes (in 3D) of the pure phases 1 and 2, Λ is the total length (in 2D) or

surface area (in 3D) of the interfaces between the two phases, and $\langle \ldots \rangle$ means averaging over all interfaces.

The big fraction in the right hand side of Eq. (9) is analogous to the quantity τ^{-1}, obtained in the 1D theory. If it is positive, then in many problems δp approaches $-\langle \mathscr{K} \rangle / g$ on the time scale of $\tau \sim L_d$. At the next, "adiabatic" stage $\delta p(t) \simeq -\langle \mathscr{K} \rangle(t)/g$, so that the dynamics will be governed solely by the mean curvature of the domain interfaces. Rubinstein and Sternberg [11] developed such an adiabatic theory for a particular example of the globally constrained RDE.

We solved several model problems of the 2D and 3D dynamics, using Eqs. (8) and (9). First, we proved the linear stability of an equilibrium planar interface and of a perfect circular (or spherical) drop with respect to deformations of their shape. In these problems, the e-fold relaxation time proves to be of order L_d^2 that is much longer than $\tau \sim L_d$. In these examples, the domains are "truly" (rather than marginally) stable with respect to the perturbations preserving the surface-to-volume ratio of the two phases. However, they remain only marginally stable with respect to translations.

Extending the treatment of [11], one can obtain two additional results, pertaining to *nonlinear* deformations of a 2D drop. First, any domain of a convex shape finally becomes perfectly circular. Second, merging singularity (drop-donut transition) can develop in a finite time in a strongly concave, two-armed drop.

For their particular example of globally constrained RDE, Schimansky-Geier *et al.* [10] predicted instability of an equilibrium drop with respect to a purely radial mode (see also [12]). Equations (8) and (9) make it possible to fully investigate the *nonlinear* dynamics of this instability. The problem is formulated in terms of the drop radius R and inhibitor mismatch δp. An equation for $\dot{R}$ is given by Eq. (8):

$$\dot{R} = c(t) = -g\,\delta p - 2R^{-1} \tag{10}$$

(again, we consider a drop of phase 1). An equation for $\dot{\delta p}$ follows directly from Eq. (9). Assume for simplicity that the drop volume is much smaller than the total volume V of the domain Ω. Then

$$\dot{\delta p} = -4\pi\epsilon R^2 (g\,\delta p + 2R^{-1}), \tag{11}$$

where

$$\epsilon = \frac{K(U_2, p_*) - K(U_1, p_*)}{V(dK/dp_*)|_2}.$$

Equations (10) and (11) are integrable. The first integral is

$$-\delta p + (4\pi/3)\epsilon R^3 = \text{const}. \tag{12}$$

Consider the dynamics on the phase plane $(R, \delta p)$. For the integral curves (IC) (12) not intersecting the line of equilibria (LE) $\delta p = -2/(gR)$, the corresponding drops never achieve an equilibrium. Instead, they shrink and disappear in a finite time. For IC that have two intersection points with the LE, the smaller-radius point is unstable, while the larger-radius point is stable, in agreement with the linear analysis [10,12]. The critical (minimum) radius R_c of a stable drop is determined by the tangency point between an IC and the LE: $R_c = (2\pi g \epsilon)^{-1/4}$. The corresponding critical volume $V_c = (4\pi/3)R_c^3$ scales like $V^{3/4}$, while the relative critical volume V_c/V scales like $V^{-1/4}$. Notice that this nontrivial radial dynamics is completely missed by the adiabatic theory [11].

Equations (8) and (9) can be also employed to analyze the dynamics of a nonsymmetric 2D (3D) "donut" of phase 1, bounded by two nonconcentric circumferences (spheres). Such a donut always develops singularity in a finite time. Depending on the initial values of the external and internal donut radii and δp, either the "hole" of the donut (phase 2) shrinks to zero, or reconnection occurs so that the hole breaks out, and the donut ultimately becomes a circular (spherical) drop [15].

Now we consider the statistical aspects of the many domains' growth as described by the globally constrained RDE. To the best of our knowledge, this important subject has not been addressed before. The dynamics of a large number of drops is conveniently described by the distribution function (DF) $F(R,t)$ of the drops with respect to their radii. In 3D we have $\int_0^\infty F(R,t)dR = n(t)$, the time-dependent volume concentration of the drops. For $\delta p \ll p_*$, no new domains can develop, so that the DF must satisfy the continuity equation

$$\frac{\partial F}{\partial t} + \frac{\partial}{\partial R}(\dot{R}F) = 0\,, \quad \dot{R} = -g\,\delta p - 2R^{-1}. \tag{13}$$

As usual, the inhibitor dynamics is governed by the global constraint [compare it with Eq. (11)]:

$$-\delta p + \frac{4\pi\epsilon_0}{3}\int_0^\infty R^3 F(R,t)dR = Q = \text{const} \tag{14}$$

with $\epsilon_0 = \epsilon V$. Equations (13) and (14) belong to the well-known set [16–18] of the mean-field models of OR. That a globally constrained RDE can be mapped into a mean-field model of OR is a remarkable fact.

Of a central importance in the problem (13) and (14) is an asymptotic similarity solution for the DF, and related scalings. Lifshitz and Slezov [16] showed (for another variant of equation for $\dot{R}$) that for any *extended* initial condition $F(R,0)$, the DF will approach at large times this similarity DF (SDF). Recently, this result has been supported by extensive numerical simulations [19]. The same problem with a *localized* initial condition has not been solved yet. In a parallel development, Brown [20] has found numerically a whole one-parameter family of SDF, with the Lifshitz-Slezov solution corresponding to a limiting value of the parameter. The role of these solutions in the general initial value problem has not been clarified yet and is a subject of long-standing controversy [19,20]. We will now resolve this controversy, working with the model (13) and (14).

First of all, in the limit of $t \to \infty$, Eqs. (13) and (14) have a one-parameter family of solutions for the SDF that can be found analytically. The similarity ansatz is $F(R,t) = t^{-2}\Phi(R/t^{1/2})$. Correspondingly, δp goes down like $-g^{-1}\beta t^{-1/2}$, n decreases like $\eta t^{-3/2}$, and the average drop radius grows like $\chi t^{1/2}$. The positive coefficients $\eta = M_0$, $\chi = M_1/M_0$, and $\beta = M_1/(2M_2)$ are determined by the moments M_k of the function $\Phi(\xi)$:

$M_k=\int_0^\infty \xi^k\Phi(\xi)d\xi, k=0, 1,$ and 2. Φ satisfies an ordinary differential equation (ODE) and a normalization condition which one obtains from Eqs. (13) and (14), respectively. Equation (14) yields

$$\int_0^\infty \xi^3\Phi(\xi)d\xi \simeq \frac{3Q}{4\pi\epsilon_0}. \tag{15}$$

Integrating the ODE for Φ, we obtain

$$\Phi(\xi)\propto \frac{\xi}{|\xi^2-2\beta\xi+4|^{5/2}}\exp\left(-3\beta\int^\xi \frac{d\xi}{\xi^2-2\beta\xi+4}\right).$$

Further results depend on β. The case $0<\beta<2$ must be ruled out, since the integral (15) diverges logarithmically. For $\beta=2$, we obtain the Wagner's SDF [17]:

$$\Phi(\xi)\propto \xi(2-\xi)^{-5}\exp\left(-\frac{6}{2-\xi}\right) \tag{16}$$

for $\xi<2$, and $\Phi(\xi)=0$ elsewhere. This solution vanishes at $\xi=2$ together with all its derivatives, and it is a counterpart of the Lifshitz-Slezov solution [16], obtained for another variant of $\dot{R}$.

In the case of $\beta>2$, formal integration yields

$$\Phi(\xi)\propto \xi|\xi_- -\xi|^{-5/2+3\beta/2\sqrt{\beta^2-4}} \times|\xi_+-\xi|^{-5/2-3\beta/2\sqrt{\beta^2-4}}, \tag{17}$$

where $\xi_\pm=\beta\pm(\beta^2-4)^{1/2}$. A positive, nonsingular solution is constructed by using Eq. (17) on the interval $(0,\xi_-)$ and setting $\Phi(\xi)=0$ for $\xi\geq\xi_-$. The condition $\Phi(\xi_-)=0$ requires that $\beta<5/2$. All SDF from (17) vanish at $\xi=\xi_-$, but their derivatives are nonzero there. Therefore, the whole family of SDF is defined on quite a narrow interval of the parameter β: $2\leq\beta<5/2$, the left border of which corresponds to the Wagner solution.

Finally, we find the selection rule for the "correct" SDF. Employing the arguments of Lifshitz and Slezov [16] for our variant of $\dot{R}$, [15] we see that it is the Wagner solution (16) that will be the attractor of any *extended* initial distribution. Now consider a *localized* initial distribution which is positive on the interval $[0,R_m(t=0)]$ and zero for $R>R_m(t=0)$. First, the solution will always remain localized on an interval $[0,R_m(t)]$. Furthermore, we find from Eq. (13) that the leading term of the expansion of $F(R,t)$ in the vicinity of $R=R_m(t)$ can be written as $A(t)[R_m(t)-R]^\lambda$, where λ is uniquely determined by the initial condition:

$$\lambda=\left.\frac{d\ \ln F(R,t=0)}{d\ \ln[R_m(t=0)-R]}\right|_{R=R_m(t=0)}. \tag{18}$$

It is easy to see that parameter λ remains invariant for *any* localized solution of the problem (13) and (14) [21]. Therefore, it is the double logarithmic derivative of the initial distribution, evaluated at $R=R_m$ that uniquely selects the "correct" SDF. Equating the power exponent of the binomial $\xi_- -\xi$ in Eq. (17) to the parameter λ immediately yields the selected β:

$$\beta=2(\lambda+5/2)(\lambda+1)^{-1/2}(\lambda+4)^{-1/2}. \tag{19}$$

The same arguments are applicable to all *mean-field* theories of OR with other equations for $\dot{R}$ (see the list in [18]). It is important that physical and physicochemical systems, described by Eqs. (1) and (3) in the limit of many drops, represent the simplest paradigm of OR, as the mean-field theory in this case becomes "almost exact" [22].

We are very grateful to I. Aranson, F.-J. Elmer, J. Fineberg, M. Marder, and A. Vilenkin for useful discussions.

[1] A.S. Mikhailov, *Foundations of Synergetics I. Distributed Active Systems* (Springer-Verlag, Berlin, 1990).
[2] J.D. Murray, *Mathematical Biology* (Springer-Verlag, Berlin, 1989), p. 481.
[3] A.Vl. Gurevich and R.G. Mints, Rev. Mod. Phys. **59**, 941 (1987).
[4] L. Pismen, Chem. Eng. Sci. **34**, 563 (1979).
[5] A.V. Nedospasov and V.D. Khait, *Oscillations and Instabilities in Low-Temperature Plasmas* (Nauka, Moscow, 1979), p. 116 (in Russian).
[6] I. Aranson, B. Meerson, and P.V. Sasorov, Phys. Rev. E **47**, 4337 (1993).
[7] F.-J. Niedernostheide, R. Dohmen, H. Willebrand, B.S. Kerner, and H.-G. Purwins, Physica D **69**, 425 (1993).
[8] U. Middya, M. Sheintuch, M.D. Graham, and D. Luss, Physica D **63**, J393 (1993).
[9] F.J. Elmer, Phys. Rev. A **41**, 4174 (1990).
[10] L. Schimansky-Geier, Ch. Zülicke, and E. Schöll, Z. Phys. B **84**, 433 (1991).
[11] J. Rubinstein and P. Sternberg, IMA J. Appl. Math. **48**, 249 (1992).
[12] I. Aranson, B. Meerson, and P.V. Sasorov, Phys. Rev. E **52**, 948 (1995).
[13] L.M. Pismen, J. Chem. Phys. **101**, 3135 (1994).
[14] Actually, such a state breaks down and gets simplified [23,6] on a very long time scale of order $\exp[O(L_d)]$. Here we are interested in faster processes.
[15] The details will be presented elsewhere.
[16] I.M. Lifshitz and V.V. Slezov, Sov. Phys. JETP **8**, 331 (1959).
[17] C. Wagner, Z. Electrochem. **65**, 581 (1961).
[18] V.V. Slezov and V.V. Sagalovich, Sov. Phys. Usp. **30**, 23 (1987).
[19] M.K. Chen and P.W. Voorhees, Model. Simul. Mater. Sci. Eng. **1**, 591 (1993).
[20] L.C. Brown, Acta Metall. **37**, 71 (1989); Scr. Metall. Mater. **24**, 963; 2231 (1990).
[21] For $R_m(t)$ one obtains the same ODE as in Eq. (13).
[22] Indeed, the limit of a small volume fraction of the drops, while rendering only a minor technical convenience in Eq. (14), is of a crucial importance for the mean-field theory of the "conventional" OR.
[23] C. Elphick, O. Regev, and E.A. Spiegel, Mon. Not. R. Astron. Soc. **250**, 617 (1991).