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Linear theory of unstable growth on rough surfaces

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Unstable homoepitaxy on rough substrates is treated within a linear continuum theory. The time dependence of the surface width $W(t)$ is governed by three length scales: The characteristic scale l_0 of the substrate roughness, the terrace size l_D and the Ehrlich-Schwoebel length l_{ES}. If $l_{ES} \ll l_D$ (weak step edge barriers) and $l_0 \ll l_m \sim l_D \sqrt{l_D/l_{ES}}$, then $W(t)$ displays a minimum at a coverage $\theta_{\min} \sim (l_D/l_{ES})^2$, where the initial surface width is reduced by a factor l_0/l_m. The role of deposition and diffusion noise is analyzed. The results are applied to recent experiments on the growth of InAs buffer layers [M.F. Gyure *et al.*, Phys. Rev. Lett. **81**, 4931 (1998)]. The overall features of the observed roughness evolution are captured by the linear theory, but the detailed time dependence shows distinct deviations which suggest a significant influence of nonlinearities.
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I. INTRODUCTION

A high symmetry crystal surface growing epitaxially from a molecular beam can become unstable towards the formation of pyramidal mounds if the mass transport between different atomic layers is reduced by additional energy barriers at step edges.[1,2] Over the last few years, this phenomenon has been observed for a wide range of metal and semiconductor surfaces, and a considerable body of theoretical work has been devoted to the description of the asymptotic (late time) evolution of the surface morphology.[3–5] In the early time regime, continuum theory predicts an exponential growth of the surface modulations. For this reason the precise initial state of the surface has commonly been disregarded, since the exponential instability should rapidly wash out the details of the substrate roughness.

In a recent paper,[6] Gyure, Zinck, Ratsch, and Vvedensky (GZRV) presented experimental and numerical results for the early time development of unstable homoepitaxy from a rough substrate, which show a more complex scenario: It was observed that the competition between smoothening of the initial roughness and the instability associated with the incipient mound structure can lead to a *minimum* in the total surface width. A similar effect was predicted previously in the context of *noise-induced* roughening,[7] and related experimental observations have been reported both for thin metal films[8] and semiconductor multilayers.[9] Qualitatively, the minimum originates from the wavelength dependence of smoothing and (deterministic or stochastic) roughening rates: If the roughness spectrum of the substrate has sufficient weight at short wavelengths, which are efficiently smoothened by capillarity effects,[10] then the decrease of the substrate contribution to the surface width can temporarily dominate the long wavelength roughening induced by growth.

The possibility to minimize the surface roughness by an appropriate choice of the buffer layer thickness and other growth parameters is of obvious interest in applications. In this paper we develop a quantitative theory of unstable growth on rough substrates, which allows us to determine the conditions under which a minimum occurs, and to estimate the layer thickness of minimal roughness in terms of microscopic length scales and parameters, such as the in-layer and interlayer diffusion barriers. Our starting point is the observation of GZRV that the time evolution of the roughness spectrum appears to be well described by the linearized continuum evolution equation for the surface. By incorporating various kinds of noise[11] into the linear theory, we can compare the influence of stochastic and deterministic roughening, and obtain a unified description of both cases. A critical discussion of our results in relation to the experiments of GZRV will be presented at the end of the paper.

II. LINEARIZED CONTINUUM THEORY

The standard phenomenological evolution equation for the continuous surface profile $H(\mathbf{r},t)$ is of the form[1,3–5]

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

where the surface current $\mathbf{J}$ incorporates both a growth-induced destabilizing contribution[3–5,12] and a stabilizing term originating in capillarity,[10] and F denotes the deposition flux, which will be assumed constant for the time being. Small fluctuations $h(\mathbf{r},t)$ around the flat singular surface $H=Ft$ then satisfy the linear equation

$$\partial_t h = -\alpha \nabla^2 h - \kappa (\nabla^2)^2 h, \tag{2}$$

with positive coefficients α, κ representing deposition (α) and smoothening (κ), respectively, whose relation to the growth parameters will be explained below.

The substrate roughness is incorporated through a spatial roughness spectrum $\langle |\hat{h}(\mathbf{k},0)|^2 \rangle = S(k,0) \equiv S_0(k)$, where $\hat{h}(\mathbf{k},t)$ is the Fourier transform of $h(\mathbf{r},t)$ and $k=|\mathbf{k}|$. Under the linear equation (2) the roughness spectrum evolves as

$$S(k,t)=S_0(k)\exp[2(\alpha k^2-\kappa k^4)t], \tag{3}$$

which implies that fluctuations with wave numbers $k>k_c=\sqrt{\alpha/\kappa}$ are damped, while those with $k<k_c$ are exponentially amplified. The surface width $W(t)$ is obtained by summing over all wave numbers,

$$W^2(t)=2\pi\int_0^\infty dkkS_0(k)e^{2(\alpha k^2-\kappa k^4)t}. \tag{4}$$

Motivated by the experimental data shown in Fig. 4 of GZRV, we choose a white noise roughness spectrum,

$$S_0(k)=\begin{cases} l_0^2W_0^2/\pi^3 & : \quad k<\pi/l_0 \\ 0 & : \quad \text{else}, \end{cases} \tag{5}$$

where the small scale cutoff l_0 is required for a finite value $W_0=W(0)$ of the initial surface width. Taking the time derivative of Eq. (4) and evaluating it at $t=0$, we find that the surface width shows an initial *decrease* if $(l_m/l_0)^2>12$, where $l_m=2\pi\sqrt{2\kappa/\alpha}$ is the wavelength[6] of those fluctuations which are maximally amplified by the linear equation (2). Thus the condition for a nonmonotonic time dependence of the surface width is that the length scale characterizing the substrate roughness, l_0, is much smaller than the typical scale l_m of the emerging mounds. This result holds also for more general initial roughness spectra, e.g., $S_0(k)=Ak^{-\rho}$ with $\rho<2$ and a small scale cutoff l_0. For substrates whose roughness is dominated by long wavelength fluctuations, in the sense that $S_0\sim k^{-\rho}$ with $\rho>2$, a large scale cutoff is needed and the time derivative $dW/dt|_{t=0}$ does usually not exist.

In the following we take $l_0\ll l_m$. Then, Eq. (4) reduces to the scaling form

$$W^2(t)=W_0^2(l_0/l_m)^2\Phi(t/\tau), \tag{6}$$

where $1/\tau=\alpha^2/4\kappa$ is the amplification rate of the maximally unstable fluctuations and the scaling function is

$$\Phi(x)=e^{2x}\sqrt{2\pi/x}[1+\text{erf}(\sqrt{2x})], \tag{7}$$

with $\text{erf}(s)=(2/\sqrt{\pi})\int_0^s\exp(-t^2)dt$. The width attains its minimum at a time $t_{\min}\approx0.18\tau$, where it has been reduced by a factor

$$W(t_{\min})/W_0\approx3.7(l_0/l_m). \tag{8}$$

Since the factor $1+\text{erf}(\sqrt{2x})$ in Eq. (7) only varies between 1 and 2, the scaling function $\Phi(x)$ is essentially the product of a decaying power and an exponentially increasing factor. The power law for small x reflects the particular smoothening mechanism (capillarity-driven surface diffusion) and its general form[7] is given by Eq. (18) below. For finite l_0/l_m, the power law sets in for times $t>t_0$ with $t_0\approx(l_0/l_m)^4\tau$.

To relate the behavior of $W(t)$ to microscopic parameters we need to express the coefficients α and κ of Eq. (2) in terms of the two length scales governing unstable homoepitaxy:[3,5] The typical terrace size[13] l_D and the Ehrlich-Schwoebel-length[12]

$$l_{ES}=a_\parallel(D/D'-1)=a_\parallel(e^{\Delta E/k_BT}-1) \tag{9}$$

defined in terms of the in-layer lattice spacing $a_\parallel$, the in-layer (interlayer) surface diffusion constant D (D') and the step edge barrier ΔE. Comparison of the two length scales makes it possible to distinguish conditions of strong ($l_{ES}\gg l_D$) and weak ($l_{ES}\ll l_D$) step edge barriers; in the first case $\alpha\approx Fl_D^2$, in the second $\alpha\approx Fl_Dl_{ES}$. The coefficient κ is traditionally associated with near-equilibrium surface diffusion,[10] however, under far-from-equilibrium growth conditions the dominant contribution to κ is believed to arise from the random nucleation process.[12] The expression $\kappa\approx Fl_D^4$ is then suggested by dimensional analysis[12] and scaling arguments.[14] It leads to a consistent picture[4] in the sense that $l_m\approx l_D$ and $\tau\approx F^{-1}$ in the strong barrier case, which implies that mounds develop on the submonolayer islands already during the growth of the first few layers (''wedding cake'' regime[15,16]). In the weak barrier case we find

$$l_m\sim l_D\sqrt{l_D/l_{ES}} \quad \text{and} \quad \tau\sim F^{-1}(l_D/l_{ES})^2. \tag{10}$$

The minimum in the surface width thus occurs at a coverage

$$\theta_{\min}\sim(l_D/l_{ES})^2\gg1 \tag{11}$$

which corresponds, not surprisingly, to the coverage where mounds first become visible for growth from a smooth substrate.[3,12] Similarly, the coverage $\theta_0=Ft_0$ at which the scaling form (6) for the width begins to hold is of the order of $\theta_0\sim(l_0/l_D)^4$ independent of l_{ES} (provided $l_{ES}\ll l_D$).

To apply these considerations to the experiment on InAs growth of GZRV, we first need to check the condition $l_0\ll l_m$. From Fig. 4 of the paper[6] we estimate that $W_0/W(t_{\min})\approx4$. Comparing this to the theoretical prediction (8) we find $l_0\approx0.07\times l_m$ and $l_m/l_0\gg1$ is true. This is in contrast to the kinetic Monte Carlo simulations of GZRV, where $W_0/W(t_{\min})\approx1.1$. The instability length in the experiment is $l_m\approx1.0$ μm, which yields $l_0\approx70$ nm for the small scale cutoff of the substrate roughness. This is consistent with the initial roughness spectrum in Fig. 4 of GZRV, which is constant at least down to a length scale of 300 nm. The minimum width is attained at a film thickness of about 0.57 μm. Using $a_\parallel\approx6$ Å and a bilayer thickness $a_\perp\approx3$ Å, we therefore estimate that $\theta_{\min}\approx1900$ and $l_m/a_\parallel\approx1700$, and hence $l_{ES}/a_\parallel\approx6$ and $l_D/a_\parallel\approx250$. At the experimental temperature of 500 °C, this implies a step edge barrier ΔE of the order of 0.1 eV, comparable to estimates[3,17] for GaAs.

III. NOISE EFFECTS

Next we include a noise term $\eta(\mathbf{r},t)$ in Eq. (2). The different sources of noise, the individual events of deposition (''shot noise'') and diffusion, enter the noise correlator with different dependence[11,18] on the wave number k. We write it in the form

$$R(k)\equiv\langle\eta(\mathbf{k},t)\eta(-\mathbf{k},t)\rangle=R_S+R_Dk^2, \tag{12}$$

with R_S and R_D denoting the strength of deposition and diffusion noise, respectively. In the linear model with noise the roughness spectrum $S(k,t)$ then contains a part reflecting the history of the noise, as well as the deterministic evolution of the initial roughness treated above. The full expression reads

$$S(k,t)=S_{\mathrm{det}}(k,t)+S_{\mathrm{noise}}(k,t)$$
$$=S_0(k)e^{2(\alpha k^2-\kappa k^4)t}+\frac{R(k)}{2(\alpha k^2-\kappa k^4)}\times[e^{2(\alpha k^2-\kappa k^4)t}-1]. \tag{13}$$

Unlike the deterministic mechanism, the noise increases the amplitude of the spectrum for every wavelength, i.e., $\partial_t S_{\mathrm{noise}}(k,t)>0$ for all k. We shall now examine whether under the experimental conditions of GZRV noise substantially contribute to the surface width.

The deposited particle flux can be seen as a Poisson process with intensity F, so $R_S=a_\perp a_\parallel^2 F$. Shot noise thus contributes to the total width by

$$W_S(t)^2=F\tau a_\perp(a_\parallel/l_m)^2\Psi(t/\tau), \tag{14}$$

with $\Psi'(x)=\Phi(x)$ for the choice (5) of $S_0(k)$. Using Eq. (10), we see that Eq. (14) can be ignored against Eq. (6) if

$$(W_0/a_\perp)^2(l_0/a_\parallel)^2 \gg (l_D/l_{ES})^2. \tag{15}$$

With our estimate of $l_0\approx 70$ nm, this condition is satisfied in the experiment. A different interpretation of Eq. (15) will be given below.

The diffusion noise strength is given by the average rate of adatom jumps on the surface,[11] so

$$R_D\sim\rho_1 D\sim l_D^2 F, \tag{16}$$

where we have used the estimate $\rho_1\sim F l_D^2/D$ for the adatom density.[13] Diffusion noise thus becomes more important than shot noise for $k>\pi/l_D$, whereas it can be neglected for long wavelengths. For large k we can approximate the contribution of diffusion noise in Eq. (13) by $R_D k^2/(\kappa k^4)$ which enters the total width as $W_D(t)^2=l_D^2/l_m^4 F\tau \log(l_D/a_\parallel)$. This means that roughly $W_D(t)^2\approx(l_D/l_m)^2 W_S(\tau)^2 \ll W_S(\tau)^2$, because $l_m \gg l_D$ in the weak barrier regime. In particular, at the time when the width minimum is attained, diffusion noise can be neglected against shot noise, and for the experiment of GZRV, Eq. (6) remains valid.

It was mentioned already that due to noise, a minimum in the surface width may occur even in the absence of step edge barriers.[7] For completeness, we provide here a simple analysis for the most general linear Langevin equation of kinetic roughening,

$$\partial_t h=-(-\nu\nabla^2)^{z/2}h+\eta, \tag{17}$$

where $z=2$ and $z=4$ correspond to evaporation-condensation and surface diffusion dominated relaxation, respectively,[10] $\nu>0$ is a constant, and η is the deposition noise. Odd or noninteger values of z describe nonlocal relaxation mechanisms and can be treated on the same grounds.[3] The linearity of Eq. (17) implies that the substrate contribution and the growth induced contribution to the roughness can be separated.[7] The substrate contribution is found to decay according to

$$W_{\mathrm{sub}}(t)\approx W_0(l_0/\xi(t))^{d/2}, \tag{18}$$

for a d-dimensional surface, for times such that the correlation length of the growth-induced roughness $\xi(t)$ exceeds l_0 (otherwise $W_{\mathrm{sub}}\approx W_0$). In terms of physical quantities, the correlation length can be written as[14] $\xi(t)\approx l_D\theta^{1/z}$. To determine the coverage $\hat{\theta}_{\min}$ of minimal surface width for purely stochastic roughening, the substrate contribution (18) should be compared to the growth induced roughness[14]

$$W_{\mathrm{growth}}\approx a_\perp(\theta/\tilde{\theta})^{(z-d)/2z}, \tag{19}$$

where $\tilde{\theta}$ is the coverage at which the width becomes of the order of $a_\perp$, and thus lattice effects (such as temporal oscillations of the step density) die out; the expression (19) holds for $\theta\gg\tilde{\theta}$. With the estimate[14,19] $\tilde{\theta}\sim(l_D/a_\parallel)^{zd/(z-d)}$, we obtain

$$\hat{\theta}_{\min}\approx(W_0/a_\perp)^2(l_0/a_\parallel)^d \tag{20}$$

independent of z. Comparing Eqs. (20) and (11) we have thus recovered the crossover condition (15) between deterministic instability and stochastic roughening from the opposite side. To neglect the growth instability in Eq. (17) is no longer justified when the minimum width coverage $\theta_{\min}$ predicted by the deterministic theory [Eq. (11)] is smaller than $\hat{\theta}_{\min}$ in Eq. (20).

IV. DISCUSSION AND CONCLUSION

The main results of this paper are Eqs. (6) and (14), which express the time dependent surface roughness in terms of the characteristic length and time scales of the problem—the substrate roughness scale l_0, the incipient mound size l_m, and the linear growth time τ—the latter two of which are, in turn, related to the microscopic growth parameters through Eq. (10). For the experiment of GZRV, the measured values of l_m and τ were seen to imply reasonable numbers for the microscopic lengths l_D and l_{ES}, and for the step edge barrier ΔE.

It is then natural to ask to what extent the linear theory can be used to *quantitatively* describe the experimentally observed roughness evolution, beyond providing consistent order-of-magnitude estimates. Inspection of the data for the surface roughness depicted in the inset of Fig. 4 of GZRV quickly leads to the conclusion that, despite a similar overall appearance, the shape of $W(t)$ is *not* well reproduced by our scaling functions (6) and (14). In fact, the data for the structure factor in Fig. 4 show a *qualitative* feature which the linear theory is unable to explain: It is an immediate consequence of Eq. (13) that $S(k,t)$ is a monotonic function of t (increasing or decreasing) for any k; in contrast, the measured structure factor shows a nonmonotonic dependence on film thickness for $k>k_c$.

This prompts the question whether the use of the linearized theory is really justified under the experimental conditions. Nonlinear terms in the surface current **J** in Eq. (1) are expected[3,4,12] to matter when the surface slope $|\nabla h|$ becomes comparable to $a_\perp/l_D$. Since typical slope values of the initial surface profile are of the order of W_0/l_0, the condition for the validity of the linear theory is

$$W_0/a_\perp<l_0/l_D. \tag{21}$$

In the experiment of GZRV, $W_0/a_\perp \approx 3$ and, from the estimates presented above, $l_0/l_D \approx 0.5$; thus the condition (21) is (weakly) violated. The analogy to phase ordering kinetics[4] suggests that the early time evolution may be qualitatively altered when nonlinearities are important.[20] This seems like an interesting problem for future study.

Note added in proof. A more accurate treatment of the numerical coefficients in Eqs. (10) and (11) yields the estimates $l_D \approx 0.10 \times l_m \theta_{\min}^{-1/4} \approx 15$ nm and $l_{ES} \approx 0.85 \times l_D \theta_{\min}^{-1/2} \approx 3$ Å for the experiment of GZRV, suggesting that the step edge barrier is only about 0.03 eV and the conditin (21) is marginally satisfied. We are grateful to Claudio Castellano for clarifying this point.

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[1] J. Villain, J. Phys. I **1**, 19 (1991).

[2] We assume here that the mounding instability is caused by step edge barriers suppressing *interlayer* transport. The possibility of *corner* barriers which bias the *in-layer* diffusion of atoms along step edges causing an instability has been pointed out recently by several groups [O. Pierre-Louis, M. R. D'Orsogna, and T. L. Einstein, Phys. Rev. Lett. **82**, 3661 (1999); M. V. Ramana Murty and B. H. Cooper, *ibid.* **83**, 352 (1999)]. However, for this scenario the continuum theory is yet to be worked out [P. Politi and J. Krug, cond-mat/9908152, Surf. Sci. (to be published)].

[3] J. Krug, Adv. Phys. **46**, 139 (1997).

[4] J. Krug, Physica A **263**, 170 (1999).

[5] P. Politi, G. Grenet, A. Marty, A. Ponchet, and J. Villain, cond-mat/9906289, Phys. Rep. (to be published).

[6] M. F. Gyure, J. J. Zinck, C. Ratsch, and D. D. Vvedensky, Phys. Rev. Lett. **81**, 4931 (1998).

[7] S. Majaniemi, T. Ala-Nissila, and J. Krug, Phys. Rev. B **53**, 8071 (1996).

[8] F. König, Ph.D. thesis, RWTH Aachen, 1995.

[9] U. Klemradt, M. Funke, M. Fromm, B. Lengler, J. Peisl, and A. Förster, Physica B **221**, 27 (1996).

[10] W. W. Mullins, J. Appl. Phys. **30**, 77 (1959).

[11] D. E. Wolf, in *Scale Invariance, Interfaces, and Non-Equilibrium Dynamics*, edited by A. McKane, M. Droz, J. Vannimenus, and D. Wolf (Plenum Press, New York, 1995), p. 215.

[12] P. Politi and J. Villain, Phys. Rev. B **54**, 5114 (1996).

[13] J. Villain, A. Pimpinelli, L. Tang, and D. Wolf, J. Phys. I **2**, 2107 (1992).

[14] M. Rost and J. Krug, J. Phys. I **7**, 1627 (1997).

[15] J. Krug, J. Stat. Phys. **87**, 505 (1997).

[16] M. Kalff, P. Šmilauer, G. Comsa, and Th. Michely, Surf. Sci. Lett. **426**, L447 (1999).

[17] P. Šmilauer and D. D. Vvedensky, Phys. Rev. B **48**, 17 603 (1993).

[18] Nucleation also consists of random events and enters $R(k)$ with a term (Ref. 11) $R_N k^4$. However, it is dominated by the other noise sources for all k up to $\pi/a_\parallel$.

[19] H. Kallabis, L. Brendel, J. Krug, and D. E. Wolf, Int. J. Mod. Phys. B **11**, 3621 (1997).

[20] C. Castellano and M. Zannetti, Phys. Rev. E **58**, 5410 (1998).