Critical Behavior of Complex Interfaces

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Dynamics of complex interfaces is investigated in a model of an oscillatory medium. The moving interfacial zone separating two phases of homogeneous oscillation consists of a phase with chaotic spatial and temporal behavior. As system parameters vary, the thickness of the interface grows until a phase transition occurs where the chaotic phase fills the entire domain. The system behavior and its critical properties are analyzed in terms of two coupled stochastic equations describing the profiles that delimit the interfacial zone. [S0031-9007(97)04091-X]

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Interfaces play a prominent role in determining the macroscopic dynamical behavior of many physical systems. Examples include magnetic systems where Ising walls separate regions with opposite magnetization, diffusion-limited growth processes [1], chemically reacting systems where fronts separate domains of different chemical composition [2]. Recently, experiments have been carried out on periodically forced, spatially distributed, oscillatory chemical systems that show frequency locked chemical patterns, where interfaces separate domains of different discrete oscillation phases [3]. The analysis carried out in the present Letter pertains to interfaces of this type.

Normally, the interface dynamics is described by a single-order parameter, the profile, which is assumed to be a single-valued function of the position along the interface. Langevin-type stochastic models for the interfacial profile have been successfully applied to many of the above systems. One of the most studied stochastic models of this type is the Kardar-Parisi-Zhang (KPZ) equation [4], which incorporates the simplest but relevant nonlinearity for the description of a wide class of growth processes.

However, there are also examples of interfaces with an internal structure which is not obviously negligible or just reducible to a renormalization of the stochastic force term. In this Letter we study one such example arising in a 2D model of an oscillatory medium, where the interface separates different phases of the same periodic, spatially homogeneous solution [5]. The interface at one time instant is shown in Fig. 1 where the internal structure of the interfacial zone and the upper and lower profiles delimiting this zone are evident. While the interface as a whole moves, the interfacial region separating the domains of homogeneous phases exhibits irregular (chaotic) dynamics. Thus, one is led to focus on the two profiles that delimit the region of irregular dynamics. The treatment of the interfacial zone as a separate chaotic phase is supported by the existence of a transition above which the interface destabilizes and the chaotic phase invades the homogeneous regions [5,6]. We show that the relevant properties of the interface dynamics are captured by two coupled stochastic equations for the upper and lower profiles, respectively, rather than by a single stochastic equation for some average profile. In the analysis presented below, we identify the interfacial thickness Δ as another relevant order parameter which satisfies a closed equation. The validity of this scheme is confirmed both by the accuracy of its predictions below the critical point and by the agreement between the predicted and numerically determined scaling exponents.

The spatiotemporal dynamics of the system is described by the set of equations

$$z_{\mathbf{i}}^{t+1} = (1 - 4\varepsilon) f(z_{\mathbf{i}}^t) + \varepsilon \sum_{\mathbf{j} \in \mathcal{N}(\mathbf{i})} f(z_{\mathbf{j}}^t), \qquad (1)$$

for the real dynamical variables $z_{\mathbf{i}}$, where $\mathbf{i} = (i, j)$ are lattice-site labels, $\mathcal{N}(\mathbf{i})$ is the von Neumann neighborhood of site $\mathbf{i}$, and ε gauges the strength of the diffusive coupling. Moreover, $f(z)$ is a piecewise linear map of the unit interval [$f(z) = bz$ for $0 \le z \le 1/b$, and $f(z) = a$, otherwise]. The map parameters are chosen so that the local dynamics converges to a (super)stable period-3 cycle $A \longrightarrow B \longrightarrow C \longrightarrow A$, with $A = a$, $B = ba$, and $C = b^2 a$. (This regime is akin to the 3:1 resonance case of Ref. [3]). An interface separating domains of any two of the three phases may be introduced using initial conditions where half of the system is in one homogeneous phase and the remainder in another phase [7]. Periodic boundary

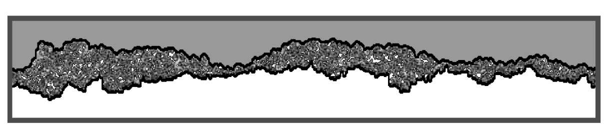

FIG. 1. Chaotic interfacial zone separating two homogeneous phases (solid gray and white regions). The thick black curves denote the upper and lower profiles.

conditions are used in the direction parallel to the interface while the system has infinite extent in the propagation direction. Periodicity of the homogeneous solution requires that the *AB* interface behaves similarly to *BC* and *CA* interfaces. Diffusive coupling induces a nontrivial dynamics of the interface separating adjacent phases. In particular, the interface can move with a nonzero velocity v_f. Even more striking is the spontaneous roughening observed in some parameter ranges (cf. Fig. 1) which emerges in spite of the determinism of the model and in the absence of any source of local chaos characterized by positive Lyapunov exponents [5]. It has been shown that the interface dynamics may be described by stochastic models driven by an effective noise resulting from the irregular motion inside the interface itself. Far from the transition, where the interfacial zone is very thin, the Edwards-Wilkinson (EW) model [8] describes the relevant features of the dynamics [9]; closer to the transition, where the interfacial zone is thick, this model breaks down. This breakdown is not due to the importance of KPZ-like nonlinear gradient terms as we shall argue in the following.

We investigate the interfacial dynamics along the line $(a = 0.1, \varepsilon = 0.173)$ in the (a, b, ε) parameter space, which illustrates all the relevant features including the growth and eventual destabilization of the interfacial zone. An examination of the velocity of the interface v_f allows one to make an accurate determination of the point b_c where the interfacial thickness Δ diverges, since this can occur only when $v_f = 0$ [6]. The velocity vanishes linearly with $b - b_c$, where $b_c = 2.54568(1)$. Figure 2 shows the stationary probability distribution $P(\Delta)$ for three values of b. The logarithmic vertical scale indicates an almost exponential decay of $P(\Delta)$ for large Δ, while the inset shows that an approximately power-law dependence is observed for small Δ. However, the most interesting feature is the superposition of the various curves after rescaling Δ by its average value $\Delta_0 \equiv \langle \Delta \rangle$ (here and in the following, angular brackets denote an average over space, time, and different realizations) and the probability density in the corresponding way. This indicates that the fluctuations of the thickness are described by a single scaling parameter [10], the critical behavior of which can be more effectively studied by computing the average thickness Δ_0 for different values of $b_c - b$. Figure 3 shows a doubly logarithmic plot of Δ_0 versus $b_c - b$ from which one may deduce that $\Delta_0 \propto (b_c - b)^{-\alpha}$ with $\alpha \approx 0.34 \pm 0.01$.

The stationary spatial correlation function of the thickness, $C_\Delta(x) = \langle \delta\Delta(x + x', t)\delta\Delta(x', t)\rangle - \langle(\delta\Delta)^2\rangle$, where $\delta\Delta = \Delta - \Delta_0$, decays exponentially well below the transition. This allows us to determine a correlation length ℓ that also appears to diverge when the critical point is approached. We find $\ell \approx (b_c - b)^{-\beta}$, with $\beta = 0.6 \pm 0.1$; the estimate of this exponent is much less reliable since each correlation length is the result of a fit to an exponential decay.

The simulation results presented above may be understood in terms of the following phenomenological model:

$$\begin{aligned} \partial_t h_1(x,t) &= D\partial_{xx}h_1 + F_1(h_1 - h_2) - v + \xi_1(x,t), \\ \partial_t h_2(x,t) &= D\partial_{xx}h_2 + F_2(h_1 - h_2) + v + \xi_2(x,t), \end{aligned} \tag{2}$$

where h_1 and h_2 denote the heights of the upper and lower profile, respectively, defined with respect to some preassigned reference position. These profiles may be unambiguously defined in view of the superstability of the homogeneous solutions. The model, consisting essentially of two coupled Edwards-Wilkinson equations, is the natural extension of the model successfully employed in Ref. [5] to reproduce the interface dynamics when the thickness can be neglected (i.e., $h_1 \approx h_2$). Symmetry arguments suggest that the diffusion coefficient D, the velocity v, and the statistical properties of the stochastic terms are the same in both equations. In fact, as long as the mutual coupling can be neglected, there is no way to distinguish the two profiles: the only difference is obviously the sign of the velocity, since the relative positions of the ordered and disordered phases are exchanged.

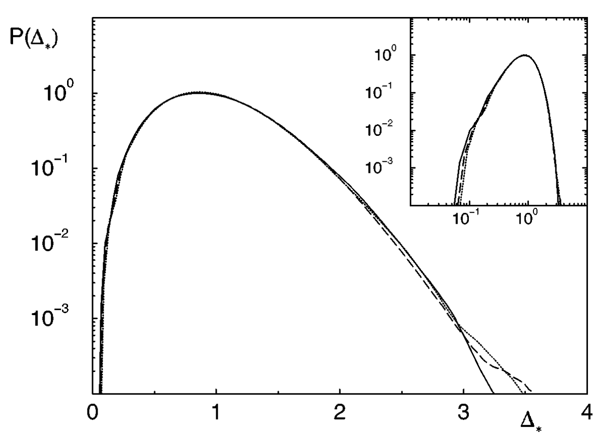

FIG. 2. $P(\Delta)$ versus $\Delta_- = \Delta/\Delta_0$ for three values of b: 2.544 (solid line), 2.542 (dashed line), and 2.54 (dotted line).

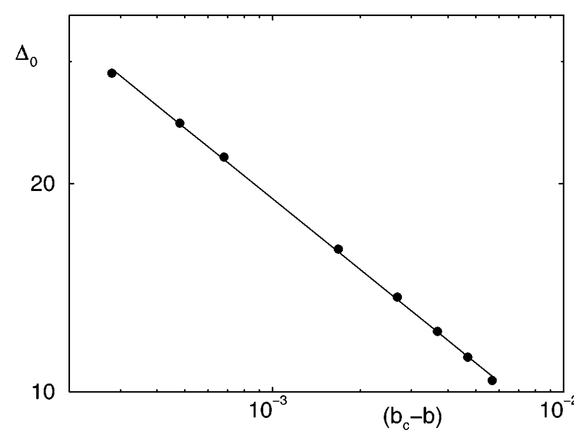

FIG. 3. Plot of Δ_0 versus $(b_c - b)$.

The coupling terms F_1 and F_2 have the net effect of producing a repulsive "force" necessary to prevent the two profiles from crossing. Because of translational invariance, F_1 and F_2 depend only on the local thickness $\Delta = h_1 - h_2$. With the above conventions on the sign of v, the oscillatory phases invade the chaotic interfacial phase; i.e., the two profiles approach one another. This happens until the short range coupling is sufficiently strong to stop the process. At that point, if and only if the coupling terms F_1 and F_2 are different, will the whole interface move with a nonzero velocity as observed in the numerical simulations. This is a genuine nonequilibrium feature of the interface dynamics.

In principle, one should add to each equation in (2) square gradient terms of the KPZ form. However, we argue below that the vanishing of v at the transition point makes such nonlinearities irrelevant. We note that this two-interface model differs from two coupled KPZ-type interfaces studied earlier in various contexts [11,12], since the two interfaces in our model move with opposite velocities and thus v cannot be removed by a simple scaling of h_1 and h_2.

Subtracting the second from the first equation in (2), one obtains a closed equation for the interface thickness,

$$\partial_t \Delta = D \partial_{xx} \Delta + F(\Delta) - u + \xi \,, \qquad (3)$$

where $F = F_1 - F_2$, $u = 2v$, and $\xi = \xi_1 - \xi_2$ is a Gaussian white noise with correlation function

$$\langle \xi(x,t)\, \xi(x',t') \rangle = 2\Gamma \delta(x - x') \delta(t - t') \,. \qquad (4)$$

This model is similar to the phenomenological equation introduced in Ref. [13] to study an equilibrium depinning transition. In the statistically stationary regime, the average interfacial thickness does not change, so that the average of Eq. (3) over the position along the interface x yields the relation $\langle F \rangle = u$.

Addition of the two equations in (2) yields an evolution equation for the mean profile which depends on Δ. The velocity v_f of the mean profile may be determined from an average over x and yields $v_f = \langle F_1 \rangle + \langle F_2 \rangle$.

In the subsequent analysis, we approximate the coupling term by a linear function centered around the average value in the stationary regime, $\langle \Delta \rangle = \Delta_0$: $F(\Delta) = F(\Delta_0) - g(\Delta - \Delta_0)$, where $g = F'(\Delta_0)$ is a positive constant. In this approximation $F(\Delta_0) = u$. Accordingly, Eq. (3) simplifies to

$$\partial_t \Delta(x,t) = D \partial_{xx} \Delta - g(\Delta - \Delta_0) + \xi(x,t) \,. \qquad (5)$$

By exploiting the periodic boundary conditions, the above equation can be solved by Fourier transforming in space, yielding an explicit expression for the space correlation function. For $x \ll L$, we find

$$C_\Delta(x) \simeq \frac{\Gamma}{4\sqrt{Dg}} \left(1 - e^{-\sqrt{g/D}x} \right). \qquad (6)$$

The purely exponential decay is in accord with numerical simulations, where it is observed even for early times. The measurements of the decay rate $\sqrt{g/D}$ and of the standard deviation $\Gamma/(4\sqrt{Dg})$ of Δ represent two independent constraints on the three parameters g, D, and Γ characterizing the model.

The additional information needed to determine all parameters can be obtained from the behavior of the temporal correlation function, $C_\Delta(t) = \langle \delta\Delta(x, t + t') \times \delta\Delta(x, t') \rangle$. An analysis similar to that performed for the spatial correlation leads to the asymptotic expression

$$C_\Delta(t) \simeq \frac{\Gamma}{4\sqrt{Dg}} \left[1 - \frac{2}{\pi} \, \mathrm{erf}(\sqrt{gt}\,) \right]. \qquad (7)$$

A one-parameter fit of $C_\Delta(t)$, together with the previous information, allows one to determine all parameters. The results for $b = 2.542$ are $D = 1.05$, $g = 0.0087$, and $\Gamma = 9.7$. These values are accurate to within 5% as confirmed by a more detailed analysis of the behavior of the low-k Fourier modes. Using these values, the theoretical and simulated correlation functions $C_\Delta(t)$ are reported in Fig. 4, where one sees that the model is able to reproduce both the asymptotic exponential behavior and the initial faster decay. This is still true rather close to the transition.

The only significant deviation between the theoretical predictions and the numerical observations concerns the tails of the probability distribution; however, this is expected in view of the failure of the linearization of F for Δ sufficiently different from Δ_0. In fact, the assumptions behind model (2) are further confirmed by the agreement between the diffusion constant as determined from the evolution of Δ and from the behavior of the Fourier modes of h_1 and h_2 (with the approach described in [5]). When the two profiles are sufficiently close to one another, the agreement is not obvious since a symmetry breaking occurs as indicated by the nonzero velocity of the entire interface.

We observed that both Δ and the correlation length ℓ parallel to the interface diverge as the critical point is approached. In view of the results presented above, we might expect that system properties will remain

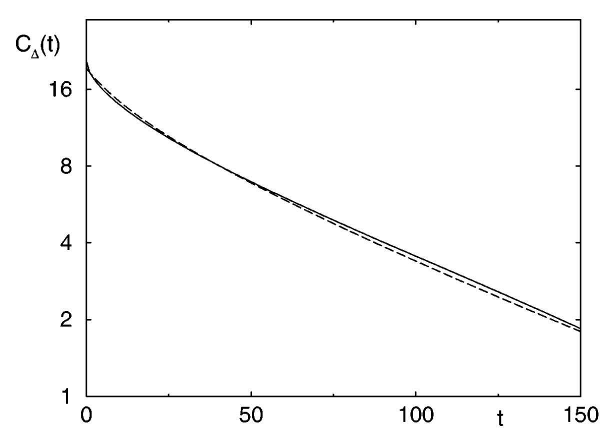

FIG. 4. Temporal correlation function $C_\Delta(t)$ versus t. Solid curve (simulation); dashed curve [Eq. (7)].

invariant in the vicinity of the critical point provided all variables are suitably rescaled. Let us first perform the scaling analysis with reference to a repulsive force of the type $F(\Delta) = c/\Delta^\eta$, since it is naturally scale invariant. Moreover, the relevant control parameter $(b_c - b)$ may be replaced by the velocity u since this also vanishes linearly with $(b_c - b)$.

We assume that the basic quantities scale with u as follows: $\Delta = \Delta'/u^\alpha$, $t = t'/u^z$, $x = x'/u^\beta$. The scaled equation is

$$u^{z-\alpha}\partial_{t'}\Delta' = u^{2\beta-\alpha}D\partial_{x'x'}\Delta' + u^{\alpha\eta}c/\Delta'^\eta - u + u^{(z+\beta)/2}\xi\,. \tag{8}$$

Requiring that the model (except for the force that will be considered separately) be invariant to this transformation yields the following relations among the exponents:

$$\begin{aligned} 2\beta - z &= 0\,,\\ 1 + \alpha - z &= 0\,,\\ \alpha - z/2 + \beta/2 &= 0\,, \end{aligned} \tag{9}$$

giving, $\alpha = 1/3$, $z = 4/3$, $\beta = 2/3$. With these values of the critical indices, one also finds that the coefficient of the force scales as $c' = cu^{\eta/3-1}$. Accordingly, if $\eta > 3$ the repulsive potential renormalizes to an infinitely high barrier at $\Delta = 0$, so that the entire equation (8) is invariant under the renormalization transformation. It is obvious that forces decaying faster than algebraically yield the same scenario.

For $\eta < 3$, the force diverges, indicating that α must be larger than 1/3: the only physically consistent solution is obtained by assuming that in this regime it is the stochastic term which is negligible. As a result, in the rescaled units ($\Delta' = u^{1/\eta}\Delta$), the probability distribution $P(\Delta)$ becomes increasingly δ-like. The excellent invariance of $P(\Delta)$ observed in Fig. 2 is thus an indirect indication that the force F decays at least as fast as $1/\Delta^3$.

The exponent α can be most confidently tested against our numerical data and the numerical value obtained from the analysis of Fig. 3, $\alpha = 0.34$, is indeed extremely close to the predicted value 1/3. The numerical estimate of the scaling exponent of the correlation length ℓ, $\beta \approx 0.6$, also agrees with the predicted value $\beta = 2\alpha = 2/3$, within the estimated error bounds.

Only for $\eta = 3$ does the force term neither vanish nor diverge: this is the scenario implicitly assumed by the model in Eq. (5), since the dynamics is determined by a balance between the repulsive force F and the attractive contribution originating from u. The accuracy of the linear model even relatively close to the transition point can be taken as an indication that the force presumably decays only slightly faster than $1/\Delta^3$.

In general, one should also include lateral-growth processes, which give rise to an additional KPZ-type nonlinearity [4]. This term may modify the critical behavior as shown in [14] for a directed percolation problem. However, in our model, the coefficient of the nonlinear term turns out to be proportional to the average velocity v, since the lateral and the forward growth of the profile are both ruled by the very same mechanisms. This feature follows from the isotropic growth of the profiles observed in Ref. [5]. Accordingly, scaling analysis shows that the nonlinear term vanishes as $u^{2/3}$ and is thus irrelevant.

The analysis shows that the critical behavior of the chaotic interfacial zone separating two phases of an oscillation may be described by a set of coupled stochastic equations for the two interfacial profiles that define the zone. Complex interfacial dynamics combined with instabilities are features common to many chemical and physical systems and the analysis presented here may find application in other contexts.

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