# Nonmonotonic roughness evolution in unstable growth

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The roughness of vapor-deposited thin films can display a nonmonotonic dependence on film thickness, if the smoothening of the small-scale features of the substrate dominates over growth-induced roughening in the early stage of evolution. We present a detailed analysis of this phenomenon in the framework of the continuum theory of unstable homoepitaxy. Using the spherical approximation of phase-ordering kinetics, the effect of nonlinearities and noise can be treated explicitly. The substrate roughness is characterized by the dimensionless parameter $Q = W_0/(k_0 a^2)$, where $W_0$ denotes the roughness amplitude, $k_0$ is the small-scale cutoff wave number of the roughness spectrum, and $a$ is the lattice constant. Depending on $Q$, the diffusion length $l_D$ and the Ehrlich-Schwoebel length $l_{ES}$, five regimes are identified in which the position of the roughness minimum is determined by different physical mechanisms. The analytic estimates are compared by numerical simulations of the full nonlinear evolution equation.

## I. INTRODUCTION

The morphology of thin-film surfaces has a decisive influence on many film properties. The control of growth-induced surface roughness is therefore a central concern in thin-film science and technology. Two types of roughening mechanisms have been extensively studied in recent years.[1,2] The term *kinetic roughening* is commonly used to refer to a *stochastic* mechanism, in which fluctuations in the deposition flux interact with thermal smoothening to generate a scale-invariant, rough morphology. This theoretically appealing but empirically rather elusive phenomenon[3] is often superseded by a second, *deterministic* mechanism, a *growth instability* associated with reduced interlayer transport and slope-dependent mass currents along the surface.[2,4,5] The hallmark of unstable growth is a morphology of more or less regular mounds with a clearly developed characteristic length scale. While in practice the distinction between the two types of roughening mechanisms may not always be so clear cut,[6] they are very different conceptually.

In addition to the growth-induced roughness, clearly also the roughness of the substrate affects the film morphology. Since the growing film covers up the small-scale details of the substrate modulations, the substrate contribution to the roughness is expected to decrease with increasing film thickness, while the growth-induced roughness component increases. Under suitable conditions this leads to the somewhat counterintuitive possibility of a *minimum* of the total surface roughness at a nonzero film thickness. This phenomenon has been observed in several growth experiments,[7–9] and a theoretical description has been worked out on the level of linear continuum theories of kinetic roughening[10] and unstable growth.[11]

In Ref. 11 a quantitative comparison with the experiments of Gyure *et al.*[9] was attempted, which indicated an important influence of nonlinearities. This motivated the present study, in which the nonlinear term in the growth equation is treated explicitly using the spherical approximation of phase-ordering kinetics.[12,13] We find that the interplay of instability, nonlinearity, and noise gives rise to a rather complex

behavior, in which the position of the roughness minimum can be determined by several distinct physical mechanisms. For a quick overview of the different regimes we refer the curious reader to Table I.

The paper is organized as follows. In the Sec. II we introduce the standard continuum equation for unstable homoepitaxial growth[13,14] and describe the strategy for its analytical solution. Section III is devoted to the roughness evolution in the absence of noise. We first recapitulate the linear analysis of Ref. 11, then provide a detailed analysis of the relevance of the nonlinearity and the nonlinear behavior, and finally discuss the influence of correlated initial roughness. The effects of noise are analyzed in Sec. IV. In Sec. V we compare the analytic estimates to a numerical evaluation of the spherical approximation, as well as to numerical simulations of the full nonlinear growth equation, finding good agreement in all cases. Finally, some conclusions are formulated in Sec. VI.

## II. THE CONTINUUM EQUATION

The evolution of a surface growing under typical molecular beam epitaxy (MBE) conditions is described by an equation of the form[4]

TABLE I. Analytical estimates of the position of the minimum depending on the substrate roughness parameter $Q = W_0/(a_\perp a_\parallel k_0)$. The other quantities appearing in the table are determined by the characteristic length scales $l_D$ and $l_{ES}$ of the growing surface. In particular, $y = l_{ES}/l_D$ and $\tau_l = 4\kappa/\alpha^2 \approx 16 F^{-1}(l_D/l_{ES})^2$. For definitions of $\tau_D$ and $\tau_{ES}$ see Eqs. (32) and (33).

| Value of $Q$ | Position of the minimum | Relevant effect |
|---|---|---|
| $Q \ll 1/2$ | $\tau_{RD} = (Q^4 y^2/8\pi)\tau_l$ | Conserved noise |
| $1/2 \ll Q \ll \sqrt{32\pi}/y$ | $\tau_{RS} = (Q^2 y^2/32\pi)\tau_l$ | Nonconserved noise |
| $\sqrt{32\pi}/y \ll Q \ll 8l_D/(a_\parallel y)$ | $0.18\tau_l$ | Linear deterministic |
| $8l_D/(a_\parallel y) \ll Q \ll 8l_D/(a_\parallel y^2)$ | $0.4(\tau_l\tau_D)^{1/2}$ | Nonlinear |
| $8l_D/(a_\parallel y^2) \ll Q$ | $0.5(\tau_l\tau_D\tau_{ES})^{1/3}$ | Nonlinear |

$$\partial_t H + \mathbf{\nabla} \cdot \mathbf{J} = a_\perp F + \eta, \tag{1}$$

where $H$ is the height, $\mathbf{J}$ is the surface current, $a_\perp$ is the monolayer thickness, $F$ is the average value of the deposition flux, and $\eta$ is a noise term describing fluctuations in the flux (shot noise) and in the diffusion of adatoms on the surface. The constant term $a_\perp F$ can be eliminated passing to the frame of reference $H = a_\perp F t + h$ moving with the average height. The noise has zero average and correlations

$$\langle \eta(\mathbf{x},t) \eta(\mathbf{x}',t') \rangle = (2\pi)^{-2} \delta(t-t')(R_S - R_D \nabla^2) \delta(\mathbf{x}-\mathbf{x}'), \tag{2}$$

with the amplitudes $R_S$ and $R_D$ representing the effect of shot and diffusion noise, respectively.

The surface current is the sum of two contributions

$$\mathbf{J} = \kappa \mathbf{\nabla}(\mathbf{\nabla}^2 h) + f[(\mathbf{\nabla} h)^2] \mathbf{\nabla} h. \tag{3}$$

The first tends to smoothen the surface and has the form of a capillarity term,[15] even though it may be dominated by non-equilibrium effects such as nucleation.[24,16] The second term models a growth-induced surface current, whose existence is often (but not necessarily[17–19]) caused by the presence of an Ehrlich-Schwoebel barrier for interlayer diffusion. We assume in-plane isotropy of the current (for a discussion of origins and consequences of anisotropy see Refs. 19 and 20). For the function $f(a)$ we use the form valid in the weak barrier limit[5]

$$f(a) = \frac{\alpha}{(1 + l_D \sqrt{a}/a_\perp)(1 + l_{ES} \sqrt{a}/a_\perp)}, \tag{4}$$

where $l_D$ is the diffusion length and $l_{ES}$ the Ehrlich-Schwoebel length. These length scales are related to the in-layer hopping rate $D$, the interlayer hopping rate $D'$, and the deposition flux $F$ through

$$l_D \approx (D/F)^\gamma a_\parallel, \quad l_{ES} = (D/D' - 1) a_\parallel. \tag{5}$$

Here $a_\parallel$ denotes the in-layer lattice constant and the exponent $\gamma$ depends on the size of the critical cluster for two-dimensional nucleation.[21] Conditions of weak and strong step edge barriers can be distinguished according to whether $l_{ES} \gg l_D$ (strong barriers) or $l_{ES} \ll l_D$ (weak barriers).[2,5,16] Here we focus on the latter case, in which a continuum description is most likely to be valid.

The coefficients $\alpha$ and $\kappa$ in Eqs. (3) and (4) are related to microscopic parameters by $\alpha \approx F l_{ES} l_D / 2$, $\kappa \approx F l_D^4$.[2,5,14,16] We will for simplicity assume that the equality sign holds in these formulas; however, numerical factors are not precisely known and the equalities should be intended instead only in order of magnitude. Also the amplitude of the noise terms is connected to microscopic parameters through $R_D = l_D^2 R_S = l_D^2 F a_\perp^2 a_\parallel^2$.

By inserting the expression (3) of the current in the equation for the height and neglecting noise one obtains

$$\partial_t h = -\kappa(\mathbf{\nabla}^2)^2 h - \mathbf{\nabla}[f((\mathbf{\nabla}h)^2)\mathbf{\nabla}h]. \tag{6}$$

This strongly nonlinear equation is reminiscent of the Cahn-Hilliard equation for phase-ordering in systems with conserved order parameter.[12] A widely used method for investigating this kind of nonlinear evolution is the large-$N$ limit, or

spherical approximation. In the present context it consists of replacing the argument of the nonlinear current $f$ with its average value $a(t) = \langle (\mathbf{\nabla} h)^2 \rangle$. In this way, Eq. (6) is effectively linearized. It is then possible to write down a closed form linear equation for the structure factor $S(k,t) = \langle \hat{h}(\mathbf{k},t) \hat{h}(-\mathbf{k},t) \rangle$

$$\partial_t S(k,t) = -2[\kappa k^4 - f[a(t)]k^2]S(k,t), \tag{7}$$

where $a(t)$ must be determined self-consistently

$$a(t) = 2\pi \int_0^{k_0} dk \, k^3 S(k,t). \tag{8}$$

The solution of this pair of coupled equations has already been derived for long times by Rost and Krug.[13] Here we concentrate on the short-time behavior, i.e., all what happens before the instability sets in. In this time range we expect the large-$N$ approximation to give a fairly accurate description of the nonlinear behavior,[22] since correlations are still small in range and amplitude. In particular we will be interested in the time evolution of the surface roughness

$$W^2(t) = 2\pi \int_0^{k_0} dk \, k S(k,t). \tag{9}$$

We usually assume as initial condition $S_0(k) = S(k=0,t)$ a white spectrum with an upper cutoff $k_0 = \pi/l_0$

$$S_0(k) = \begin{cases} W_0^2/(\pi k_0^2) & \text{for } k < k_0 \\ 0 & \text{for } k > k_0, \end{cases} \tag{10}$$

which implies $W^2(0) = W_0^2$ and $a(0) = W_0^2 k_0^2 / 2$. The dimensionless number

$$Q = W_0/(k_0 a_\perp a_\parallel) \tag{11}$$

will turn out to provide a useful measure for the strength of the initial roughness; note that it involves both the amplitude ($W_0$) and the small-scale cutoff. Other types of initial roughness spectra will be treated in Sec. III C.

## III. SOLUTION IN THE DETERMINISTIC CASE

Equation (7) can be formally integrated

$$S_{det}(k,t) = 2\left(\frac{W_0}{k_0}\right)^2 \exp[-2\kappa k^4 t + 2k^2 b(t)], \tag{12}$$

where

$$b(t) = \int_0^t ds \, f[a(s)]. \tag{13}$$

By defining $k_m^2(t) = b(t)/(2\kappa t)$ one can rewrite

$$W^2(t) = 2\left(\frac{W_0}{k_0}\right)^2 \int_0^{k_0} dk \, k \exp\left\{2t\kappa k^4\left[2\left(\frac{k_m}{k}\right)^2 - 1\right]\right\} \tag{14}$$

$$= W_0^2 \sqrt{\frac{\pi}{2}} \exp(2\kappa k_m^4 t)$$

$$\times \frac{\{\mathrm{erf}(k_m^2 \sqrt{2\kappa t}) + \mathrm{erf}[(k_0^2 - k_m^2)\sqrt{2\kappa t}]\}}{2\sqrt{\kappa t} k_0^2}, \quad (15)$$

with $\mathrm{erf}(s) = (2/\sqrt{\pi}) \int_0^s \exp(-t^2) dt$. The wave number $k_m$ is the position of the structure function peak (when it is a real number, otherwise the peak is for $k = 0$). Within linear theory its value is constant, $k_m = k_l \equiv \sqrt{\alpha/2\kappa}$, while in general $k_m$ is a function of time.

The derivative of $W^2$ with respect to $t$ at $t = 0$ is

$$\left. \frac{dW^2(t)}{dt} \right|_{t=0} = \frac{2}{3} W_0^2 \kappa k_0^2 (3\tilde{k}^2 - k_0^2), \quad (16)$$

where $\tilde{k} \equiv k_m(0) = \sqrt{f[a(0)]/2\kappa} = \sqrt{f(W_0^2 k_0^2/2)/2\kappa}$. Hence if $\tilde{k}/k_0 > 1/\sqrt{3}$ the roughness grows from the beginning and there is no minimum in the behavior of $W^2(t)$: The instability is immediately at work. Notice, however, that the condition for the existence of the minimum involves $\tilde{k}$ and not $k_l$. When the initial roughness is large $\tilde{k} \ll k_l$ and it may occur that $\tilde{k} \ll k_0 \ll k_l$: In such a case a minimum occurs even if the linear theory does not predict it. This fact, together with the observation that $k_l/k_0$ does not depend on $W_0$, implies that if $k_0$ is sufficiently large a minimum exists even for very small amplitude of the initial fluctuations. If $k_0$ is not large, only a strong initial roughness can originate a nonmonotonic behavior of the width.

We will assume in the following that a minimum exists. To study the detailed behavior of the system one should in principle consider Eqs. (12) and (13) simultaneously. However, expanding the expression of $W^2(t)$ for small $k_m/k_0$

$$W^2(t) = \left(\frac{W_0}{k_0}\right)^2 \sqrt{\frac{\pi}{2}} \frac{\mathrm{erf}(k_0^2 \sqrt{2\kappa t})}{2\sqrt{\kappa t}} + W_0^2[1 - \exp(-2\kappa k_0^4 t)]$$

$$\times \left(\frac{k_m}{k_0}\right)^2 + O\left(\frac{k_m}{k_0}\right)^4, \quad (17)$$

one finds that after a transient time $t_0 = 1/(2\kappa k_0^4)$ the width starts decreasing as $1/\sqrt{\kappa t}$ and it does so until $1/\sqrt{\kappa t} \simeq k_m^2$. During this time interval the width decreases in time and depends only on $\kappa$ and $k_0$, not on $k_m$ (hence not on the form of the current, not even its linear expansion). The system is effectively described by an evolution equation (6) where only the relaxational term proportional to $\kappa$ matters.[10,23] This fact is crucial for all the following calculations: The structure factor is known

$$S_{det}(k,t) = S_0 \exp(-2\kappa k^4 t) \quad (18)$$

and can be used to compute $a(t)$ and $k_m(t)$, which change nonlinearly in time, but do not affect significantly $S_{det}(t)$. This situation persists up to a time such that other terms in the expansion (17) become large. Since the other terms grow with $t$, it is around this time that $W^2(t)$ reaches a minimum. The only role played by the current $f$ is to determine the time

evolution of $k_m(t)$ and hence when the decay of the initial fluctuations ends: But $f$ does not affect the way $W^2(t)$ decreases.

The form (18) of the structure factor may be interpreted as that of a system which is coarsening with a typical correlation length growing as $L(t) \sim (\kappa t)^{1/4}$. The initial condition creates ''domains'' of size $k_0^{-1}$, much smaller than the length of the instability $k_m^{-1}$: the system evolves by reducing the amplitude of fluctuations and increasing the correlation length. This explains why the initial decrease of $W^2$ is seen only for small $\tilde{k}/k_0$. This coarsening process continues until

$$L \simeq k_m^{-1} . \quad (19)$$

From this time on the evolution proceeds by amplifying fluctuations of scale $L$ and the instability sets in. Notice that while in the linear case $k_m = k_l$ is constant in time, when nonlinearities are taken into account $k_m$ grows in time but remains always smaller than $k_l$, because $f(a) \le \alpha$ for all $a$. Therefore the time where the minimum occurs in the linear theory is a lower bound for the same quantity in the nonlinear case.

We now compute in detail how the position of the minimum depends on the amplitude of the initial fluctuations. For reference it is useful to summarize first the results of the linear theory.[11]

### A. Linear theory

The assumption of linearity for the current implies $a(t) = 0$. Hence $f[a(t)] = f[0] = \alpha$, $b(t) = \alpha t$, and $k_m^2 = k_l^2 = \alpha/2\kappa$. Then the temporal evolution of the width is fully specified for all times by Eq. (15). Letting $k_0 \to \infty$ such a formula can be cast as

$$W^2(t) = \frac{W_0^2}{4} \left(\frac{k_l}{k_0}\right)^2 \Phi(t/\tau_l), \quad (20)$$

where

$$\tau_l = 4\kappa/\alpha^2 \quad (21)$$

is the inverse amplification rate of the maximally unstable fluctuations and the scaling function is

$$\Phi(x) = e^{2x} \sqrt{2\pi/x} [1 + \mathrm{erf}(\sqrt{2x})]. \quad (22)$$

This formula is valid only for times greater than $t_0 = 1/2\kappa k_0^4 \sim (k_l/k_0)^4 \tau_l \ll \tau_l$.

The width attains a minimum at a time

$$t_{min}^l \approx 0.18 \ \tau_l, \quad (23)$$

where it has been reduced by a factor

$$W^2(t_{min}^l)/W_0^2 \approx 3.42 \ (k_l/k_0)^2. \quad (24)$$

This minimum marks the transition between the initial power-law decrease and the eventual exponential increase due to the linear instability.

### B. Nonlinear theory

The initial condition implies that $a(0) = W_0^2 k_0^2/2$. Then, for very small $t$

$$b(t) = f(W_0^2 k_0^2/2)t \equiv \tilde{\alpha} t. \tag{25}$$

By comparing $\tilde{\alpha}$ with $\alpha$ one recovers the condition (21) of Ref. 11 for the irrelevance of the nonlinearity

$$\frac{W_0 k_0 l_D}{a_\perp} \ll 1. \tag{26}$$

However, this condition turns out to be too restrictive. The reason is that it assesses the relevance of the nonlinearity from its importance in the expression of the unstable current at the initial time. But, as discussed above, $f$ does not play any role in the initial evolution of the width. The relevance of the nonlinearity must instead be established from its influence on the position of the minimum of $W^2(t)$, i.e., for long times. By that time the initial fluctuations have already been reduced significantly.

Expanding $a(t)$ as a function of $k_m/k_0$

$$a(t) = \frac{W_0^2}{4\kappa k_0^2 t}[1 - \exp(-2\kappa k_0^4 t)] \tag{27}$$

$$+ \frac{W_0^2}{4}\left[ -4\exp(-2\kappa k_0^4 t)k_0^2 + \sqrt{\frac{2\pi}{\kappa t}}\mathrm{erf}(k_0^2\sqrt{2\kappa t}) \right]$$

$$\times \left(\frac{k_m}{k_0}\right)^2 + O\left(\frac{k_m}{k_0}\right)^4, \tag{28}$$

one can see that $a(t)$ is constant only for times of the order of $t_0 = 1/(2\kappa k_0^4)$, indicating that Eq. (25) soon loses validity.

For longer times (up to $t \simeq 1/[\kappa k_m(t)^4]$),

$$a(t) = \frac{W_0^2}{4\kappa k_0^2 t}. \tag{29}$$

Inserting Eq. (29) in the expression (13) we can compute the long-time behavior of $b(t)$

$$b(t) = \int_0^t ds \frac{\alpha}{\left(1 + \frac{l_D W_0}{2a_\perp k_0 \sqrt{\kappa s}}\right)\left(1 + \frac{l_{ES} W_0}{2a_\perp k_0 \sqrt{\kappa s}}\right)} \tag{30}$$

$$= \int_0^t ds \frac{\alpha}{(1 + \sqrt{\tau_D/s})(1 + \sqrt{\tau_{ES}/s})}, \tag{31}$$

where we have introduced two time scales

$$\tau_D = \left(\frac{l_D W_0 k_l^2}{2a_\perp k_0}\right)^2 \tau_l = Q^2 y^2 \left(\frac{a_\parallel}{8l_D}\right)^2 \tau_l \tag{32}$$

and

$$\tau_{ES} = \left(\frac{l_{ES} W_0 k_l^2}{2a_\perp k_0}\right)^2 \tau_l = Q^2 y^4 \left(\frac{a_\parallel}{8l_D}\right)^2 \tau_l. \tag{33}$$

In the right equalities we have introduced the quantity $y = l_{ES}/l_D$ and $Q$ is defined in Eq. (11). It is important to stress that since we are considering weak Ehrlich-Schwoebel barriers $y \ll 1$ and hence $\tau_{ES} \ll \tau_D$. Moreover, realistic values of the diffusion length are such that $l_D/a_\parallel \gg 1$. Depending on the initial roughness via the parameter $Q$, the time scales $\tau_{ES}$

and $\tau_D$ will be larger or smaller than the initial one, $\tau_l$, giving rise to different scenarios.

### 1. Irrelevant nonlinearity (intermediate initial roughness)

Consider first the case of a fairly small initial roughness, i.e., $Qya_\parallel/8l_D \ll 1$, so that $\tau_{ES} \ll \tau_D \ll \tau_l$. Then for $t_0 \ll t \ll \tau_{ES}$ one can neglect the constant term in the denominator of Eq. (31)

$$b(t) \approx \int_0^t ds \frac{\alpha s}{\sqrt{\tau_D \tau_{ES}}} = \frac{\alpha t^2}{2\sqrt{\tau_D \tau_{ES}}}. \tag{34}$$

For $\tau_{ES} \ll t \ll \tau_D$ instead

$$b(t) \approx \int_0^t ds \frac{\alpha s^{1/2}}{\sqrt{\tau_D}} = \frac{2\alpha t^{3/2}}{3\sqrt{\tau_D}}, \tag{35}$$

while for $\tau_D \ll t \ll \tau_l$

$$b(t) \approx \alpha t. \tag{36}$$

$b(t)$ undergoes several changes during the time evolution passing through two intermediate behaviors. However, it is easy to see that these variations in the form of $b(t)$ are too short lived to affect the time evolution of $W^2$ [or $a(t)$]. The minimum width is reached when the effect of the nonlinearity is already lost, and is well described by linear theory. The condition for the irrelevance of the nonlinearity is therefore that $\tau_D \ll \tau_l$, that is

$$\frac{W_0 k_0 l_D}{2a_\perp}\left(\frac{k_l}{k_0}\right)^2 \ll 1. \tag{37}$$

Comparing with Eq. (26) it is clear that the relevance of the nonlinearity is strongly reduced when $k_l \ll k_0$. Notice, moreover, that the role of $k_0$ is opposite compared to the condition (26): An initial substrate rough down to very small length scales (large $k_0$) makes the nonlinearity less relevant.

### 2. Relevant nonlinearity (large initial roughness)

Let us assume instead $\tau_{ES} \ll \tau_l \ll \tau_D$, i.e., $8l_D/(ya_\parallel) \ll Q \ll 8l_D/(y^2 a_\parallel)$. One has for $\tau_{ES} \ll t \ll \tau_D$

$$b(t) = \frac{2\alpha}{3\sqrt{\tau_D}}t^{3/2} \ll \alpha t. \tag{38}$$

With this expression of $b(t)$ the value of $k_m$ is much smaller than the linear value $k_l$ and the condition (19) for the minimum is attained on time scales larger than $\tau_l$. To estimate $t_{min}$ more precisely the form of $W^2(t)$ can be written in the scaling form

$$W^2(t) = \frac{W_0^2}{4}\left(\frac{k_m(t)}{k_0}\right)^2 \Phi[t\kappa k_m(t)^4], \tag{39}$$

where $\Phi$ is defined in Eq. (22). Now the minimum value of $W^2$ is not reached where $\Phi'(x) = 0$, because $k_m$ depends on $t$. The condition for the minimum of $W^2$ is instead

$$x\Phi'(x)(2b't - b) + \Phi(x)(b't - b) = 0. \tag{40}$$

Using the expression (38) for $b(t)$ one gets

$$\frac{x\Phi'(x)}{\Phi(x)} = -\frac{1}{4}, \tag{41}$$

whose solution is $x_{min} = t^D_{min}/\tau(t^D_{min}) \approx 0.06$, yielding

$$t^D_{min} \approx 0.4(\tau_l \tau_D)^{1/2} = 0.4 \ Qy\left(\frac{a_\parallel}{8l_D}\right)\tau_l. \tag{42}$$

Hence $t^D_{min}$ is larger than $\tau_l$ but smaller than $\tau_D$. The value of the width at the minimum is

$$W(t^D_{min})/W_0 \approx 1.3\left(\frac{k_l}{k_0}\right)\left(\frac{\tau_l}{\tau_D}\right)^{1/2}. \tag{43}$$

If instead $Q \gg 8l_D/(y^2 a_\parallel)$, then $\tau_l \ll \tau_{ES} \ll \tau_D$. By means of an analogous procedure, one finds that the minimum occurs for $x_{min} \approx 0.03$, yielding

$$t^{ES}_{min} \approx 0.5(\tau_l \tau_D \tau_{ES})^{1/3} = 0.5 \ Q^{4/3}y^2\left(\frac{a_\parallel}{8l_D}\right)^{4/3}\tau_l. \tag{44}$$

The value of the width at the minimum is

$$W(t^{ES}_{min})/W_0 \approx 1.1\left(\frac{k_l}{k_0}\right)\left(\frac{\tau_l^2}{\tau_D \tau_{ES}}\right)^{1/3}. \tag{45}$$

### C. Correlated initial conditions

The previous results can be easily extended to the case of a substrate with correlated roughness, i.e., with

$$S_0(k) = \begin{cases} A & \text{for } k < k^* \\ A(k^*/k)^\theta & \text{for } k^* < k < k_0 \\ 0 & \text{for } k > k_0, \end{cases} \tag{46}$$

with $\theta > 0$ and $k_0/k^* \gg 1$.

In this case the condition for an initial decrease of the roughness is, in the limit $k_0/k^* \to \infty$,

$$\left(\frac{\tilde{k}}{k_0}\right)^2 < \begin{cases} \dfrac{4-\theta}{2(6-\theta)} & \text{for } \theta < 4 \\ 0 & \text{for } \theta > 4. \end{cases} \tag{47}$$

Hence for $\theta > 4$ the width can only increase monotonically from the beginning.

#### 1. Linear theory

For $\theta < 2$ one can safely take $k^* \to 0$ and find, for $t \gg 1/(2\kappa k_0^4)$ and to second order in $k_l$,

$$W^2(t) = \pi A k^{*\theta}(2\kappa t)^{\theta/4}\left[\frac{1}{(8\kappa t)^{1/2}}\Gamma\left(\frac{2-\theta}{4}\right) + k_l^2\Gamma\left(1-\frac{\theta}{4}\right)\right]. \tag{48}$$

Estimating the position of the minimum from the time when the second term equals the first, one obtains

$$t^l_{min}(\theta) = \frac{1}{8\kappa k_l^4}\left[\frac{\Gamma\left(\dfrac{2-\theta}{4}\right)}{\Gamma\left(1-\dfrac{\theta}{4}\right)}\right]^2, \tag{49}$$

while the minimum width reached is

$$\frac{W^2[t^l_{min}(\theta)]}{W_0^2} = (2-\theta)2^{-(\theta/2+1)}\Gamma\left(\frac{2-\theta}{4}\right)^{\theta/2}$$

$$\times \Gamma\left(1-\frac{\theta}{4}\right)^{1-\theta/2}\left(\frac{k_l}{k_0}\right)^{2-\theta}. \tag{50}$$

Both $t^l_{min}(\theta)$ and $W^2[t^l_{min}(\theta)]$ are growing functions of $\theta$: The minimum is delayed and made shallower by the presence of correlations in the roughness. This occurs because the roughness is concentrated on large length scales and the damping of small scale fluctuations provided by the relaxational dynamics is less effective in the reduction of the surface width. Such effect is most evident when $\theta$ approaches 2: $t^l_{min}(\theta)$ diverges, while $W^2[t^l_{min}(\theta)]/W_0^2$ goes to 1, since for $\theta = 2$ all the roughness is concentrated on the macroscopic length scale $(k^*)^{-1}$.

For $2 < \theta < 4$, the origin of the minimum is different. In this case one can take $k_0 \to \infty$, and find for $t \ll 1/(2\kappa k^{*4})$ and small $k_l$

$$W^2(t) = W_0^2 + 2\pi A k^{*\theta/4}(2\kappa t)^{\theta/4}$$

$$\times\left[-\frac{1}{(\theta-2)(2\kappa t)^{1/2}} + \frac{2k_l^2}{4-\theta}\right]. \tag{51}$$

Differently from Eq. (48), here $t$ has, in the first contribution to the second term, a positive (and small) exponent but a negative prefactor. Therefore in this case the initial decrease of the roughness is much weaker and this is reflected by the minimum width, which is close to $W_0$. The time when the minimum is reached is

$$t^l_{min}(\theta) = \frac{1}{8\kappa k_l^4}\left(\frac{4-\theta}{\theta-2}\right)^2 \tag{52}$$

and vanishes, as expected, in the limit $\theta \to 4$.

#### 2. Nonlinear theory

With correlated initial conditions the evolution of the average square slope, considering $S(k,t) = S_0(k)\exp(-2\kappa k^4 t)$ and $k^* \to 0$, is for $t \gg 1/(2\kappa k_0^4)$

$$a(t) = \frac{\pi A k^{*\theta}\Gamma(1-\theta/4)}{2(2\kappa t)^{1-\theta/4}}. \tag{53}$$

Hence $a(t)$ is for all $\theta < 4$ a decreasing function of time but its rate of reduction vanishes as $\theta$ approaches 4. All the previous treatment of the nonlinearity can be repeated. The only difference in the results is that the time scales $\tau_D$ and $\tau_{ES}$ are modified. In particular

$$\tau_D(\theta) = \frac{1}{2K}\left[\frac{\pi A k^{*\theta}\Gamma(1-\theta/4)}{2}\frac{l_D^2}{a_\perp^2}\right]^{4/(4-\theta)}. \tag{54}$$

With this expression one can assess the relevance of the nonlinearity by comparison with the time scale of linear theory $t^l_{min}(\theta)$. For $\theta < 2$ one finds that the nonlinearity is irrelevant $[\tau_D(\theta) \ll t^l_{min}(\theta)]$ for
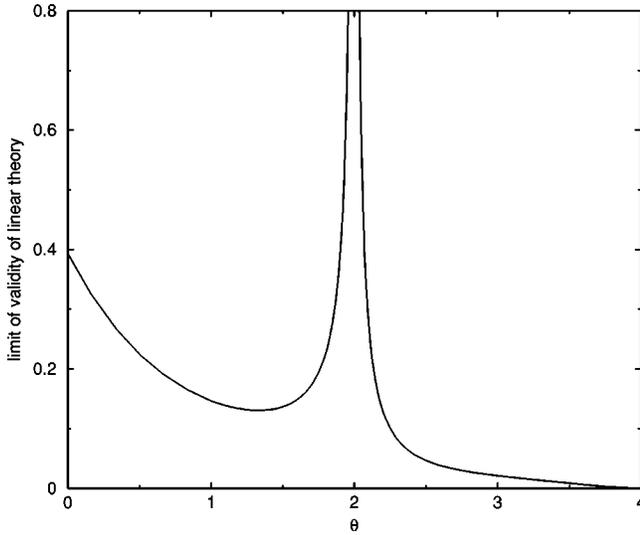
FIG. 1. Plot of the right hand side of Eqs. (55) and (56) vs $\theta$ for $k_l/k_0 = 1/10$ and $k^*/k_0 = 1/100$.

$$\frac{W_0^2 k_0^2 l_D^2}{4a_{\parallel}^2}\left(\frac{k_l}{k_0}\right)^4 \ll \left(\frac{k_l}{k_0}\right)^{\theta}\frac{1}{(2-\theta)\Gamma(1-\theta/4)}\left[1-\frac{\theta}{2}\left(\frac{k^*}{k_0}\right)^{2-\theta}\right]$$
$$\times\left\{\frac{\Gamma[(2-\theta)/4]}{2\Gamma(1-\theta/4)}\right\}^{(4-\theta)/2}. \qquad (55)$$

The right-hand side of the previous inequality is of the order of one for $\theta \to 0$, in agreement with Eq. (37). For small $\theta$ it decreases as $(k_l/k_0)^{\theta}$. For $\theta \to 2$ it diverges, as a consequence of the fact that $t_{min}^l(\theta)$ goes to infinity.

For $2 < \theta < 4$ the condition for the irrelevance of the nonlinearity becomes

$$\frac{W_0^2 k_0^2 l_D^2}{4a_{\parallel}^2}\left(\frac{k_l}{k_0}\right)^4 \ll \left(\frac{k_l}{k_0}\right)^{\theta}\frac{1}{(2-\theta)\Gamma(1-\theta/4)}\left[1-\frac{\theta}{2}\left(\frac{k^*}{k_0}\right)^{2-\theta}\right]$$
$$\times\left[\frac{4-\theta}{2(\theta-2)}\right]^{(4-\theta)/2}. \qquad (56)$$

The right-hand side diverges for $\theta \to 2$ and vanishes for $\theta \to 4$, as expected since $\tau_D(\theta)$ diverges in that limit: For $\theta \to 4$ the nonlinearity is always relevant.

A more immediate perception of the meaning of Eqs. (55) and (56) is given by Fig. 1, showing the right-hand sides of the inequalities as a function of $\theta$. Nonlinearity is irrelevant for values of $[W_0 k_l^2 l_D/(2a_{\parallel}^2 k_0)]^2$ smaller than the function plotted. Except for a small region around $\theta = 2$, where it diverges (because $t_{min}^l \to \infty$), the function is always smaller than its value for $\theta \to 0$.[25] This means that correlations increase the effect of the nonlinearity for almost all values of $\theta$.

## IV. SOLUTION IN THE NOISY CASE

So far we have neglected the presence of noise in Eq. (1). We now turn to the study of the problem in the presence both of deposition and diffusion noise. It will turn out that noise affects the position of the minimum only for small initial roughness. For large and intermediate values of $W_0$ the deterministic theory of Sec. III is sufficient.

The inclusion of noise in the problem changes Eq. (7) to

$$\partial_t S(k,t) = -2[\kappa k^4 - f[a(t)]k^2]S(k,t) + R(k), \qquad (57)$$

with $R(k) = R_S + R_D k^2$.

The formal solution is

$$S(k,t) = S_{det}(k,t) + S_{noise}(k,t), \qquad (58)$$

where $S_{det}(k,t)$ is again given by Eq. (12) while

$$S_{noise}(k,t) = R(k)S_{det}(k,t)\int_0^t ds\, S_{det}^{-1}(k,s). \qquad (59)$$

As before the key point is to realize that provided $k_m/k_0 \ll 1$, after a short transient of duration $t_0 = 1/(2\kappa k_0^4)$, one can safely take $S_{det}(k,t) \approx S_0\exp(-2\kappa k^4 t)$. Using this expression

$$S_{noise}(k,t) = \frac{R(k)}{2\kappa k^4}[1 - \exp(-2\kappa k^4 t)]. \qquad (60)$$

With this formula one can compute the additive contributions of the noisy part of the structure factor to $a(t)$ and to the roughness $W^2(t)$ which arise due to shot noise $(S)$ and diffusion noise $(D)$, respectively,

$$a(t) = a_{det}(t) + a_S(t) + a_D(t) \qquad (61)$$

and

$$W^2(t) = W_{det}^2(t) + W_S^2(t) + W_D^2(t). \qquad (62)$$

The results are

$$a_S(t) \approx \frac{\pi R_S}{4\kappa}\ln(2\kappa k_0^4 t), \qquad (63)$$

$$a_D(t) \approx \frac{\pi R_D k_0^2}{2\kappa}, \qquad (64)$$

and

$$W_S^2(t) \approx \pi R_S\sqrt{\frac{\pi t}{2\kappa}}, \qquad (65)$$

$$W_D^2(t) \approx \frac{\pi R_D}{4\kappa}\ln(2\kappa k_0^4 t). \qquad (66)$$

The determination of the temporal evolution of the system is now more complicated than in the noiseless case. There, the form of $a(t)$ was always the same and the minimum for $W^2$ changed depending on the various approximations for $f[a(t)]$. Here, even the expression of $a(t)$ varies in time. Moreover, in the noiseless case, the minimum in $W^2$ occurs when the condition (19) is fulfilled. Here, there are three independent contributions to the width: A minimum may appear much before the instability starts to play any role, simply because of the interplay between the different contributions to $W^2$. However, since $W_{det}^2$ is the only decreasing contribution, it is clear that a minimum in $W^2$ can occur only if it already existed in the deterministic case. Noise cannot create a minimum and, as will be shown below, cannot de-

stroy it, but can only shift it to shorter times. We will assume in the following that a minimum in the noiseless case exists.

The complication related to the different contributions to $a(t)$ turns out to be unimportant for physical values of the parameters. Inserting the expressions for $a_S$ and $a_D$ in Eq. (4) one sees that, under the physically sensible assumptions $l_D \gg a_\parallel$ and $k_0^{-1} \gg a_\parallel$, $f(a_S) \approx f(a_D) \approx \alpha$. Hence, even if for long times it may happen that $a_S$ or $a_D$ become larger than $a_{det}$, their value is so small that the theory is not affected.

The analysis of $W^2$ is instead quite involved. Let us define the time scales

$$\tau_{RS} = \frac{W_0^2}{2k_0^2 \pi R_S} = \frac{Q^2 y^2}{32\pi} \tau_l, \tag{67}$$

$$\tau_{RD} = \frac{2\kappa W_0^4}{\pi k_0^4 R_D^2} = \frac{Q^4 y^2}{8\pi} \tau_l, \tag{68}$$

$$\tau_{DS} = \left(\frac{R_D}{R_S}\right)^2 \frac{1}{8\pi\kappa} = \frac{y^2}{128\pi} \tau_l. \tag{69}$$

$\tau_{RS}$ is the time scale when $W_S$ becomes greater than $W_{det}$; $\tau_{RD}$ is the time scale when $W_D > W_{det}$; $\tau_{DS}$ is the time scale when $W_S > W_D$. Let us carry out the analysis of the interplay of these different time scales assuming, for the moment, that the deterministic part is well described by linear theory, i.e., $t_{min} = t_{min}^l \approx 0.18\tau_l$.

For small initial roughness ($Q \ll 1/2$) one finds that $\tau_{RD} \ll \tau_{RS} \ll \tau_{DS} \ll \tau_l$ and this implies that $W = W_{det}$ for $t \ll \tau_{RD}$, $W = W_D$ for $\tau_{RD} \ll t \ll \tau_{DS}$, $W = W_S$ for $\tau_{DS} \ll t \ll \tau_l$, and $W \approx \exp(t)$ for $\tau_l \ll t$. Hence the minimum occurs for $t_{min} \approx \tau_{RD}$ but before the instability sets in there is another change for $t \approx t_{DS}$.

If instead $Q \gg 1/2$ there are two possibilities. If the initial roughness is not very large ($Q^2 y^2 \ll 32\pi$), one has $\tau_{DS} \ll \tau_{RS} \ll \tau_l$ and $W = W_{det}$ for $t \ll \tau_{RS}$, $W = W_S$ for $\tau_{RS} \ll t \ll \tau_l$, and $W \approx \exp(t)$ for larger times. Otherwise, if $Q^2 y^2 \gg 32\pi$, then $\tau_{DS} \ll \tau_l \ll \tau_{RS} \ll \tau_{RD}$: $W = W_{det}$ always, the noise is irrelevant and $t_{min} = t_{min}^l \approx 0.18\tau_l$. Notice that the condition $Q^2 y^2 \gg 32\pi$ is, apart from the numerical factor, the condition (15) of the paper by Krug and Rost.[11]

We have so far assumed that $t_{min}^{det} = t_{min}^l$ of the same order of magnitude of $\tau_l$. When deterministic nonlinearities are strong they increase the value of $t_{min}^{det}$, which becomes much greater than $\tau_l$. As shown above, this starts to happen for $\tau_D \gg \tau_l$ which corresponds to $Q^2 y^2 \gg (8l_D/a_\parallel)^2 \gg 1$. Then it might in principle happen that $t_{min}^{det}$ becomes larger than $\tau_{RS}$ in the last of the cases above. However, while $t_{min}^{det}$ grows for large $Q$ as $Q$ [Eq. (42)] or as $Q^{4/3}$ [Eq. (44)], $\tau_{RS}$ is proportional to $Q^2$: $\tau_{RS}$ always remains larger than $t_{min}^{det}$ and nothing in the previous discussion changes.

In Table I is a summary of the different regimes found depending on $Q$.

## V. NUMERICAL RESULTS

The results presented above are obtained by analytically estimating the behavior of the large-$N$ equation (7), which in turn is an approximation of the fully nonlinear Eq. (6). In order to check the validity of these results we have solved
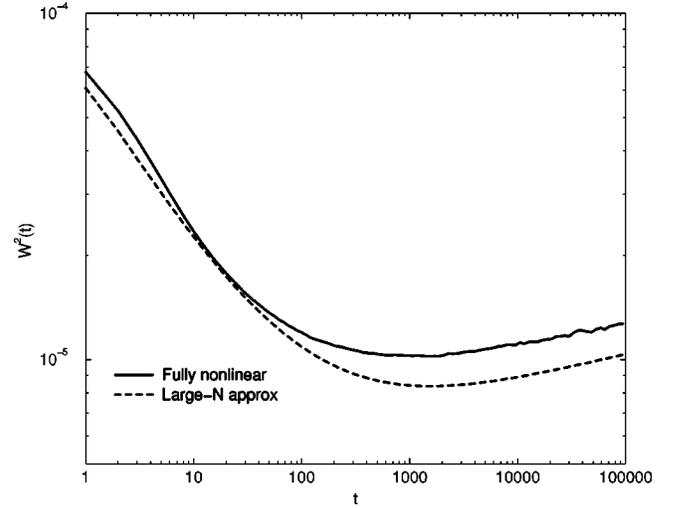


FIG. 2. Double logarithmic plot of $W^2(t)$ vs $t$ for a system with $k_0 = 1/10$, $W_0 = 1/100$, $Q = 1/10$, $F = 10^{-7}$, $l_D = 1000$, and $y = 1/100$. Here and in all other plots $a_\perp$ and $a_\parallel$ are taken to be equal to 1. For these values of the parameters $Q \ll 1/2$ and conserved noise dictates the position of the minimum.

numerically the fully nonlinear equation for values of the parameters corresponding to the different possible regimes of Table I and compared the results with the numerical integration of the large-$N$ equation[26] and with the analytical estimates. The numerical integration was performed by the simple first-order Euler scheme, on a lattice of size $512 \times 512$. The temporal stepsize was chosen to be 1, while the lattice spacing was equal to $\pi/k_0$, with $k_0$ specified in the figure captions.

We start by considering the limit of very small initial roughness, so that $Q = 0.1$ and the minimum is due to conserved noise roughening the surface (Fig. 2). The minimum width for the fully nonlinear solution occurs for a time compatible with the analytical prediction $t_{min} = \tau_{RD} \approx 1600$. Despite having a minimum around the same time the solution of the fully nonlinear case and the large-$N$ approximation differ noticeably for large times. This poor agreement is, however, only apparent and is due to a technical subtlety: The numerical solution of the full equation is performed on a square lattice, while the analytical calculations assume a circular Brillouin zone, $k < k_0$. When noise is irrelevant, since the structure factor decays exponentially for large wave vectors, the difference in the Brillouin zones does not really matter after the initial transient $t_0$. In the noise dominated cases instead, the structure factor has a power-law tail: The effect of the different Brillouin zones persists in time, cannot be eliminated easily,[27] and leads to a systematic overestimate of the value of $W^2(t)$. This is why for long times the numerical solution is not in agreement with the large-$N$ result. This problem is most evident in this case dominated by conserved noise, as the power-law tail of the $S(k,t)$ is broader.

In Fig. 3 the value of the roughness of the fully nonlinear case and of the large-$N$ approximation are plotted in the case where nonconserved noise dominates. Again, the analytical value $t_{min} = \tau_{RS} \approx 15\,700$ matches quite well the numerical results.

The same quantities are reported in Fig. 4 for values of the parameters such that both noise and nonlinearities are
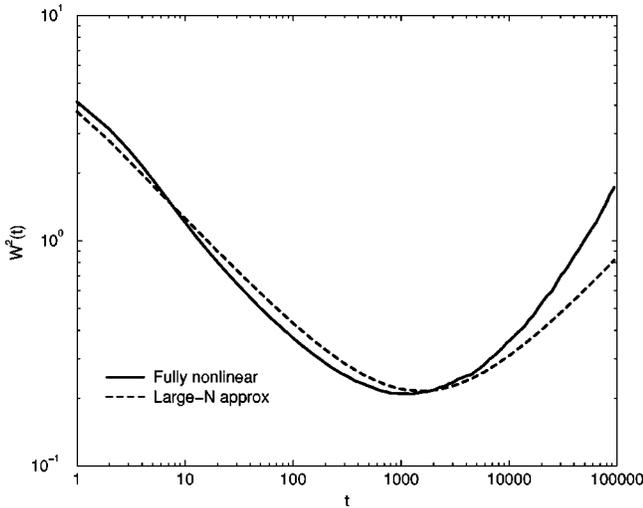
FIG. 3. Double logarithmic plot of $W^2(t)$ vs $t$ for a system with $k_0 = 1/40$, $W_0 = 10/4$, $Q = 100$, $F = 1$, $l_D = 40$, and $y = 1/100$. For these values of the parameters $1/2 \ll Q \ll \sqrt{32\pi}/y$: The minimum is due to nonconserved noise.



FIG. 5. Double logarithmic plot of $W^2(t)$ vs $t$ for a system with $k_0 = 1/100$, $W_0 = 50\,000$, $Q = 5 \times 10^6$, $F = 10^4$, $l_D = 10$, and $y = 1/1000$. The inset shows the log-log plot of $b(t)$ along with a line of slope 3/2. For these values of the parameters $8l_D/(a_\| y) \ll Q \ll 8l_D/(a_\| y^2)$ and the nonlinearity is relevant.

irrelevant. In this case, the analytical prediction for the position of the minimum is the one provided by linear theory $t_{min}^l \approx 0.18\tau_l \approx 14\,400$. Also in this case, the agreement between numerics and the theoretical prediction is good. Notice that for these values of the parameters the naive condition (26) for the irrelevance of nonlinearity is violated. However, the nonlinear contribution to the current (4) is initially large but rapidly decays, so that it does not influence the position of the minimum. The initial deviation from linearity can also be seen in the behavior of $b(t)$, plotted in the inset of Fig. 4.

The position of the minimum is instead determined by the nonlinearity in Fig. 5: Here linear theory would predict $t_{min}^l \approx 300$, while the roughness keeps decreasing until $t \approx 60\,000$ in reasonable agreement with the analytical estimate $t_{min} \approx 40\,000$ given by Eq. (42). The nonlinear behavior
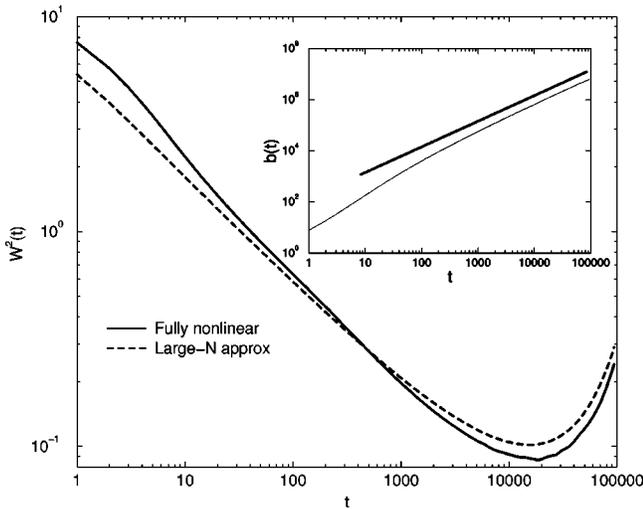
is also evident in the inset, where $b(t)$ is plotted: it is proportional to $t^{3/2}$ as predicted by Eq. (35).

Finally, in Fig. 6 the roughness in the most nonlinear case $[Q \gg 8l_D/(a_\| y^2)]$ is shown. In this case the linear theory would predict $t_{min} \approx 0.03$, while Eq. (44) yields $t_{min} \approx 5000$. The numerical result $t_{min} \approx 8000$ is again close to the estimate provided by nonlinear theory. Consistently $b(t)$ grows as $t^2$ (Fig. 6, inset).

In summary, for initial values of the roughness ranging from very small to very large we find that the evolution of the fully nonlinear equation is well approximated by the large-$N$ limit and that the analytical estimates found above agree with the numerical results. The large-$N$ limit describes quite precisely this early stage behavior because, up to the time when the minimum is reached, the dynamics makes the



FIG. 4. Double logarithmic plot of $W^2(t)$ vs $t$ for a system with $k_0 = 1/100$, $W_0 = 3$, $Q = 300$, $F = 1/50$, $l_D = 265.9$, and $y = 1/10$. The inset shows the log-log plot of $b(t)$ along with a line of slope 1. For these values of the parameters $\sqrt{32\pi}/y \ll Q \ll 8l_D/(a_\| y)$: linear noiseless theory holds.
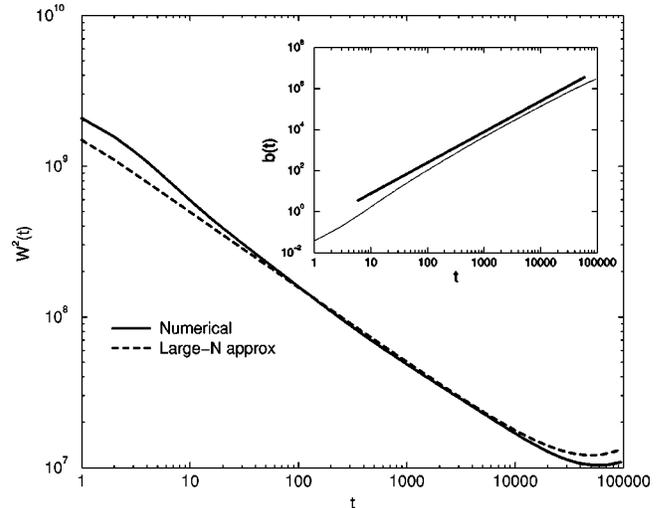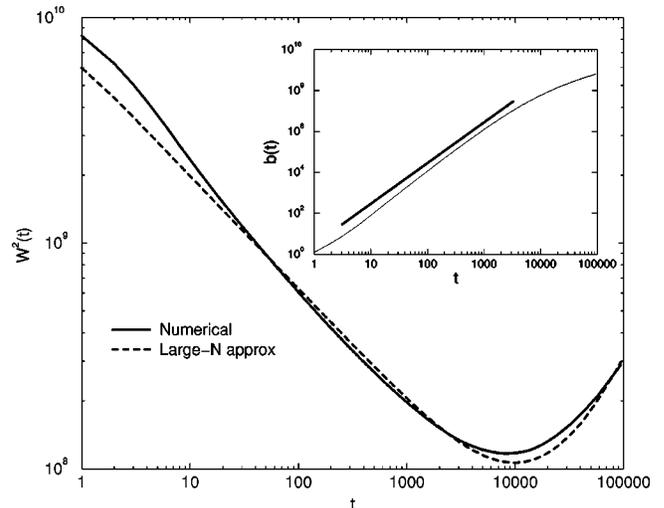


FIG. 6. Double logarithmic plot of $W^2(t)$ vs $t$ for a system with $k_0 = 1/1000$, $W_0 = 10^5$, $Q = 10^8$, $F = 10^4$, $l_D = 100$, and $y = 1/10$. The inset shows the log-log plot of $b(t)$ along with a line of slope 2. For these values of the parameters $8l_D/(a_\| y^2) \ll Q$.

surface smoother, reducing slope fluctuations and making the approximation $(\nabla h)^2 = \langle (\nabla h)^2 \rangle$ increasingly more accurate. Only after the minimum, when the instability takes over, slope fluctuations grow, leading to the breakdown of the large-$N$ approximation.

## VI. DISCUSSION

In the previous sections we have carried out a rather complete analysis of the nonmonotonic behavior of the roughness of a surface which evolves according to the continuum equation for unstable growth. It must be stressed that an initial decrease of the roughness is not necessarily due to a ''rough'' substrate, in the sense of surface width exceeding some threshold. No matter how small the substrate fluctuations, if they extend to a length scale smaller than that of the linear instability, the roughness will initially decrease. Only if substrate fluctuations are limited to relatively large length scales, big amplitudes are needed.

The initial decrease of the roughness is always governed by the relaxational Mullins-like term. The moment when this initial decrease ends depends instead crucially on the value of $Q$ (i.e., on the initial roughness): the minimum may be accounted for by linear (noisy or deterministic) or nonlinear theory. Interestingly, for realistic values of the diffusion length $l_D$ and of the ratio $y = l_{ES}/l_D$, the different regimes are nonoverlapping: It is in principle possible to see each behavior by simply changing $W_0$, i.e., the initial roughness. It is clear, of course, that this is not necessarily true in practice, since experimentally realizable values of $Q$ are limited. Notice, however, that $Q$ depends on two quantities, $W_0$ and $k_0$, and also the variation of the latter could help in expanding the range of variation of experimentally realizable values of $Q$. Moreover, the presence of correlations in the initial roughness enhances the effect of nonlinearity.

With regards to the experimental relevance of the present results it is natural to compare them with the recent data of Gyure *et al.*[9] The comparison of linear theory with the same data[11] pointed out the violation of the naive condition for the irrelevance of nonlinearity, Eq. (26). As shown above the correct condition for the irrelevance is different [Eq. (37)] and turns out to be fulfilled: The substrate is such that nonlinearity does not matter. On the other hand, the experimental parameters give $Q \approx 110$ and $\sqrt{32\pi}/y \approx 450$, indicating that nonconserved noise mostly dictates the position of the minimum. This conclusion is at odds with the results of Ref. 11. The mismatch is due to a different treatment of numerical prefactors and should not be taken too seriously: The precision of values determined from experimental data is poor and already introduces large uncertainty in the physical parameters. However, even if precise experimental data were available a very detailed comparison between theory and experiment would not be possible, because formulas linking parameters of the continuum equation $(\alpha, \kappa)$ with physical quantities $(l_D, l_{ES})$ are known only in order of magnitude. Improved determination of the numerical prefactors would surely be an important contribution to this field of research.

We have considered here only the form (4) of the unstable current, which is valid in the limit of small ES barriers and does not vanish for finite slopes. Other forms of the current are commonly used for large ES barriers $(l_{ES} \gg l_D)$ or when the current vanishes for some ''magic'' value of the slope. The previous analysis can be performed along the same lines for these alternative currents. We do not expect qualitatively different results. In particular, no special behavior should be induced by the presence of magic slopes, since the initial decay of fluctuations governed by the Mullins-like term quickly washes out large slopes independently from the expression of the current.

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[25] The limit of the function for $\theta \to 0$ is here $\pi/8$ and not 1 [as in Eq. (37)] because $t_{min}^l$ is derived in a different way and has a different numerical prefactor.

[26] In the numerical integration of the large-$N$ equation, we solved

Eq. (7) with $a(t)$ given by the spatial average of $(\nabla h)^2$ resulting from the solution of the fully nonlinear equation.

[27] In principle one can compute $W^2(t)$ by integrating the spherically averaged structure factor only for $k < k_0$. However, the aliasing phenomenon implicit in taking the discrete Fourier transform

(Ref. 28), leads to a systematic overestimate of $S(k,t)$ for large wave vectors, inducing a spuriously large roughness.

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