

## Interfacial instability induced by external fluctuations

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(Received 4 January 1995)

The dynamics of an interface separating the two coexistent phases of a binary system in the presence of external fluctuations in temperature is studied. An interfacial instability is obtained for an interface that would be stable in the absence of fluctuations or in the presence of internal fluctuations. Analytical stability analysis and numerical simulations are in accordance with an explanation of these effects in terms of a quenchlike instability induced by fluctuations.

PACS number(s): 05.40.+j, 68.10.Cr

### I. INTRODUCTION

The study of the effects of fluctuations on interfacial instabilities under nonequilibrium conditions [1–9] constitutes an active field of research both experimentally [10–14] and theoretically [15–21]. The amplification of fluctuations seems to be an important ingredient in the explanation of phenomena such as secondary fingering in Hele-Shaw fingers [9], instabilities in Rayleigh-Benard convection [20] and Taylor-Couette flow [21], and directional solidification and dendritic growth [10–19]. Nevertheless, a theoretical framework which includes the effects of fluctuations on instability dynamics is only starting to be developed. Moreover, there has been some theoretical work in the context of external noise on spatially extended systems [22–25] in the direction to explain the discrepancies between the experimental and theoretical results. First, it has been predicted a shift in the transition point of the Swift-Hohenberg [22,23] and Ginzburg-Landau models [23,24]. It has also been discussed the existence of an instability point induced by noise [25].

In fact, studies on several systems have shown that in most cases the intensity of thermal noise is not large enough to account for the observed dynamics [16,20,21], so other sources of noise (like inhomogeneities or external fluctuations) have to be invoked. In any case external fluctuations can be imposed on a system through a control parameter and, in a certain degree, are unavoidable when a system is kept out of equilibrium. In this context, there are some experiments in which isolated or periodic heat pulses [11,12] or modulation of some other parameter [13,14] have been employed. Also, the introduction of fluctuations in a experimentally controlled way seems to be an important step in order to clarify the role of noise.

In a problem of interfacial stability, the state of the system is usually controlled by the value of a suitable control parameter. In the usual case the relevant problem is the morphological stability of a planar interface given a fixed and constant value of the control parameter. Nevertheless a quench, that is a sudden change in the value of

the control parameter, leads the system to evolve to a different state, and this evolution can affect in several cases the transient stability of the interface. For example, in a directional solidification problem, where after a sudden change in the pulling velocity the system evolves to a cellular pattern, the dynamical evolution of the concentration profile determines the periodicity of the first unstable mode of the interface [18,26]. In the case of a binary mixture, with an interface separating two phases, after a quench in temperature the final equilibrium state is a flat interface separating the two coexisting phases, but the evolution to this state induces a transient interface instability. [27]

Now we are interested in the effects of temperature fluctuations on the morphological stability of the interface separating the two phases of a binary system. The experimental situation we are thinking about corresponds to a system that is kept at a given temperature  $T$  near the critical point  $T_c$  by an external device, in such a way that this temperature fluctuates around its mean value  $T_0$ . These fluctuations are external, in the sense that are caused by the experimental setup and do not correspond to the thermal fluctuations of the binary system.

In Sec. II of this paper we obtain a Ginzburg-Landau model for the dynamics of a binary mixture in the presence of external fluctuations in temperature. These fluctuations appear as a multiplicative noise in the equation and drive the system to a new nonequilibrium state with a stable interface. The interesting point is that such dynamics, like in the case of a quench, does induce the transient appearance of an interfacial instability. This is shown in Sec. III, where we perform a stability analysis of the interface separating the two phases of the mixture. These results should apply on an equal basis to any system governed by an equation of the Ginzburg-Landau type, with a phase transition with conserved order parameter, in which external fluctuations are present in the corresponding control parameter. In Sec. IV we present numerical simulations of the model, obtaining a

good agreement with the theoretical analysis. The conclusions are presented in Sec. V.

## II. MODEL

We start from the conserved time-dependent Ginzburg-Landau equation,

$$\partial_{\bar{t}}\bar{c}(\bar{\mathbf{x}}, \bar{t}) = M\bar{\nabla}^2[-r\bar{c}(\bar{\mathbf{x}}, \bar{t}) + u\bar{c}(\bar{\mathbf{x}}, \bar{t})^3 - k\bar{\nabla}^2\bar{c}(\bar{\mathbf{x}}, \bar{t})], \quad (2.1)$$

where  $\bar{c}$  is the local concentration,  $M$  is the mobility, and  $r$ ,  $u$ , and  $k$  are phenomenological parameters. Near the critical point  $T_c$  the parameter  $r$  is usually assumed to be linear in temperature in the form

$$r(T) = \alpha(T_c - T). \quad (2.2)$$

This kind of equation has been widely used for the study of the dynamics of first order phase transitions with conserved order parameter [28], and in the context of binary mixtures was introduced by Cahn and Hilliard [29]. In the classification of Hohenberg and Halperin this is known as the model *B* of critical dynamics [30].

In an experiment the temperature  $T$  can be externally imposed on the system. In such a situation  $T$  acts as a control parameter. When it is externally changed the system gets different equilibrium values for the bulk concentrations. For a temperature below the critical point, the equilibrium situation corresponds to two coexisting phases with the usual kink concentration profile in the normal direction to the interface, which is flat and morphologically stable.

We now consider local fluctuations in temperature in the following way:

$$T(\bar{\mathbf{x}}, \bar{t}) = T_0 + \delta T(\bar{\mathbf{x}}, \bar{t}), \quad (2.3)$$

$\delta T(\bar{\mathbf{x}}, \bar{t})$  is a stochastic field with zero mean and variance  $\langle \delta T^2 \rangle$  that represents the fluctuating variations of the local temperature. Spatial and temporal correlations in the temperature field are given by the correlation length  $\lambda$  and the correlation time  $\tau$ , which should be finite but that will be assumed to be smaller than the typical scales of the system. In this limit correlations of  $\delta T$  could be written in the form

$$\langle \delta T(\bar{\mathbf{x}}, \bar{t}) \delta T(\bar{\mathbf{x}}', \bar{t}') \rangle = 2\bar{\epsilon} \delta(\bar{\mathbf{x}} - \bar{\mathbf{x}}') \delta(\bar{t} - \bar{t}'), \quad (2.4)$$

where  $\bar{\epsilon} \sim \tau \lambda^d \langle \delta T^2 \rangle$ ,  $d$  being the dimension of the system. In spite of using Eq. (2.4), a finite but small correlation length will be implicitly assumed through the employ of a lattice in the numerical calculation. This does not change any fundamental point of our analysis. We see that external thermal fluctuations appear in the Ginzburg-Landau equation (2.1) in the coefficient that multiplies the linear term around its mean value  $r_0 = \alpha(T_c - T_0)$ . Fluctuations coming from this term are thus multiplicative,

which physically means that are coupled to the state of the system.

With the changes of variables

$$\begin{aligned} \mathbf{r} &= \left(\frac{r_0}{k}\right)^{1/2} \bar{\mathbf{x}}, \\ t &= \frac{Mr_0^2}{k} \bar{t}, \\ c &= \left(\frac{u}{r_0}\right)^{1/2} \bar{c}, \end{aligned} \quad (2.5)$$

we are lead to the following dimensionless equation:

$$\begin{aligned} \partial_t c(\mathbf{r}, t) &= \nabla^2[-c(\mathbf{r}, t) + c(\mathbf{r}, t)^3 - \nabla^2 c(\mathbf{r}, t)] \\ &\quad - \nabla^2 [\xi(\mathbf{r}, t) c(\mathbf{r}, t)], \end{aligned} \quad (2.6)$$

where the field  $\xi(\mathbf{r}, t) = \delta T / (T_c - T_0)$  is taken as Gaussian fluctuations with zero mean and correlation given by

$$\langle \xi(\mathbf{r}, t) \xi(\mathbf{r}', t') \rangle = 2\epsilon \delta(\mathbf{r} - \mathbf{r}') \delta(t - t'). \quad (2.7)$$

Due to the multiplicative character of the noise term in Eq. (2.6), it has a nonzero mean value. This equation can be used now to study the stability of the interface separating the two phases of the system in the presence of external fluctuations. The usual derivation of the dispersion relation for the Mullins-Sekerka instability [31] assumes a constant ramp for the concentration profile in time. In the next section we obtain that these fluctuations change the concentration profile from the usual kink solution of a stable interface to a ramp structure which decreases in time. This ramp is analogous to what would be obtained after a quench between two temperatures below the critical point [27], and induces a flux of matter across the interface which makes the interface unstable. Note that the interface would be stable in the presence of internal fluctuations, which would appear as an additive noise in the model, or in the absence of fluctuations.

## III. THEORETICAL ANALYSIS

The formal integration of Eq. (2.6) gives the following expression for the concentration  $c_\mu(t + \Delta t)$  at a point  $\mu$  [32]:

$$\begin{aligned} c_\mu(t + \Delta t) &= c_\mu(t) + \Delta t \nabla_{\mu\alpha}^2 [-(1 + D)c_\alpha + c_\alpha^3 - \nabla_{\alpha\sigma}^2 c_\sigma] \\ &\quad + \nabla_{\mu\alpha}^2 c_\alpha X_\alpha(t), \end{aligned} \quad (3.1)$$

where  $D \equiv 4\epsilon / (\Delta x)^4$  and  $X_\alpha(t) = \frac{\sqrt{2\epsilon\Delta t}}{\Delta x} \theta_\alpha$ .  $\theta_\alpha$  are Gaussian random numbers with zero mean and variance equal to 1. The presence of a multiplicative noise term gives rise to two terms in Eq. (3.1). The one proportional to  $D$  is the so-called Stratonovich term. This contribution corresponds to the mean value of the noise term of Eq. (2.6), which is nonzero owing to the multiplicative character of the noise. The second term introduced by the multiplicative noise, the last one in Eq. (3.1), is a random term with zero mean.

By inspection of Eq. (3.1) we see that the Stratonovich

term gives rise to an increase in the coefficient of the linear term. In the absence of the random number term, the corresponding bulk values for the concentration would be  $c_{eq} = \pm\sqrt{1+D}$ . The presence of the random number term simply corrects these concentration values  $c_{eq}$  to lower values. We conclude that the effect of the noise is equivalent to a quench  $\Delta T$  in temperature, proportional to the amplitude of the external fluctuations. The system, that is initially placed at the concentration values  $\pm 1$  has to evolve to the new concentration values  $\pm c_{eq}$ . Then, a flux of matter will be generated through the interface in such a way that very close to the interface the concentration should take very shortly the values  $\pm c_{eq}$ . That will lead to the appearance during a certain time of a ramp in the concentration profile and, as a result, an interfacial instability is expected to occur. The final nonequilibrium steady state generated by the noise corresponds to a kink solution with bulk values  $\pm c_{eq}$ . It is worth mentioning that the change in the concentration value  $c_{eq}$  associated to a fix quench is larger near the critical temperature. In fact, it could be calculated that  $D$  is proportional to  $\Delta T/(T_c - T_0)$ .

In Fig. 1 we present numerical results obtained from Eq. (3.1) for the concentration profile of a planar interface separating two coexisting phases with initial concentration values  $\pm 1$ . After a short transient, we obtain a ramp structure. In this figure the results for two different cases, with and without the random number term, are shown. We obtain that the ramps are qualitatively similar, but the slope is lower in the second case.

In view of the above discussion, we expect that the effects of the noise on the interfacial instability come essentially from the Stratonovich term in Eq. (3.1) and that the random term will not introduce any new relevant effect, apart from numerical corrections. Therefore in the

following analysis we will employ Eq. (3.1) without the random term:

$$c_\mu(t + \Delta t) = c_\mu(t) + \Delta t \nabla_{\mu\alpha}^2 \left[ -(1+D)c_\alpha + c_\alpha^3 - \nabla_{\alpha\sigma}^2 c_\sigma \right]. \quad (3.2)$$

The Cahn-Hilliard equation associated to Eq. (3.2) is

$$\frac{\partial c}{\partial t} = \nabla^2 (-c_{eq}^2 c + c^3 - \nabla^2 c). \quad (3.3)$$

One of the ingredients of the derivation of the dispersion relation corresponding to Eq. (3.3) is the assumption of a dynamic expression for the concentration profile of a planar interface,  $c_p(y, t)$ . To do so, first we study the temporal evolution of this magnitude numerically using Eq. (3.2). In Fig. 2 we present the results for  $\epsilon = 0.004$ . We obtain that the ramp decreases in time. We can write

$$c_p(y, t) = c_0(y) + u(y, t), \quad (3.4)$$

where  $c_0(y)$  is the equilibrium concentration profile, the kink solution:

$$c_0(y) = c_{eq} \tanh \left( \frac{c_{eq}}{\sqrt{2}} y \right). \quad (3.5)$$

By substituting Eq. (3.4) in Eq. (3.3) we obtain, up to linear order in  $u$ :

$$\frac{\partial u}{\partial t} = 2c_{eq}^2 \nabla^2 u. \quad (3.6)$$

This is a diffusion equation with the boundary condi-

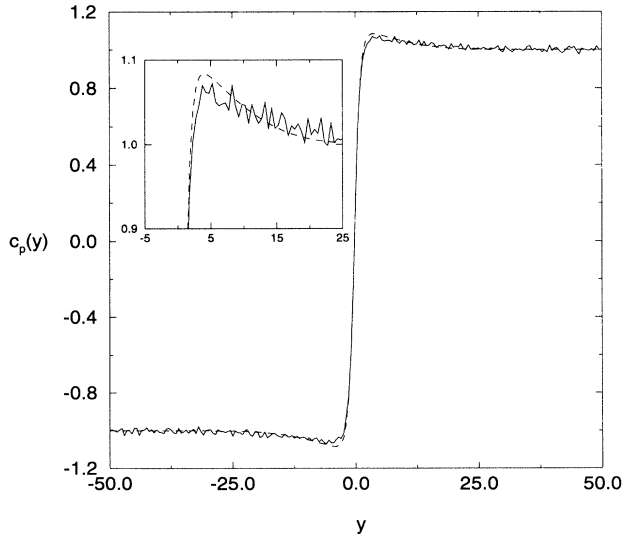


FIG. 1. Profiles  $c_p(y)$  for time  $t = 25$  and intensity  $\epsilon = 0.004$ . Solid line corresponds to Eq. (3.1); dashed line corresponds to Eq. (3.2).

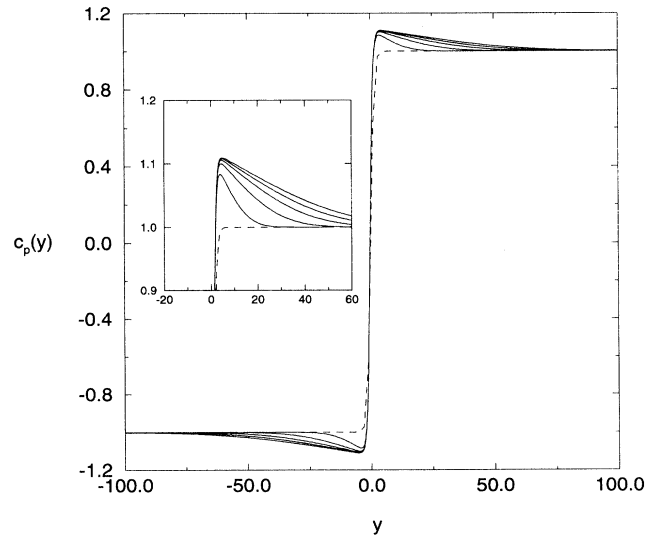


FIG. 2. Temporal evolution of the concentration profile from Eq. (3.2) for  $\epsilon = 0.004$  at  $t = 25, 100, 200, 300, 400$ . Dashed line is the kink solution of the deterministic system.

tions  $u(0^\pm, t > 0) = 0$ ,  $u(\pm\infty, t) = \mp(c_{eq} - 1)$  and  $\frac{\partial u}{\partial y}(\pm\infty, t) = 0$ . The exact solution of Eq. (3.6) with appropriate initial conditions has been obtained in Ref. [27]. Near the interface, we can write

$$c_p(y, t) \sim c_0(y) - A(t)y \quad (3.7)$$

for  $y \in [-L(t), L(t)]$ , where  $A(t)$  is the slope at the origin:

$$A(t) = \left. \frac{\partial u(y, t)}{\partial y} \right|_{y=0} = \frac{c_{eq} - 1}{c_{eq}\sqrt{2\pi}} t^{-1/2}. \quad (3.8)$$

In Fig. 3 we present the comparison between  $c_p(y)$  obtained from Eq. (3.2) and our theoretical assumption, Eqs. (3.7) and (3.8), for different times. We obtain a good accordance.

To determine the dispersion relation, first we derive a macroscopic model from the mesoscopic model, Eq. (3.3). The macroscopic model consists of three equations. The first one is the diffusion equation for  $\delta c(\mathbf{r}, t)$ , the deviation of the concentration profile with respect to the flat interface:

$$\delta c(\mathbf{r}, t) = c(\mathbf{r}, t) - c_p(y, t) = c(\mathbf{r}, t) \mp c_{eq} + A(t)y, \quad (3.9)$$

where in Eq. (3.9) we have taken the limit of sharp interface [33] for  $c_p(y, t)$  and  $\mp$  applies to each bulk phase, namely,  $A$  and  $B$ , respectively. The equation for  $\delta c(\mathbf{r}, t)$  is

$$\frac{\partial}{\partial t} \delta c(\mathbf{r}, t) = 2c_{eq}^2 \nabla^2 \delta c(\mathbf{r}, t). \quad (3.10)$$

The second equation is associated to the boundary

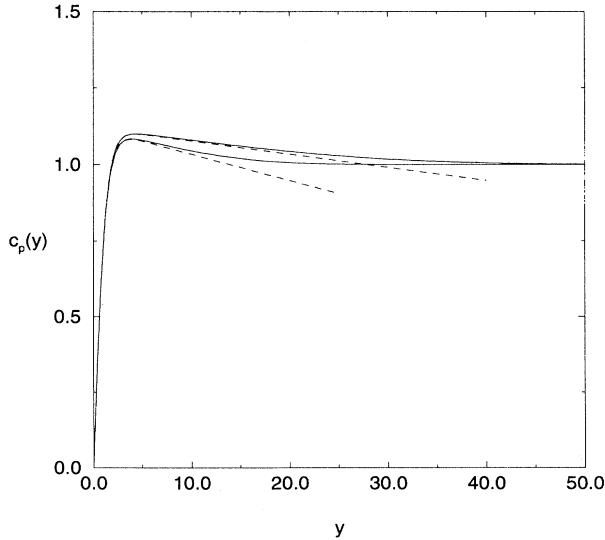


FIG. 3. Comparison between the concentration profiles  $c_p(y)$  from the numerical integration of Eq. (3.2) (solid lines), and the approximation given by Eq. (3.7) (dashed lines), for  $\epsilon = 0.004$  and  $t = 25, 100$ .

conditions at the interface. This is the so-called Gibbs-Thomson equation, which relates the deviation of the concentration at the interface owing to the local curvature  $\mathcal{K}$ . From Eq. (3.3) we obtain [34]

$$\delta c(\mathbf{r}, t)|_{int} = \frac{\sigma}{4c_{eq}^3} \mathcal{K}, \quad (3.11)$$

where  $\sigma = \int (\frac{dc_0(y)}{dy})^2 dy = \frac{2\sqrt{2}}{3} c_{eq}^3$  is the surface tension and  $c_0(y)$  is given by Eq. (3.5). Finally, the third equation is the continuity equation, which yields the motion of the interface due to an imbalance of fluxes. The normal velocity to the interface  $v$  is

$$2c_{eq}v = \hat{\mathbf{n}} \cdot (\mathbf{J}_B - \mathbf{J}_A)|_{int}, \quad (3.12)$$

where  $\hat{\mathbf{n}}$  is the unitary vector normal to the interface directed toward the  $B$  phase.  $\mathbf{J}_A$  and  $\mathbf{J}_B$  are the fluxes [ $\mathbf{J} = -2c_{eq}^2 \nabla c(\mathbf{r}, t)$ ] when one moves toward the interface from the  $A$  and  $B$  phases, respectively. To do the stability analysis, one considers a perturbation of the planar interface  $h(x, t) = h_k(t) \exp(ikx)$  where  $k$  is the wave vector of the perturbation and looks for a solution of the form:

$$\delta c(\mathbf{r}, t) = \delta c_0(t) \exp(\mp qy) h(x, t). \quad (3.13)$$

By substituting Eq. (3.13) in Eq. (3.10) and using the usual quasistatic approximation ( $\partial \delta c / \partial t = 0$ ) [7] we obtain  $q = k$ . Furthermore, to determine  $\delta c_0(t)$ , we substitute Eq. (3.13) in the Gibbs-Thomson relation and we use a linearized expression for the curvature  $\mathcal{K} = h_{xx} = -k^2 h_k \exp(ikx)$ :

$$\delta c_0(t) = -\frac{\sigma}{4c_{eq}^3} k^2 + A(t). \quad (3.14)$$

Finally, by taking into account that  $v = \partial h(x, t) / \partial t = (dh_k / dt) \exp(ikx)$ , we could write the continuity equation as

$$\frac{1}{h_k} \frac{dh_k}{dt} = -\frac{\sigma}{2c_{eq}^2} k^3 + 2c_{eq} A(t)k. \quad (3.15)$$

By using Eq. (3.8), the integration of Eq. (3.15) gives

$$h_k \sim \exp \left[ -\frac{\sigma}{2c_{eq}^2} k^3 t + \frac{4(c_{eq} - 1)}{\sqrt{2\pi}} k t^{1/2} \right]. \quad (3.16)$$

Equation (3.16) is one of the most important results of this paper [35]. We obtain that apart from the usual stabilizing mechanism of surface tension, associated to the term proportional to  $k^3 t$  in the exponential, there is a destabilizing mechanism, associated to the term proportional to  $kt^{1/2}$ . This last term is zero when there are no external fluctuations acting on the system. Furthermore, the effect of the destabilizing mechanism decreases in time. Then, the result implies that a stable interface in an equilibrium state would become unstable if an external noise acts on it and the system would evolve to a new nonequilibrium steady state. This result is different from the usual Mullins-Sekerka instability which gives for long wavelength a purely exponential growth

in time for the linear regime. In a real experiment, the gradients of concentration are reduced with time and for long times the interface would become finally stable as we have obtained in Eq. (3.16). Additionally, we could define a crossover time between the two different regions,  $t_c$ , as the maximum of Eq. (3.16):

$$t_c^{1/2} = 2\sqrt{\frac{2}{\pi} \frac{(c_{eq} - 1)c_{eq}^2}{\sigma}} k^{-2}. \quad (3.17)$$

We obtain that  $t_c$  increases with the perturbation wavelength.

#### IV. NUMERICAL RESULTS AND DISCUSSION

We have performed simulations of Eq. (2.6) in a two dimensional lattice of  $L_x = 112$  and  $L_y = 400$  with a mesh size  $\Delta x = \Delta y = 0.5$  and  $\Delta t = 0.001$ . In Fig. 4 we present the evolution of  $h_k$  given by Eqs. (3.1) and (3.2) with an initial perturbation  $h_k(0) = 10 \Delta x$  and wave number  $k = 0.112$ . Initially the concentration of the two phases is  $\pm 1$  and then we apply an external noise of different intensities. The results (a) and (b) of Fig. 4 correspond to the numerical integration of Eq. (3.2) for  $\epsilon = 0.003$  and  $\epsilon = 0.004$ , respectively, while (c) corresponds to the numerical integration of Eq. (3.1) for  $\epsilon = 0.004$ . In the three cases the system is unstable during a transient, in accordance to the theoretical result of Eq. (3.16), indicating that the mechanisms drawn in Sec. III are correct. However the actual evolutions are quantitatively different in (b) and (c), in spite of corresponding to the same value of  $\epsilon$ . This disagreement should come from their different equilibrium values  $c_{eq}$ . In fact, results from Eq. (3.1) should be compared to the numerical integration of

TABLE I. Numerical and theoretical values of parameters  $C_1$  and  $C_2$ , for cases (a) and (b) of Fig. 4.

$\epsilon$	$C_1$ (theor.)	$C_1$ (numer.)	$C_2$ (theor.)	$C_2$ (numer.)
0.003 (a)	0.00073	0.00074	0.015	0.019
0.004 (b)	0.00077	0.00079	0.020	0.025

Eq. (3.2) for a different value of  $\epsilon$  chosen to give the same value of  $c_{eq}$ . We show as (d) in Fig. 4 the evolution of Eq. (3.2) for  $\epsilon = 0.0026$ , for which  $c_{eq} = 1.08$  as in case (c). The agreement between both results is quite good. In Fig. 4 we also include a numerical fit of an expression of the form:

$$h_k = C_0 \exp[-C_1 t + C_2 (t - t_0)^{1/2}], \quad (4.1)$$

where  $t_0$  has been included to take into account the initial transient in which the ramp is formed. In Table I we present the numerical values of  $C_1$  and  $C_2$ . Furthermore, we include the theoretical values of  $C_1$  and  $C_2$  obtained from Eq. (3.16) corresponding to the cases (a) and (b) of Fig. 4.

#### V. SUMMARY

In summary, we have introduced external temperature fluctuations in a system constituted by two phases separated by an interface. These fluctuations appears as a multiplicative noise in the corresponding Ginzburg-Landau equation. The interfacial stability analysis shows that, apart from the usual stabilization term due to surface tension, there is a transient term that acts as a destabilizing mechanism and it is due to the external fluctuations. This theoretical result has been corroborated by numerical simulations of the model. It would be of interest to consider the effect of fluctuations of the type considered in this paper in the case of domain growth in phase separation.

#### ACKNOWLEDGMENTS

We thank J. Casademunt for helpful discussions. J.L.M. and A.H.M. thank the Direcció General de Investigaci6 Científica y T6cnica (Spain) (Project No. PBR93-0054-C02-01) and NATO (Collaborative Research Grant No. 931018) for support. A.M.L. and L.R.P. thank the Direcció General de Investigaci6 Científica y T6cnica (Spain) (Project No. PB93-0769-C02-02) for support. We also acknowledge the Centre de Supercomputaci6 de Catalunya (CESCA) for computing support.

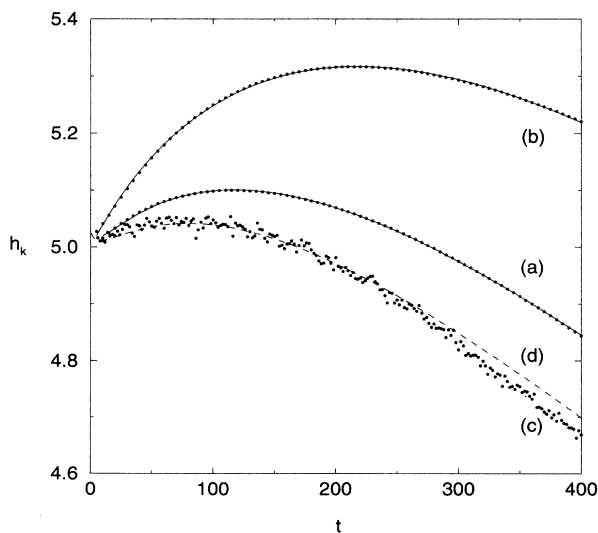


FIG. 4. Temporal evolution of  $h_k$  from the numerical integration of Eq. (3.2) for (a)  $\epsilon = 0.003$ , (b)  $\epsilon = 0.004$ , and (d)  $\epsilon = 0.0026$ ; and Eq. (3.1) for (c)  $\epsilon = 0.004$ . The solid lines correspond to a fit given by Eq. (4.1).

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