Dynamical universality of the nonlinear conserved current equation for growing interfaces

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A nonlinear growth equation recently proposed as a model for molecular beam epitaxy, $\partial h/\partial t = -\nu_4\nabla^4 h + \lambda_{22}\nabla^2(\nabla h)^2 + \lambda_{13}\nabla(\nabla h)^3 + \eta$, where ν_4, λ_{22}, and λ_{13} are growth parameters and η is the deposition white noise, has been studied by a direct integration method. The standard deviation of the surface height for nonzero λ_{13} increases as $t^{1/4}$ in 1+1 dimensions, where t is time, which is consistent with the universality class of the Edwards- and Wilkinson-type [Proc. R. Soc. London Ser. A **381**, 17 (1982)] growth equation $\partial h/\partial t = \nu_2\nabla^2 h + \eta$, where ν_2 is the Edwards-Wilkinson growth parameter. This disagrees with the previous results obtained by dimensional scaling analysis. For $\lambda_{13} = 0$, our results are in agreement with known analytic results. Possible implications of our results for conserved growth are discussed.

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Kinetic surface roughening associated with the nonequilibrium growth of interfaces has been a subject of great current interest [1]. Recently, much attention has focused on the vacuum deposition growth of thin films, where a beam of particles is normally incident on a flat substrate, and the spatiotemporal random stochastic noise inherently present in the impinging flux causes the growing interface to roughen kinetically. Root mean square fluctuation W in the dynamical height of the growing surface (or, equivalently, W is the dynamical surface roughness of the thin film) is often found to exhibit generic scale invariance, obeying the dynamic scaling hypothesis [2],

$$W(t) \sim L^\alpha f(L/t^{1/z}), \tag{1}$$

where L is the lateral size of the substrate, t is the growth time, and the correlation length $\xi(t) \sim t^{1/z}$ denotes how lateral correlations spread over the substrate. In Eq. (1), α and z are the roughening and dynamical exponents, respectively, which define the universality class of the nonequilibrium growth process, and $f(y = L/t^{1/z})$ is the scaling function with the asymptotic properties $f(y \ll 1) \sim 1$ and $f(y \gg 1) \sim y^{-\alpha}$, so that $W(L \gg \xi(t)) \sim t^\beta$ and $W(L \ll \xi(t)) \sim L^\alpha$, where $\beta = \alpha/z$ is the growth exponent. Determining the critical exponent α and β (as well as $z = \alpha/\beta$) for various surface growth mechanisms has been extensively pursued in the recent literature using theory, experiment, and computer simulation [1–18].

An important class [3, 4] of growth processes, molecular beam epitaxy (MBE) being a famous and well-studied example, is conservative in nature (at least, in an idealized sense), meaning that the growth process conserves the total mass and volume of the growing film (after deposition), which immediately implies that evaporation and formation of overhangs and voids must be negligibly small in these types of surface growth. Such conserved growth processes obey a current conservation equation for the dynamical height fluctuation $h(\mathbf{x}, t)$ given by

$$\frac{\partial h}{\partial t} = -\boldsymbol{\nabla}\cdot\mathbf{j} + \eta, \tag{2}$$

where $\mathbf{j}$ is the particle current on the surface, $\boldsymbol{\nabla}$ is the divergence operator along the surface, and η is the nonconserved stochastic white random noise associated with the incident particle flux. [Note that the well-studied Kardar-Parisi-Zhang (KPZ) equation [15] cannot be written in the form of Eq. (2) and is *not* a conserved growth equation.] Under the most general conditions consistent with the symmetry of the problem (i.e., translational invariance in the growth direction, rotational and translational invariance in the surface plane), the leading-order current conserving growth equation is [3]

$$\frac{\partial h(\mathbf{x},t)}{\partial t} = \nu_2\nabla^2 h - \nu_4\nabla^4 h + \lambda_{22}\nabla^2(\boldsymbol{\nabla}h)^2 + \lambda_{13}\boldsymbol{\nabla}\cdot(\boldsymbol{\nabla}h)^3 + \eta(\mathbf{x},t), \tag{3}$$

where ν_2, ν_4, λ_{22}, λ_{13} are the coefficients of the various linear (ν_2, ν_4) and nonlinear (λ_{22}, λ_{13}) terms in the growth equation. When $\nu_2 \neq 0$, the other gradient terms in the growth equation are higher orders, and, therefore, irrelevant from a critical phenomena viewpoint. Thus, for $\nu_2 \neq 0$, the critical exponents (α, β, z) are entirely determined by the linear $\nu_2\nabla^2 h$ Laplacian term (the other terms, if present, may very well be quantitatively significant in producing complicated crossover behavior in real finite size, finite time experiments), which dominates the large distance ($L \to \infty$) and long time ($t \to \infty$) asymptotic behavior. In the presence of finite ν_2, therefore, at least for the asymptotic critical properties of the model, one could drop all the other gradient terms in Eq. (3), and the resulting simple linear growth equation ($\nu_2 \neq 0$, $\nu_4 = \lambda_{22} = \lambda_{13} = 0$) was introduced by Edwards and Wilkinson (EW) in the context of studying sedimentation [5]. A more nontrivial and interesting case

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is the situation with $\nu_2 = 0$ but ν_4, λ_{22}, $\lambda_{13} \neq 0$ in Eq. (3). Such a fourth-order conserved growth equation (and its various special cases such as λ_{22}, $\lambda_{13} = 0$) has been extensively studied in the last few years in the context of developing a theoretical conceptual framework for MBE growth [3,4,6–13,17]. In this paper we provide a direct numerical solution of Eq. (3) with $\nu_2 = 0$, finding the seemingly surprising (and unanticipated) result that the values of the critical exponents α and β (and $z = \alpha/\beta$) for the full fourth-order conserved growth equation (i.e., $\nu_4, \lambda_{22}, \lambda_{13} \neq 0; \nu_2 = 0$) are exactly the same as those of the second-order EW equation. Thus, the full nonlinear conserved MBE growth equation, i.e., Eq. (3) with $\nu_2 = 0$, belongs to the EW universality class, which is Eq. (3) with $\nu_2 \neq 0$ and $\nu_4 = \lambda_{22} = \lambda_{13} = 0$. The implications of our result, which we believe to be exact, are far reaching. In addition to explaining why a number of surface diffusion driven discrete growth models [12,13] seem to give EW exponents even though there is no obvious explicit source for producing a $\nu_2 \neq 0$ term, our results go a long way in providing an explanation for the important technological issue of why MBE growth on a flat singular substrate is capable of producing smooth thin films of very small kinetic roughness (EW growth in $d = 2 + 1$ has α, $\beta = 0$, implying smooth growth).

Before presenting our numerical results, we briefly discuss a context and a background for our model, namely, the conserved current continuum growth equation defined by Eq. (3), which was introduced for the surface growth problem by Lai and Das Sarma [3], who however, mostly concentrated, in their work, on the situation $\nu_2 = \lambda_{13} = 0$, a scenario also independently considered by Villain [4]. The case $\nu_2 = \lambda_{22} = \lambda_{13} = 0$, i.e., the fourth-order linear ($\nu_4 \neq 0$) growth equation, which is also the Mullins-Herring equilibrium surface diffusion equation, was independently invoked by Das Sarma and Tamborenea [7] and Wolf and Villain [11] in the context of kinetic surface roughening. Combining equations (2) and (3), the most general form for the nonequilibrium surface current $\mathbf{j}(\mathbf{x}, \mathbf{t})$ in the context of kinetic growth can be written in the leading order as

$$\mathbf{j}(\mathbf{x},t) = -(\boldsymbol{\nabla} h)[\nu_2 + \lambda_{13}(\nabla h)^2 + \cdots] + \boldsymbol{\nabla}[\nu_4 \nabla^2 h - \lambda_{22}(\boldsymbol{\nabla} h)^2 + \cdots]. \quad (4)$$

These two terms in the surface current, namely, a nonequilibrium gradient term proportional to the slope $\boldsymbol{\nabla} h$, and a term proportional to $\boldsymbol{\nabla}\mu_{\rm ne}$, where $\mu_{\rm ne} \sim \nu_4 \nabla^2 h - \lambda_{22}(\boldsymbol{\nabla} h)^2$ is a "nonequilibrium" chemical potential, have been separately justified in the context of MBE growth by Krug *et al.* [13] and Villain [4], respectively. Our approach in this paper, following the original work of Lai and Das Sarma [3], is somewhat formal in that we insist on keeping all the symmetry allowed conserving terms up to fourth order in the growth equation, which gives us Eq. (3). We point out that while $\nu_2 \neq 0$ is the most relevant term in Eq. (3), the λ_{13} nonlinear term is the most relevant term in the absence of ν_2 as a simple power counting would show [3].

In the rest of this paper, we put $\nu_2 = 0$ in Eq. (3) to get the fourth-order conserved continuum growth equation:

$$\frac{\partial h(\mathbf{x},t)}{\partial t} = -\nu_4 \nabla^4 h + \lambda_{22} \nabla^2 (\boldsymbol{\nabla} h)^2 + \lambda_{13} \boldsymbol{\nabla} \cdot (\boldsymbol{\nabla} h)^3 + \eta(\mathbf{x}, t). \quad (5)$$

We consider in this paper Eq. (5) as well as two of its limiting forms with $\lambda_{13} = 0$ and with $\lambda_{22} = 0$. Our reasons for considering the $\nu_2 = 0$ situation are several: (i) As emphasized before, the $\nu_2 \neq 0$ situation gives us the EW equation, which being linear is trivially solved to produce $z = 2$, $\alpha = (3 - d)/2$, where d denotes the total system dimensionality (substrate dimension plus the growth direction). We emphasize that in the presence of nonzero ν_2 all other terms in Eq. (3) are irrelevant and do not affect growth criticality. In this situation (i.e., for $\nu_2 \neq 0$) our results may help to clarify the crossover behavior. (ii) More importantly, for MBE growth without desorption on a flat substrate the ν_2 term has been argued on physical grounds to be vanishingly small [3, 4, 13] so that Eq. (5) may actually be the leading-order conserved current continuum growth equation in many experimentally relevant situations [10]. This, in fact, is our principal motivation for studying Eq. (5) in detail. Before presenting our numerical results, we mention that the linear fourth-order equation [i.e., Eq. (5) with $\nu_4 \neq 0$ but $\lambda_{22} = \lambda_{13} = 0$] is also trivially solved to give $z = 4$ and $\alpha = (5 - d)/2$.

Now we present our numerical results, which are based on a direct integration of Eq. (5) as well as the special limiting cases of $\lambda_{13} = 0$ and $\lambda_{22} = 0$. Our simulations are performed in $d = 1 + 1$ starting from a flat substrate and using periodic boundary conditions along the surface. The time t corresponds to the number of Monte Carlo steps in our integration. We have tested our algorithm by first integrating the linear fourth-order equation ($\lambda_{22} = \lambda_{13} = 0$), exactly obtaining the known result $\beta = 3/8$ in $d = 1 + 1$ dimensions. We carry out our integration on a discrete grid using a simple Euler integration method [14]. The noise η is taken to be a uniform

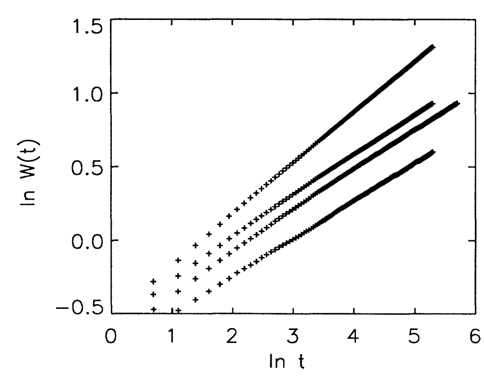

FIG. 1. Surface width W as a function of time in log-log plot of Eq. (5) for various values of λ_{13} and λ_{22} (with $\nu_4 = 1$). The curves are from the top to the bottom: $\lambda_{13} = 0$ and $\lambda_{22} = 1$ ($\beta \approx 0.34$), $\lambda_{13} = 1$ and $\lambda_{22} = 0$ ($\beta \approx 0.25$), $\lambda_{13} = 1$ and $\lambda_{22} = 1$ ($\beta \approx 0.25$), where -0.1 is added in ln W to avoid data overlap, and the bottom data is for $\lambda_{13} = 10$ and $\lambda_{22} = 0$ ($\beta \approx 0.25$) with uniform noise distribution and $\Delta t = 0.01$.

white noise or a Gaussian white noise with no difference in the results. We have tried different integration time steps (e.g., $\Delta t = 0.01,\ 0.001$), obtaining the same critical exponents.

We show in Fig. 1 our calculated dynamical surface width W ($\equiv$ root mean square deviation in the surface height fluctuations) as a function of time t for $L = 5\times10^4$, averaging over 50 independent runs. We show results for four different possibilities (from top to bottom in Fig. 1) in Eq. (5): $\lambda_{13} = 0$, $\lambda_{22} = 1$; $\lambda_{13} = 1$, $\lambda_{22} = 0$; $\lambda_{13} = \lambda_{22} = 1$; and, finally, $\lambda_{13} = 10$, $\lambda_{22} = 0$. All the curves use $\nu_4 = 1$. In Fig. 2, we show the saturated surface width $W(L, t \to \infty)$ as a function of the substrate size L for the limiting cases $\lambda_{13} = 0$, $\lambda_{22} = 1$ and $\lambda_{13} = 1$, $\lambda_{22} = 0$. We have carried out calculations for other values of the parameters ν_4, λ_{13}, λ_{22} obtaining very consistent results similar to the ones shown in Figs. 1 and 2. From these log-log plots, we conclude that the critical exponents α, β, and z in $d = 1+1$ dimensions for the various nonlinearities in Eq. (5) are given by

$$\begin{aligned} &\nu_4,\ \lambda_{22} \neq 0;\ \lambda_{13} = 0 : \beta = 0.34 \pm 0.01; \alpha = 1.02 \pm 0.03;\ z \approx 3.0; \\ &\lambda_{22} = 0;\ \nu_4,\ \lambda_{13} \neq 0 : \beta = 0.25 \pm 0.01; \alpha = 0.50 \pm 0.02;\ z \approx 2.0; \\ &\nu_4,\ \lambda_{22}, \lambda_{13} \neq 0 : \beta = 0.26 \pm 0.01; \\ &\nu_4,\ \lambda_{22} = 0;\ \lambda_{13} \neq 0 : \beta = 0.24 \pm 0.01. \end{aligned} \tag{6}$$

In Fig. 3, we show some typical saturated surface growth morphologies for these continuum growth models. We first discuss the $\lambda_{22}\nabla^2(\boldsymbol{\nabla} h)^2$ nonlinearity [which we note is the conserved KPZ nonlinearity [15,16]], which has earlier been analytically studied using the dynamical renormalization group (DRG) technique, yielding [3]

$$z = (7+d)/3;\ \alpha = (5-d)/3. \tag{7}$$

Our numerical results for $\lambda_{22} \neq 0$, $\lambda_{13} = 0$ agree with these DRG predictions, which have been argued to be exact [17] even though the original calculation was a single loop calculation [3]. Our numerical results showing the exactness of the DRG exponents for $\lambda_{13} = 0$ are significant because the issue has been somewhat controversial [8].

While our results for the λ_{22} nonlinearity are reassuring in the sense that they verify the exactness of existing perturbative DRG analytic results, our results for $\lambda_{13} \neq 0$ are surprising to say the least because our calculated exponents agree with the known EW exponents $z = 2$, $\alpha = 0.25$ in $d = 1+1$, leading to the conclusion that the $\boldsymbol{\nabla} \cdot (\boldsymbol{\nabla} h)^3$ nonlinearity, by itself, is sufficient to generate the linear EW term $\nabla^2 h$ upon renormalization (even though $\nu_2 = 0$ to start with), which then dominates the critical properties of the system, driving the model into the EW universality class. Since $\boldsymbol{\nabla} \cdot (\boldsymbol{\nabla} h)^3$

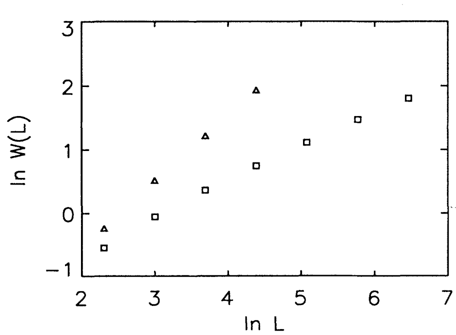

FIG. 2. Saturation surface width W as a function of substrate size L in log-log plot for (triangles) $\lambda_{13} = 0$ and $\lambda_{22} = 1$ ($\alpha \approx 1.0$), and (squares) $\lambda_{13} = 1$ and $\lambda_{22} = 0$ ($\alpha \approx 0.50$), both with $\nu_2 = 0$ and $\nu_4 = 1$.

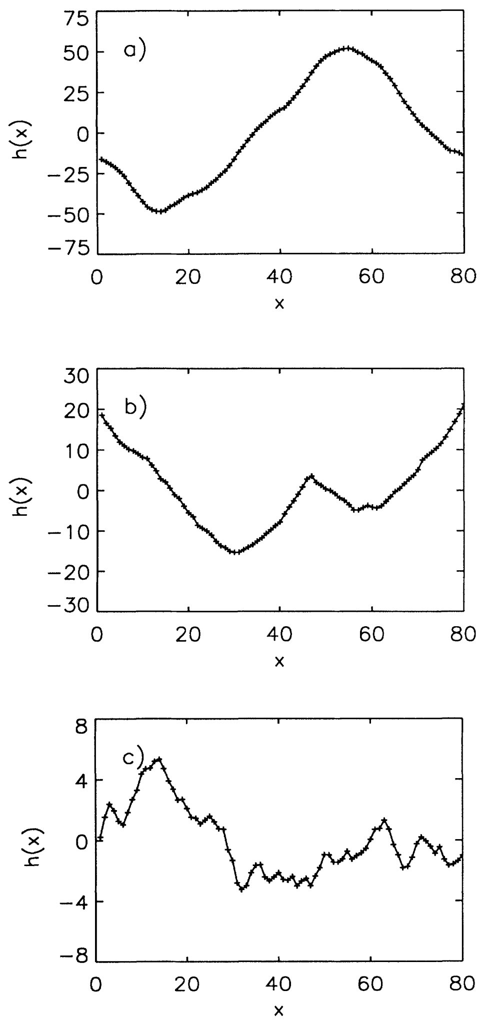

FIG. 3. Typical surface configurations in the saturation regime for various values of λ_{13} and λ_{22} ($\nu_2 = 0$ and $\nu_4 = 1$): (a) $\lambda_{13} = \lambda_{22} = 0$; (b) $\lambda_{13} = 0$ and $\lambda_{22} = 1$; and (c) $\lambda_{13} = 1$ and $\lambda_{22} = 0$. In curve (a) the curvature remains small and the height configuration is smooth. In (b) $h \to -h$ symmetry is broken so there is a sharp peak in contrast to the smooth valley. In (c) the surface width remains small.

nonlinearity is the most relevant term in Eq. (5), we conclude that both the conserved growth equations, Eqs. (3) and (5), belong to EW universality, independent of whether ν_2 is finite or zero. In discussing this result, we note that dimensional scaling analysis gives the following results for the $\nabla \cdot (\nabla h)^3$ nonlinearity [3]:

$$z = \frac{3+d}{2}, \qquad \alpha = \frac{5-d}{4}, \tag{8}$$

leading to $\alpha = 3/4$ and $\beta = 3/10$ in $d = 1+1$. We mention that an analysis of higher-order diagrams for the $\nabla^2(\nabla h)^2$ nonlinearity explicitly shows that vertex corrections vanish and the interaction remains unrenormalized in contrast to the $\nabla \cdot (\nabla h)^3$ nonlinearity, where the interaction term λ_{13} is clearly affected by vertex corrections. We emphasize that such a failure of a dimensional scaling analysis is quite uncommon — for example, a scaling analysis of the $\nabla^2(\nabla h)^2$ nonlinearity yields the correct critical exponents in contrast to our finding for the $\nabla \cdot (\nabla h)^3$ nonlinearity. Because Eq. (5) contains all possible leading-order symmetry-allowed conserved growth terms (except for the EW term $\nu_2 \nabla^2 h$), we have to conclude that MBE growth, in the absence of void formation, may belong (asymptotically) to EW universality either because of the explicit presence of the ν_2 term [as in Eq. (3)] or because of the renormalization of the λ_{13} term [as in Eq. (5)], except for the extremely unlikely scenario that both $\nu_2 = \lambda_{13} = 0$, whence we have the universality given by Eq. (7).

While we do not have a formal theoretical proof for the renormalization of the $\lambda_{13}\nabla \cdot (\nabla h)^3$ term into the $\nabla^2 h$ term, we have a persuasive argument based on the Hamiltonian $H_{13} \sim \frac{\lambda_{13}}{4} \int d^{d-1}x (\nabla h)^4$, which produces the $\lambda_{13}\nabla \cdot (\nabla h)^3$ term in the continuum growth equation via the Langevin equation approach. In the equilibrium situation, H_{13} will obviously generate [19] a lower-order $(\nabla h)^2$ term upon renormalization, leading to the EW $\nabla^2 h$ term in the growth equation. The situation is formally similar to the equilibrium restricted solid-on-solid model [18], which is well described by the continuum Hamiltonian $H_{\rm RSOS} \sim \int d^{d-1}x (\nabla h)^n$ with very large n, which, on renormalization, however, leads to the EW equation corresponding to $n = 2$. The presence of nonconserved noise in Eq. (5) should not pose a formal problem to this argument because the noise remains exactly unrenormalized in the conserved current model, thereby not affecting this argument. [The fact that the noise remains unrenormalized in the conserved growth model is supported by the agreement between our numerical exponents and DRG exponents for the $\nabla^2(\nabla h)^2$ nonlinearity.] This argument for the renormalization of the $\lambda_{13}\nabla \cdot (\nabla h)^3$ nonlinearity into a $\nu_2 \nabla^2 h$ linear term [20] is quite subtle, however because one is dealing with a nonequilibrium situation — our direct integration of Eq. (5) shows that the argument is valid in the nonequilibrium condition. We emphasize that a complete failure of the dimensional scaling analysis as we find here for the $\nabla \cdot (\nabla h)^3$ nonlinearity is an extremely unusual occurrence in surface growth phenomena. In this context, it is worthwhile to emphasize that both the EW $\nabla^2 h$ term and the nonlinear $\lambda_{13}\nabla \cdot (\nabla h)^3$ term in the growth equation can be derived from the general surface tension Hamiltonian $H \sim \int d^{d-1}x \sqrt{1 + (\nabla h)^2}$ as the first two growth terms in a series expansion of the square root within a dynamical Langevin equation approach [17]. We surmise that the universality class of all the higher order terms in this expansion is EW as well, for the same reason. Since EW universality produces very smooth growth ($\beta = \alpha = 0$) in $d = 2+1$, we speculate that conserved MBE growth is generically smooth independent of whether $\nu_2 = 0$ or not (as long as $\lambda_{13} \neq 0$).

Note added in proof. After our work was submitted for publication, Das Sarma and Kotlyar [21] carried out an analytical DRG calculation, obtaining agreement with both of our numerical conclusions, showing (i) the generation of the EW $\nabla^2 h$ term upon renormalization of the $\nabla \cdot (\nabla h)^3$ term in Eq. (3), and (ii) the exactness of the Lai–Das Sarma result [3] for the $\nabla^2 (\nabla h)^2$ term to all loop orders.

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