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Disrupted coarsening in complex Cahn-Hilliard dynamics

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Predicting the pattern formation in a system maintained far from equilibrium is a complex task. For a given dynamics governed by the evolution of a conservative order parameter, recent investigations have demonstrated that the knowledge of the long time expression of the order parameter is sufficient to predict the existence of disrupted coarsening, i.e., the pinning of the inhomogeneities wavelength to a well defined value. However, there exists some dynamics for which the asymptotic form of the order parameter remains unknown. The Cahn-Hilliard-like equation used to describe the stability of solids under irradiation belongs to this class of equations. In this paper, we present an alternative to predict the patterning induced by this equation. Based on a simple ansatz, we calculated the form factor and proved that a disrupted coarsening takes place in such dynamics. This disrupted coarsening results from the bifurcation of the implicit equation linking the characteristic length of the dynamics $(k_m^\infty)^{-1}$ to a control parameter describing the irradiation. This analysis is supported by direct simulations. From this paper, it clearly appears that the bifurcation of k_m^∞ is a criterion for disrupted coarsening.

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I. INTRODUCTION

When a system initially at equilibrium is subject to an external perturbation, small spatial inhomogeneities in the order parameter can lead to nanometric domains. This patterning results from the balance between a short range order interaction and a long range order perturbation. For example, magnetic systems, dipolar fluids [1], block copolymers [2–4], and chemical reactions exhibit striped lamellar domains [5]. This phenomenon, ubiquitous in physics and chemistry, has been extensively studied. The modeling of this patterning is mainly based on the Cahn-Hilliard equation [6–8]. For unperturbated systems, the patterning associated with this equation is now clearly understood [9–11].

For systems submitted to long range interactions, the external force may disrupt the Ostwald ripening and then forbids the formation of large domains as pointed out by some authors in one dimension [12] ($d = 1$). In these cases, the characteristic size of such domains no more evolves with time and remains pinned to a well defined value leading to the appearance of a disrupted coarsening. The dynamics associated with this disrupted coarsening is not clearly understood. Based on the knowledge of the stationary states, Politi and Mishbah [12] give a useful criterion to discuss this interrupted coarsening. A central problem in nonequilibrium pattern formation associated with the Cahn-Hilliard-like dynamics is to identify criteria predicting the appearance of this disrupted coarsening when stationary states remain unknown.

In this paper, we address this problem studying the behavior of alloys under irradiation. Under irradiation, random displacements of atoms over a few angstroms regardless of their chemical identity lead to a solid solution in well defined areas called displacement cascades [13]. This random displacement of atoms acts as an external force able to counterbalance the attractive interactions between atoms. A balance between this long range interaction over a few nanometers and the thermodynamic short range interaction due to the minimization of the free energy between nearest neighbors leads to a complex dynamics modeled within the Cahn-Hilliard equation framework [14]. This competition leads to the appearance of a specific modulation of the concentration wave and disrupted coarsening. Unfortunately, no analytical expression for the stationary states of Cahn-Hilliard-like equations exists for this system. In this paper, calculations, valid whatever the space dimension d is, are supported by numerical simulations performed only in two dimensions ($d = 2$).

II. FORMULATION OF THE PROBLEM

A. Modeling of the atomic jumps

To describe the irradiation effects in a solid, the probability density function $p_R(\mathbf{x})$ associated with the ejection of atoms at a given distance x has been introduced [14]. The term $p_R(\mathbf{x})$ results from an average of atomic collisions between atoms set in motion by impinging atoms over the displacement cascade. From a coarse graining procedure [15,16], the effect of highly nonuniform collision events reduces to a probability to eject atoms at a distance x from its initial position. Obviously, $p_R(\mathbf{x})$ is radial and then symmetric. From Monte Carlo and molecular dynamic simulations performed on numerous solids with different incident particles [16,17], it appears that $p_R(\mathbf{x})$ exhibits a cut-off radius roughly equal to 2 nm. This value does not strongly depend on the nature and the energy of the incident particle nor the target atoms [17]. In a first approximation, $p_R(\mathbf{x})$ can thus be represented by a simple exponential decay associated with the cut-off R. By analogy with the kinetic theory of gases, R can then be understood as the mean free path of ejected atoms. On the other hand, the strength of the atomic mixing is defined by γ, the ratio between the number of atoms ejected and the number of atoms in the volume of the

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cascade per time unit [15]. The term γ is related to the flux of incident particle ψ and can be easily calculated from the slowing down of particles in matter [15,16].

The effect of this atomic mixing due to irradiation induces a long range perturbation in the displacement cascade. This perturbation can be written as [15–19]

$$\gamma[\eta(\mathbf{x},t) - (p_R * \eta)(\mathbf{x},t)], \qquad (1)$$

where $*$ denotes the convolution product and $\eta(\mathbf{x},\mathbf{t})$ is the conservative scalar order parameter [20]. For a binary mixture AB, η reduces to the difference between the atomic concentrations $c_A - c_B$. It is thus possible to describe the behavior of solids under irradiation adding this term in the usual Cahn-Hilliard equation [14]. Assuming the mobility of defects does not depend on $\eta(\mathbf{x},t)$ [21], the nonlinearities associated with this dynamics are only due to the nonlinearity of F, the standard Cahn-Hilliard free energy functional. Even if the mobility M does not depend on the conservative order parameter $\eta(\mathbf{x},t)$, its evolution as a function of the atomic mixing is far from being understood. This last point precludes a direct comparison between the time evolution of the calculated and measured patterning. Despite this point, it remains possible to predict the asymptotic behavior of the coarsening induced by irradiation.

B. The modified Cahn-Hilliard equation

For a free energy density f with a double well structure below the critical temperature [20,22], $f(\eta)$ reduces to $\alpha\eta^2 + \beta\eta^4$. The phenomenological coefficient $\alpha = a(T - T_c)$ is negative below the critical temperature and positive above; the coefficients a and β are always positive. Moreover, the energetic cost of interfaces is taken into account by the term $\mu|\nabla\eta|^2$. In this expression, μ is always positive. Substituting dimensionless variables $\mathbf{x}' = \mathbf{x}\sqrt{\frac{-\alpha}{\mu}}$, $R' = R\sqrt{\frac{-\alpha}{\mu}}$, $t' = t(\frac{M\alpha^2}{\mu})$, $\phi = \eta(\frac{-\beta}{\alpha})$, and $W' = \gamma(\frac{\mu}{M\alpha^2})$, the modified Cahn-Hilliard type equation can be written in a dimensionless form [23]:

$$\frac{\partial\phi}{\partial t} = \nabla^2(-\phi + \phi^3 - \nabla^2\phi) - W(\phi - p_R * \phi). \qquad (2)$$

In this equation, the primes were dropped for simplicity. Only two independent parameters, W and R, parametrize this equation. R is associated with the second moment of $p_R(x)$ and W is related to the strength of the irradiation γ.

When $W = 0$ or $R \to 0$, this equation reduces to the usual Cahn-Hilliard equation [23]. When $R \to \infty$, this equation has been proposed to describe phase separation in both symmetric diblock copolymers and chemically reactive binary mixtures [24].

To handle the dynamics of patterning, thermal fluctuations as well as fluctuations induced by irradiation must be introduced in Eq. (2) [25]. As clearly pointed out by some authors [10,23], the noise induced by fluctuations has no effect on the long time patterning. In the following, we only discuss the dynamics of the nonstochastic modified Cahn-Hilliard equation given by Eq (2).

III. CALCULATION OF THE FORM FACTOR

The symmetry of $p_R(\mathbf{x})$ insures the existence of a Lyapounov functional and the uniqueness of $\phi(\mathbf{x},t)$. However, no expression for the stationary solution of Eq. (2) exists [26].

A. A mean field approximation

To overcome this difficulty, we assume that the pair correlation function $\langle\phi(\mathbf{x},t)\phi(\mathbf{y},t)\rangle$ is invariant by translation ($\langle . \rangle$ denotes the ensemble average over the initial configurations). From this assumption, $\langle\phi(\mathbf{x},t)^2\rangle = \langle\phi(\mathbf{0},t)^2\rangle = S(t)$. Moreover, we assume that $\phi(\mathbf{x},t)^3 \approx \langle\phi(\mathbf{x},t)^2\rangle\phi(\mathbf{x},t)$. Our ansatz leads to a "linearization" of Eq. (2) as first applied by previous authors on the study of the diblock copolymer segregation [2,23]. It is straightforward to determine the equation followed by the form factor $S(\mathbf{k},t)$, the Fourier transform of $\langle\phi(\mathbf{x},t)\phi(\mathbf{y},t)\rangle$:

$$\frac{\partial S(\mathbf{k},t)}{\partial t} = \left(-2k^2[k^2 - 1 + S(t)] - \frac{2WR^2k^2}{1 + R^2k^2}\right) S(\mathbf{k},\mathbf{t}), \qquad (3)$$

where $\mathbf{k}$ is the reduced wave factor of modulus k. As $p_R(\mathbf{x})$ is an exponential function, its Fourier transform reduces to $\frac{1}{1+R^2k^2}$. The term $\frac{2WR^2k^2}{1+R^2k^2}$ is thus the Fourier transform of Eq (1). The total integrated scattering intensity $S(t)$ must be determined self-consistently:

$$S(t) = \int S(\mathbf{k},t)\frac{d\mathbf{k}}{(2\pi)^d}, \qquad (4)$$

where d is the spatial dimension of the system ($d = 2$ in this paper).

From our ansatz, the function $S(\mathbf{k},t)$ is thus formally given by

$$S(\mathbf{k},t) = S(\mathbf{k},0)\exp[-2t\, g(k,t,R,W)], \qquad (5)$$

where $S(\mathbf{k},0)$ is the initial form factor and

$$g(k,t,R,W) = \frac{k^2}{t}\int_0^t (S(u) - 1)du + k^4 + \frac{WR^2k^2}{1 + R^2k^2}.$$

It clearly appears that $g(k,t,R,W)$ only depends on the modulus of $\mathbf{k}$, k. This implies that $S(\mathbf{k},t)$ exhibits a spherical symmetry. On the other hand, $g(k,t,R,W)$ exhibits a minimum for $k = k_m(t)$. The wave vector modulus $k_m(t)$ is obviously associated with the maximum of the function $S(k,t)$. The existence of a maximum for $S(k,t)$, assessed by numerical simulations, is not surprising for a Cahn-Hilliard-like equation. The linear part of Eq. (2) acts as a filter to select a characteristic wave vector [23]. However, the nonlinear term of Eq. (2) can no more be neglected below the critical temperature. This point implies that $k_m(t)$ for large times is different from the one derived for small times associated with the linear part of Eq. (2).

Expanding $g(k,t,R,W)$ to the fourth order in the neighborhood of $k_m(t)$ and applying the steepest descent method to calculate $S(t)$ for large t values, it is easy to show that

$$k_m(t)^d S(k,t) \propto \exp\left[-2t\, k_m^4(t)b_4(k_m(t))\left(\left(\frac{k}{k_m(t)}\right)^2 - 1\right)^2\right]$$

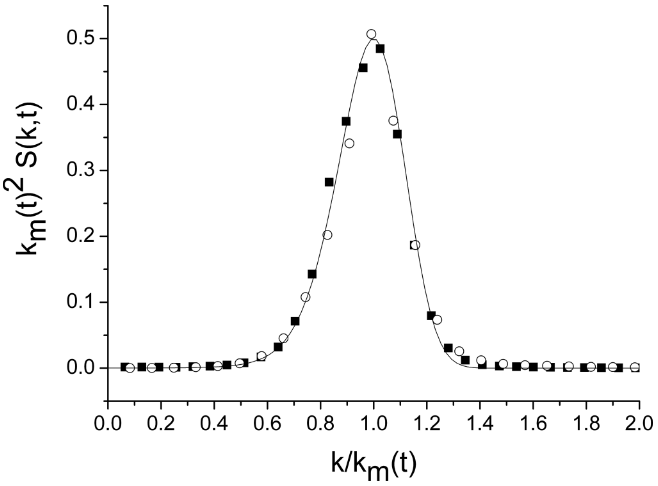

FIG. 1. Comparison between $S(k,t)$ derived from our ansatz (full line) and extracted from numerical simulations of Eq. (2) (black squares and open circles). Computations were performed in two dimensions ($d = 2$) using a finite difference scheme in the Fourier space with a 512×512 grid size for large times [the reduced times were equal to 2000 (black squares) and 5000 (open circles); for each time, simulations were averaged over 100 random initial configurations].

exhibits a scaling behavior as a function of the reduced variable $\frac{k}{k_m(t)}$, where $b_4(k_m(t))$ is the fourth order coefficient of the expansion of $g(k,t,R,W)$ as a function of the variable k.

Figure 1 displays the comparison between $S(k,t)$ calculated and extracted from numerical calculations. The fair agreement between these two calculations assesses the validity of our ansatz. Moreover, this figure highlights the scaling law followed by $S(k,t)$.

B. Patterning of stationary states

Applying the steepest descent method [27], it is possible to derive the asymptotic behavior of $k_m(t)$. As $S(k,t)$ exhibits a maximum for large times, the asymptotic value k_m^∞ of $k_m(t)$ for large times allows defining the characteristic length $L = 1/k_m^\infty$ of the atoms modulations due to irradiation in the real space, i.e., the microstructure induced by irradiation in the material. The knowledge of k_m^∞ thus allows characterizing the patterning of stationary states. The simple application of the steepest descent method allows one to calculate $S(t)$:

$$S(t) = B\, k_m(t)^{-1+d} \exp[-2t\, g(k_m(t),t,R,W)],$$
$$g(k_m(t),t,R,W) = 2k_m^4(t)\left[-1 + \frac{WR^4}{[1+R^2k_m(t)^2]^2}\right], \qquad (6)$$

where $B = \frac{\Gamma(\frac{d}{2})}{S(k_m(t),0)2^{\frac{3}{2}-d}\pi^{1-\frac{d}{2}}}$ is a normalization factor and Γ is the gamma function. On the other hand, $S(t)$ is linked to R, W, t, and $k_m(t)$ via the first derivative of $g(k,t,R,W)$:

$$S(t) = 1 + \frac{d}{dt}\left(\int_0^t (S(u)-1)du\right)$$
$$= 1 - \frac{d}{dt}\left(-2tk_m(t)^2 - \frac{WR^2t}{[1+R^2k_m(t)^2]^2}\right). \qquad (7)$$

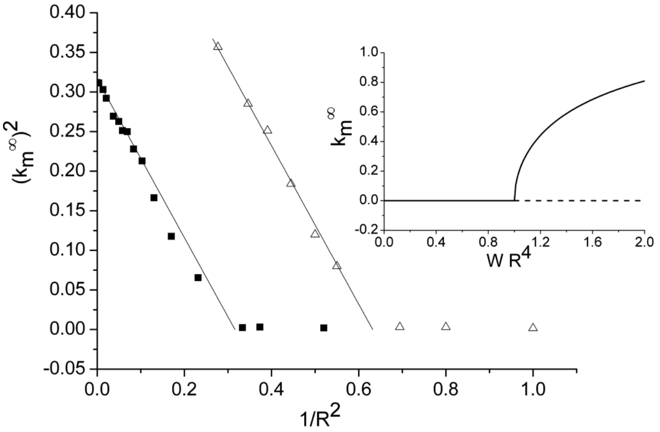

FIG. 2. Variation of $(k_m^\infty)^2$ extracted from numerical simulations (black squares: $W = 0.1$, open triangles: $W = 0.4$) for large times ($t > 100\,000$) as a function of R^{-2}. The evolution of $(k_m^\infty)^2$ exhibits a linear variation as expected (full lines), for $WR^4 > 1$. For $WR^4 < 1$, $(k_m^\infty)^2$ is null. The inset displays the evolution of k_m^∞ with WR^4.

Satisfying Eqs. (6) and (7) leads to an implicit equation for $k_m(t)$. Moreover, if a nonnull stationary state exists, from Eq. (6), the two conditions $\lim_{t\to\infty} g(k_m(t),t,R,W) = 0$ and $\lim_{t\to\infty} S(t) > 0$ must be verified. Using Eq. (7), these conditions imply $W < (\frac{R^2+1}{2R^2})^2$. Two distinct cases can then be distinguished:

(a) for $WR^4 < 1$, Eq. (6) is only satisfied for $k_m(t) \approx t^{-\frac{1}{3}}$, as expected for the usual Cahn-Hilliard equation out of irradiation;

(b) for $WR^4 > 1$, two different values of $k_m(t)$ are solutions of the implicit equation. When time goes to infinity, either $k_m(t)$ tends to zero or to a nonnull value

$$k_m^\infty = \sqrt{\frac{\sqrt{WR^4}-1}{R^2}}. \qquad (8)$$

It can be noticed that for $R \to \infty$, our expression of k_m^∞ is equal to the one derived for the patterning of diblock copolymers' formation [24]. Figure 2 displays the evolution of $(k_m^\infty)^2$ derived from numerical simulations as a function of R^{-2} for different values of W.

IV. DISRUPTED COARSENING

A. Stability analysis

The different kinks observed on Figure 2 assess that two distinct regimes take place as a function of WR^4 which can be understood as a control parameter. For $WR^4 > 1$, it must be noticed that a long time [$t \gg (WR^4-1)^{-1}$] is needed to reach the stationary state described by the wave vector k_m^∞ when WR^4 tends to one.

A simple linear stability analysis of Eq. (5) around k_m^∞ for $WR^4 > 1$ and $WR^4 < 1$ clearly shows that only $k_m^\infty = 0$ leads to a stable solution of $S(k,t)$ for $WR^4 < 1$ (full line in the inset on Fig. 2). For $WR^4 < 1$, the atomic mixing induced by irradiation is not sufficient to counterbalance the ordering of the solid below the critical temperature. An Ostwald ripening takes place and the characteristic length of the problem reduces to $k_m(t)^{-1} \propto t^{\frac{1}{3}}$ for $d = 2$, as expected for a classical Cahn-Hilliard equation. For $WR^4 = 1$, a bifurcation occurs as clearly shown in the inset on Fig. 2. For $WR^4 > 1$, the solution associated with $k_m^\infty = 0$ becomes unstable (dotted line) and only the form factor $S(k,t)$ associated with k_m^∞ obtained from Eq. (8) is stable (full line). This nonnull value of k_m^∞ for $WR^4 > 1$ implies that the coarsening is disrupted. A new patterning resulting from the balance between the atomic mixing and the ordering energy takes place. This patterning is defined by a constant characteristic length $(\frac{\sqrt{WR^4-1}}{R^2})^{-\frac{1}{2}}$. As R tends to infinity, $k_m^\infty = W^{\frac{1}{4}}$, as calculated for a diblock copolymer's separation [24].

This analysis clearly demonstrates that the bifurcation of $k_m(t)$ implies disruption of the ripening. This bifurcation appears as an alternative criterion to predict a disrupted coarsening in dynamics for which no asymptotic expression for the order parameter exists.

B. Derivation of a phase diagram

From the knowledge of the asymptotic behavior of $k_m(t)$, it is possible to draw a "phase diagram" versus the two parameters R and W as shown on Fig. 3. The phase diagram derived from the analysis of $k_m(t)$ is qualitatively similar to the one obtained from a classical minimization of the Lyapounov energy [14]. This point then assesses our ansatz. Above the line $W = (\frac{R^2+1}{2R^2})^2$, the atomic mixing induced by irradiation cannot be counterbalanced by the ordering energy and a solid solution is created. The form factor $S(k,t)$ then reduces to a Dirac distribution. We thus obtain a solid solution and $\phi(\mathbf{x},t)$ obviously does not depend on $\mathbf{x}$. A second line can be drawn on the phase diagram defining the bifurcation of k_m^∞ (dotted line $WR^4 = 1$). Below this line, the atomic mixing cannot counterbalance the chemical ordering and $S(\mathbf{k},t)$ tends to zero according to an inverse power law as expected from the usual Cahn-Hilliard equation. A phase separation is thus observed. Between these two lines, the coarsening is disrupted. The form factor is peaked to a nonnull value of $k_m(t) = k_m^\infty$ inducing patterning at the nanometric scale. Figure 3 summarizes these different cases. From this analysis, it clearly appears that the wave vector associated with the disruptive patterning k_m^∞ is proportional to $(\psi)^{\frac{1}{4}} M(T)^{-\frac{1}{4}}$. The value of the wave vector is only a function of the flux of incident particles ψ and the temperature of irradiation via the mobility. This dependence of k_m^∞ with the flux and the temperature can be clearly understood. This is the effect of the competition between the ejection of atoms (the ψ term) and the healing of these ejected atoms via a diffusion process.

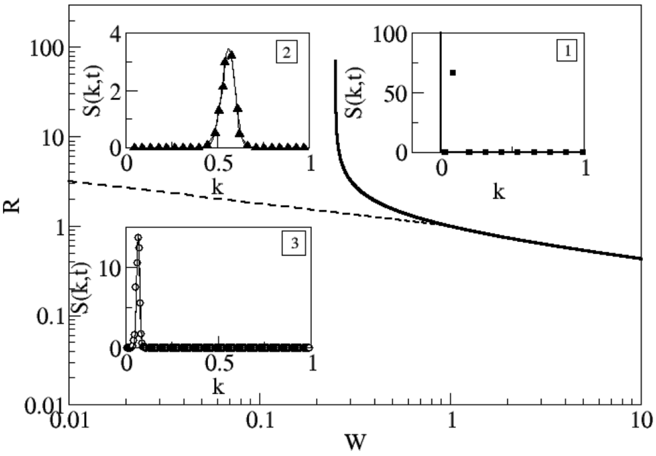

FIG. 3. Schematic phase diagram describing different patterns for large times ($t > 1000$). Above the full line, $S(k,t)$ (black squares: $W = 0.4$, $R = 3$, inset 1) is a Dirac function. Below the full line, $S(k,t)$ exhibits a Gaussian-like shape with a nonnull maximum (black triangles: $W = 0.4$, $R = 1.6$, inset 2). Below the dashed line, $S(k,t)$ tends to zero (open circles: $W = 0.4$, $R = 1$, inset 3).

TABLE I. Values of the mobility, WR^4, and the wave vector k_m^∞ in CuCo irradiated by 1 MeV Kr ions with an incident flux of 10^{10} cm^{-2}s^{-1} at different temperatures.

T(K)	M(cm^{2}s^{-1}eV^{-1})	WR^4	k_m^∞(nm^{-1})
1000	7.1410^{-11}	250	2
500	7.810^{-14}	2.3410^5	11

C. Application

From this analysis, it is possible to estimate typical values of k_m^∞ at different temperatures in immiscible binary alloys submitted to irradiation. From the Bragg-Williams model, a, β, and the critical temperature T_c can be easily computed for the CuCo alloy [20]. These values are equal to 1.710^{-4}eV K^{-1}, 1.33 eV and 1928 K, respectively. On the other hand, μ is equal to 0.443 eV A^2, derived from the Krivoglaz-Clapp-Moss formalism [28]. From Monte Carlo simulations of the chemical diffusion of species in this alloy, the mobility $M(T)$ out of irradiation can also be determined [29]. Submitted to a 1 MeV krypton irradiation and a flux ψ of 10^{10} cm^{-2}s^{-1}, displacement cascades are created in this alloy over a characteristic length of tens of nanometers and γ reduces to 500 s^{-1} [16]. Moreover, the value of the cut-off radius R is equal to 2 nm. Assuming the mobility $M(T)$ does not evolve under irradiation, it is possible to estimate the value of the wave vector associated with the patterning for large times at different temperatures. Table I summarizes the evolution of the mobility (assumed to be the mobility in the nonirradiated alloy), WR^4, and k_m^∞ at two distinct temperatures. For these two temperatures, WR^4 is always greater than 1 and a disrupted patterning would occur. For both temperatures, patternings of a few nanometers appear for such fluxes. From the values of k_m^∞ derived from this paper, it appears that only small angle scattering experiments are able to detect the patterning induced by irradiation in CuCo. In fact, from the value of k_m^∞ at 1000 K, it is possible to determine the Bragg angle associated with the patterning in an x-ray diffraction experiment. For Cu$K\alpha$ radiation ($\lambda = 0.154$ nm) and k_m^∞ of 2 nm^{-1}, the Bragg angle associated with the diffraction is $2\theta_B = \frac{k_m^\infty \lambda}{2\pi} \approx 3°$.

V. CONCLUSIONS

In this paper, we present an alternative to predict the disruption of the coarsening for a complex Cahn-Hilliard-like dynamics, for which no analytical solution of the stationary order parameter exists. We clearly demonstrate by a direct comparison with numerical simulations that a simple ansatz allows one to overcome this problem. Applying this ansatz, we show that the patterning induced by Eq. (2) is characterized by a well defined wave vector $k_m(t)$. Despite the irradiation, $S(\mathbf{k},t)$ also exhibits a scaling behavior as it is the case out of irradiation as displayed on Fig. 1. Thanks to this ansatz, it clearly appears that $k_m(t)$ satisfies an implicit equation as a function of the parameters W and R. This equation exhibits a bifurcation as a function of the control parameter WR^4. Above the critical value $WR^4 = 1$, the asymptotic expression of $k_m(t)$ for large times is nonnull. This nonnull value of k_m^∞ implies a disrupted coarsening of the patterning in agreement with numerical simulations. The main interest of this paper in comparison with previous works [23,24] is that the disrupted patterning clearly depends on the bifurcation of $k_m(t)$ in the two dimensional control parameters space as pointed on Eq. (3). This result confirms and generalizes previous investigations on the disrupted patterning in one dimension.

A puzzling question is whether for more complex dynamics with a translation symmetry coarsening with or without frozen periodicity are the only possible scenarios. This question appears to be a perspective for this work. Another perspective will be to consider if there is any simple link between simple symmetries and frozen coarsening. The last perspective is to extend our analysis to nonlocal equations as they arise, for example, in solidification or viscous fingering or to more complex dynamics as the Kuramoto-Shivashinki [30] one in which the mobility becomes a function of the order parameter.

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