Numerical simulation of a continuum model of growth of thin composite films

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We present the results of extensive numerical integration in $(1+1)$ dimensions of a set of equations that couple the Kardar-Parisi-Zhang (KPZ) equation to the time-dependent Ginzburg-Landau (TDGL) equation, recently proposed for modeling the growth of thin composite solid films. We find that for times t shorter than a crossover time t_c the mean domain size $L(t)$ grows logarithmically with the time, whereas for $t \gg t_c$ $L(t)$ grows as t^{1/z_m}, with z_m being *nonuniversal* and depending on the parameters of the model. The roughness exponent is also found to be nonuniversal. Thus, neither the dynamics of the domains' growth is governed by the TDGL equation, nor is the scaling of the surface roughness described by the KPZ equation.

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I. INTRODUCTION

Thin solid films with specific electronic, optical, and mechanical properties have wide applications in science and technology. To produce such films, molecular beam epitaxy (MBE) and vapor deposition have been used in the past. In MBE, for example, particles are deposited [1] on a surface through a directed beam, which then diffuse on the surface until they reach an energetically favorable position. However, due to a variety of reasons, deposition of only one type of particle may not yield a thin solid film with the desired electrical and optical properties. For example, a continuous flux of the incoming particles may cover a deposited particle which is not, however, in a minimum energy state. As a result, the surface of the film may reach a steady state in which the width of the film is saturated, but the surface itself is not in equilibrium. Due to such shortcomings, it has become a common practice to grow thin composite solid films using more than one type of particle [1,2].

Formation of thin composite solid films indicates that their growth may give rise to interesting and nontrivial problems and, therefore, in addition to the practical applications of such films, modeling of their growth is also of great current interest. Several models have recently been proposed. In particular, Desai and co-workers [3,4] have proposed models for the growth of thin composite films by a MBE process, made of two types of particles, say A and B, in which the interaction of the two types of the particles leads to phase separation and formation of domains. More recently, Drossel and Kardar [5,6] proposed an interesting model for the growth of thin composite films by vapor deposition. In their model, phase separation is characterized by an order parameter $m(\mathbf{x},t)$ which represents the difference between the densities of the A and B particles at the surface at position $\mathbf{x}$ at time t. The growth of the surface height $h(\mathbf{x},t)$ is described by the Kardar-Parisi-Zhang (KPZ) equation [7],

$$\frac{\partial h}{\partial t} = \nu\nabla^2 h + \frac{1}{2}\lambda(\nabla h)^2 + \frac{1}{2}\chi m^2 + \eta_h(\mathbf{x},t), \tag{1}$$

which is the standard KPZ equation, together with the coupling term $\frac{1}{2}\chi m^2$. Here, η_h represents thermal fluctuations of the incoming particles' flux with

$$\langle \eta_h(\mathbf{x},t)\cdot\eta_h(\mathbf{x}',t')\rangle = 2D_h\delta^d(\mathbf{x}-\mathbf{x}')\delta(t-t'). \tag{2}$$

The governing equation for the order parameter is the time-dependent Ginzburg-Landau (TDGL) equation,

$$\frac{\partial m}{\partial t} = K(\nabla^2 m + rm - um^3) + a\,\nabla h\cdot\nabla m + bm\nabla^2 h + \frac{1}{2}\mathrm{cm}(\nabla h)^2 + \eta_m(\mathbf{x},t), \tag{3}$$

where the general forms of the three physically motivated coupling terms are obtained by symmetry arguments, and η_m represents the fluctuations in the density of the incoming particles with

$$\langle \eta_m(\mathbf{x},t)\cdot\eta_m(\mathbf{x}',t')\rangle = 2D_m\delta^d(\mathbf{x}-\mathbf{x}')\delta(t-t'). \tag{4}$$

The signs of the various coefficients that appear in Eqs. (1) and (3) are important and, thus, are described here. For stability, one must have $\nu>0$, $u>0$, and $K>0$. The mapping, $h\to -h$, changes the sign of λ and some of the other terms. With this mapping in mind, one can set $\lambda>0$, although, in principle, one may also set $\lambda=0$. The same is true for the coefficient c, as it plays the same role in the TDGL equation that λ plays in the KPZ equation. The coefficient χ determines whether growth takes place mostly on ordered domains or on top of the domain walls. Thus, the two possible signs of χ correspond to different possible physical situations. As in the case of the standard TDGL model, the phase transition at the mean-field level takes place when the sign of r changes from positive to negative. However, since the one-dimensional (1D) phase transition is very much affected by fluctuations, $r=0$ will not be the marginal case, rather the phase transition occurs for some $r>0$. Thus, it is prudent to set r at a positive value (which is what we do in

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this work; see below). The parameter a determines whether domain walls move uphill $(a<0)$ or downhill $(a>0)$. On the other hand, the coefficient b determines the effect of curvature on the domain growth, and hence one typically has, $b>0$.

Drossel and Kardar [5] did not study the behavior of their model by direct numerical integration of Eqs. (1) and (3). Instead, they simulated a discrete model in $(1+1)$ dimensions using a "brick wall" restricted solid-on-solid model, which supposedly simulates the growth of the same thin composite film modeled by Eqs. (1) and (3). In their model, one starts from a flat surface and adds particles such that no overhangs are created, with the center of each particle being atop the edge of the two particles in the layer below. If the two particles in the layer below are of the same type, say A, then a newly incoming particle will become of type B with probability $p=\exp(-4J/k_BT)$, and of type A with probability $1-p$, where J is the 1D Ising chain coupling constant, k_B is the Boltzmann's constant, and T is the temperature. Thus, the dynamics of the order parameter in the Drossel-Kardar rule-based model is the same as that of the 1D Glauber model for which the order parameter dynamic critical exponent z_m, defined by

$$L(t) \sim t^{1/z_m}, \tag{5}$$

is $z_m=2$, where $L(t)$ is the average domain size at time t. Drossel and Kardar [5] computed z_m and the roughness exponent α for their rule-based model and found that $z_m \simeq 1.85$, and that $\alpha=1$ for length scales that are up to ξ, the correlation length of the order parameter (which is also the mean size of the domains), whereas $\alpha=1/2$ for length scales $\gg \xi$, which is the usual roughness exponent for the KPZ equation in $1+1$ dimension.

In this paper, we investigate the scaling properties of the Drossel-Kardar continuum model by direct numerical integration of Eqs. (1) and (3). We show that, at least in $(1+1)$ dimensions, the evolution of the system governed by Eqs. (1) and (3) gives rise to *nonuniversal* scaling properties for the growing films, and that there are interesting crossover effects in the model that need to be studied carefully. The plan of this paper is as follows. In Sec. II we describe the numerical technique that we use to integrate Eqs. (1) and (3). The results are then presented and discussed in Sec. III.

II. THE NUMERICAL INTEGRATION

To carry out the numerical integration of Eqs. (1) and (3), we first note that because the amplitudes D_h and D_m have different dimensions ($[D_h]\equiv L^3t^{-1}$ and $[D_m]\equiv Lt^{-1}$), we can use them for making these equations dimensionless. Hence, defining,

$$\begin{cases} \tilde{x} = x\sqrt{\dfrac{D_m}{D_h}} \\ \tilde{t} = tD_m\sqrt{\dfrac{D_m}{D_h}} \\ \tilde{h} = h\sqrt{\dfrac{D_m}{D_h}} \\ \tilde{\nu} = \dfrac{\nu}{\sqrt{D_mD_h}} \\ \tilde{\lambda} = \dfrac{\lambda}{D_m} \\ \tilde{\chi} = \dfrac{\chi}{D_m} \end{cases} \quad \text{and} \quad \begin{cases} \tilde{K} = \dfrac{K}{\sqrt{D_mD_h}} \\ \tilde{r} = r\dfrac{D_h}{D_m} \\ \tilde{u} = u\dfrac{D_h}{D_m} \\ \tilde{a} = \dfrac{a}{D_m} \\ \tilde{b} = \dfrac{b}{D_m} \\ \tilde{c} = \dfrac{c}{D_m}\sqrt{\dfrac{D_h}{D_m}} \end{cases} \tag{6}$$

yields a set of two dimensionless equations which are identical in forms to Eqs. (1) and (3), except that the noise amplitudes D_h and D_m in the dimensionless forms of Eqs. (2) and (4) are 2 (instead of $2D_m$ or $2D_h$). We then discretize these equations with a fully implicit finite-difference method and solve the resulting set of nonlinear equations using the Newton-Raphson and biconjugate-gradient methods. The coupled equations were integrated up to the time $t=1.5\times 10^5$ (with a dimensionless time step, $\Delta t=0.2$). The Gaussian noises η_h and η_m were generated using the Box-Muller transformation [8]. Most of the simulations were carried out for a grid size $L=1024$, and we made at least 100 realizations for each case that was studied and averaged the results over all the realizations.

III. RESULTS AND DISCUSSION

Since the simulations involve solving a set of two highly nonlinear equations, we first tested the accuracy of our integration method by deleting the coupling terms and solving the KPZ and TDGL equations separately, using a grid size $L=10\,240$, in order to reproduce the known results for these two equations. Figure 1 presents the logarithmic plot of the width of the growing surface, defined by

$$w(L,t) = (\langle h^2\rangle - \langle h\rangle^2)^{1/2}, \tag{7}$$

in the KPZ model. One expects to have,

$$w(L,t) \sim t^{\beta}, \tag{8}$$

with the exact value, $\beta=1/3$, and Fig. 1 yields $\beta \simeq 0.331$. We also confirmed that our numerical integration of the KPZ equation does reproduce the roughness exponent $\alpha=1/2$, hence yielding the exact dynamic exponent $z=3/2$ for this model.

Figure 2 presents the time dependence of the average domain size $L(t)$ in the TDGL model, both at short (the inset in the figure) and long times, for two values of the noise amplitude (or, equivalently, the temperature), $D_m=0$ and 1. For the noiseless TDGL model [9,10] one has,

$$L(t) \sim \ln t, \tag{9}$$

(hence, $z_m=\infty$) and, therefore, at long times, the average domain size changes very little with time. These are completely

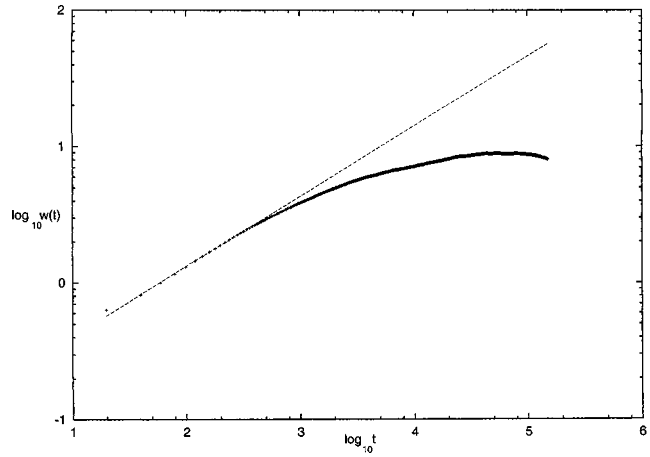

FIG. 1. Logarithmic plot of the surface width $w(t)$ vs time for the KPZ equation. The theoretical value of the dynamic exponent of the interface growth is $\beta=1/3$, while the results shown yield, $\beta \simeq 0.331$. Dashed line shows the function $w(t)=at^{\beta}$ that was fitted to the numerical results at short times.

consistent with the results shown in Fig. 2. In the noisy TDGL model, $L(t)$ grows logarithmically with t for times less than a crossover time t_c, but for $t \gg t_c$ it crosses over to a power-law regime [9,10],

$$L(t) \sim t^{1/2}, \tag{10}$$

and, hence, $z_m=2$, independent of the noise amplitude and values of the parameters K, r, and u in Eq. (3). However, t_c does depend on these parameters and can be quite large. These are also consistent with the results shown in Fig. 2 and, in particular, we obtain $z_m \simeq 2.01$, hence confirming the accuracy of our numerical integration. We remind the reader

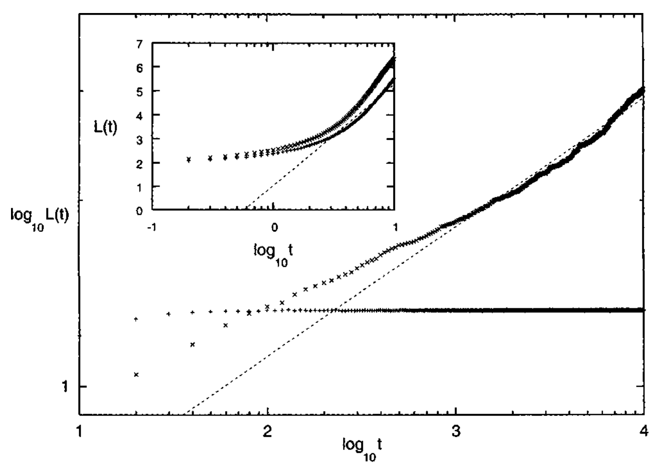

FIG. 2. Logarithmic plot of the average domain size $L(t)$ vs time, in the TDGL model, for the noise amplitutes $D_m=0(+)$ and $1(\times)$. For $D_m=0L(t)$ is constant, while for the noisy case $L(t)$ has, after a sufficiently long time, a power-law behavior, with the dashed line showing the fit of the results to $L(t)=at^b$. The inset shows the short-time results, indicating logarithmic growth of $L(t)$ for both cases, with the dashed line showing the fit of the results to $L(t)=a\ln(bt)$.

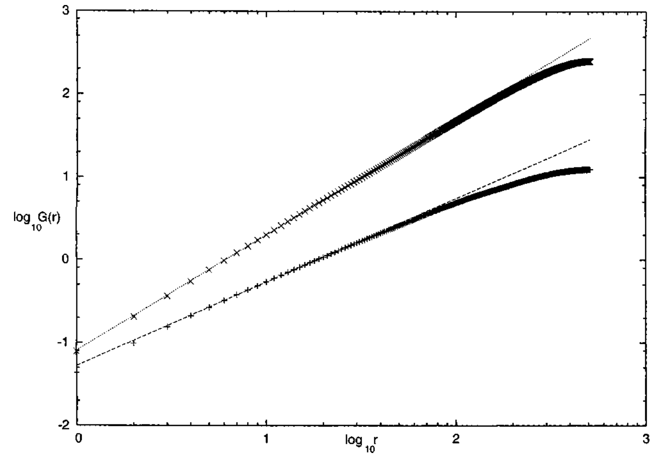

FIG. 3. Logarithmic plot of the height-height correlation function $G(r)$ vs r for $\chi=0(+)$ and $1(\times)$.

that for the 1D Glauber model one also has [11] $z_m=2$. We also compared these results with those obtained with a grid size $L=1024$, in order to confirm that the size effects are negligible, so that a grid size $L=1024$, which we use for the numerical integration of the coupled Eqs. (1) and (3), is large enough.

We now turn our attention to the set of the coupled equations, the dimensionless forms of Eqs. (1) and (3). The height-height correlation function is given by,

$$G(r,r') = \langle [h(r,t) - h(r',t')]^2 \rangle \sim |r-r'|^{2\alpha} g\left(\frac{|t-t'|}{|r-r'|^z} \right), \tag{11}$$

where α is the roughness exponent of the growing surface, z is the dynamic exponent for surface growth described by the KPZ equation, and $g(x)$ is a scaling function, so that for small r one must have, $G(r) \sim r^{2\alpha}$. Figure 3 presents the logarithmic plot of $G(r)$ versus r for two values of the coupling constant, $\chi=0$ and 1, from which we obtain, $\alpha \simeq 0.50$ and 0.70, respectively, indicating possible nonuniversal scaling behavior for the model. Note that while the correlation length ξ and the domain size L are generally the same for discrete systems, such as the Ising model, the same is not necessarily true for continuous models, such as what we consider here, as the correlation length ξ is the width of kink, or the domain wall. Therefore, in the composite films that are obtained by direct numerical integration of Eqs. (1) and (3), the roughness exponent α cannot be defined for short times and length scales up to the domain sizes, as the domains are small, typically less than ten grid points wide (i.e., the correlation length ξ is typically less than 10). However, after sufficiently long times, we find that the domain sizes saturate with $L(t) \gg \xi$ and, hence, the roughness exponent can be clearly defined and estimated.

To futher check the possible nonuniversal scaling behavior of the model, we also computed the dependence of the average domain size $L(t)$ on the time t, for $a=0$, 0.5, and 1 (together with $b=c=0$, $\lambda=5$, $\nu=u=K=1$, and $r=1.6$). Figure 4 presents the results, both for time less than a crossover time

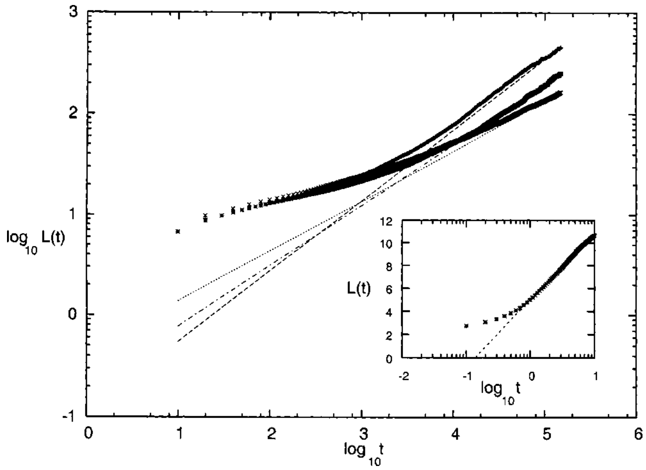

FIG. 4. Logarithmic plot of the average domain size $L(t)$ vs time t. At early times (the inset), the domains grow logarithmically with t for both $a=0(+)$ and $a=1(\times)$, with the dashed line showing the fit of the numerical results to $L(t)=a\ln(bx)$. For long times $L(t)$ grows as t^{1/z_m}, but z_m is not universal.

t_c (the inset in the figure) as well as for $t \gg t_c$. For $t < t_c$ the average domain size grows as $\ln t$; a fit of the numerical results to this functional form (the inset) confirms this. For $t \gg t_c$ there is a crossover to a power-law regime with a dynamic exponent z_m which, similar to the roughness exponent α, is nonuniversal; we obtain, $z_m \simeq 2.01$, 1.66, and 1.43 for $a=0$, 0.5, and 1, respectively. The crossover time t_c depends on the values of the various parameters of the model. The nonuniversal behavior of the model is presumably due to the nondecaying nature of the correlations that are built up as the composite film grows.

In a recent paper, Drossel and Kardar [12] carried out renormalization group analysis of the set of Eqs. (1) and (3) for dimensions $d \geqslant 4$, and also simulated their rule-based model in $(1+1)$ dimensions. They found that, for a range of the parameters of their rule-based model that was inaccessible to their perturbative analysis, the dynamic exponent z_m is nonuniversal and varies continuously with the model's parameters. Although the exact relation between the parameters in the rule-based model of Drossel and Kardar and those of Eqs. (1) and (3) is not clear, their result that z_m is nonuniversal is in agreement with our results. It would be most interesting to study the dependence of the crossover time t_c on the parameters of the model, as well as integrate Eqs. (1) and (3) in $(2+1)$ dimensions. Work in this direction is in progress.

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