Damping of Growth Oscillations

Harald Kallabis(1,2,5), Lothar Brendel(1,2), Pavel Šmilauer(3), Joachim Krug(4), and Dietrich E. Wolf(2,5)

(1) Höchstleistungsrechenzentrum, Forschungszentrum Jülich, 52425 Jülich, Germany
(2) FB 10, Theoretische Physik, Gerhard–Mercator–Universität GH Duisburg, 47048 Duisburg, Germany
(3) Institute of Physics, Czech Academy of Sciences, Cukrovarnická 10, 162 53 Praha 6, Czech Republic
(4) Fachbereich Physik, Universität GH Essen, D-45117 Essen, Germany
(5) Center for Polymer Studies, Boston University, Boston, MA 02215, USA

November 21, 2018

Computer simulations and scaling theory are used to investigate the damping of oscillations during epitaxial growth on high-symmetry surfaces. The crossover from smooth to rough growth takes place after the deposition of \((D/F)^\delta\) monolayers, where \(D\) and \(F\) are the surface diffusion constant and the deposition rate, respectively, and the exponent \(\delta = 2/3\) on a two-dimensional surface. At the transition, layer-by-layer growth becomes desynchronized on distances larger than a layer coherence length proportional to \(\ell^2\), where \(\ell\) is a typical distance between two-dimensional islands in the submonolayer region of growth.

PACS numbers: 81.10.Aj, 81.15-z, 68.55-a, 05.70.Ln

I. 1 INTRODUCTION

Layer-by-layer or Frank-van der Merwe growth (1) is a growth mode observed in molecular beam epitaxy and other deposition methods which allows precise control of chemical composition of layers down to atomic thickness. It is therefore particularly well suited for the fabrication of novel electronic devices.

The key microscopic processes in layer-by-layer growth are deposition of atoms onto a high-symmetry surface and diffusion of adatoms on the surface. The adatoms meet and form dimers which then grow into islands of monoatomic height whose edges capture most of the adatoms during the deposition of one monolayer. When the island edges become less available due to coalescence of islands, formation of dimers and islands in the next layer begins. The density of atomic steps – and all other quantities sensitive to the surface morphology – thus oscillates in time.

Generically, these oscillations are damped: Layer-by-layer growth is only a transient. Possible reasons (2) include (i) cessation of periodic formation of islands on the surface and transition to step flow growth (3), or (ii) roughening of the surface (4). If the substrate temperature is increased, damping becomes stronger in the first and weaker in the second case, allowing to discriminate between the two. Ignoring the possibility of inhomogeneous deposition (cf. Ref. (3)), surface roughening can have two different sources. If interlayer transport is inhibited by step-edge barriers, one obtains the growth instability predicted by Villain (5). If no such instability occurs, the surface may still roughen due to fluctuations in the intensity of the deposition rate. Only the latter case is considered in this paper.

The question of how long layer-by-layer growth persists is of immediate practical importance. If the answer is known, one can devise and optimize annealing schedules for growing thicker films while maintaining a smooth surface. For this purpose, it is important to know both the damping time \(t\) and the length scale over which layer-by-layer growth is synchronized. This layer coherence length \(\ell\) is a new characteristic length that determines, e.g., the annealing time needed to reestablish a flat surface (6) before growth can be continued.

A theory has been proposed recently (7,8) which predicts that the damping time and the layer coherence length depend on the typical distance \(\ell\) between islands in the submonolayer region of growth as

\[
F \ell \propto \ell^{4(d/4-d)} \quad \text{and} \quad \tilde{\ell} \propto \ell^4(4-d).
\]

In this paper, we present the theory of oscillations and detailed numerical evidence of the validity of its predictions based on extensive computer simulations of a minimal, one-parameter model at surface dimension \(d = 2\). The methods of extracting the damping time and the layer coherence length from the surface morphology evolution are outlined and thoroughly discussed.

The characteristic length \(\ell\) (and thus also \(\tilde{t}\) and \(\tilde{\ell}\)) has a power-law dependence on the ratio \(D/F\) of the surface diffusion constant to the deposition rate:

\[
\ell \propto (D/F)^{\gamma}
\]

(see (2) and references given in (4)). The exponent \(\gamma\) depends on the dimensionality \(d\) of the surface and the (possibly non-integer) dimension \(d_f\) of the islands. It also depends on whether or not desorption of adatoms or diffusion of dimers or larger clusters is negligible. Finally, \(\gamma\) is a function of the critical cluster size \(i^*\) for the formation of a stable nucleus. For the case considered here
II. 2 THEORETICAL RESULTS

Coarse-graining the surface configuration at a given time over a length scale of the order of \( \ell \), one can write down an evolution equation for the variable \( h(x,t) \). Since particle desorption can be neglected under conditions typical for molecular beam epitaxy, the equation can be written in the form of a conservation law,

\[
\partial_t h(x,t) = -\nabla j(x,t) + \eta(x,t). \tag{4}
\]

\( j \) is the surface diffusion current, and \( \eta \) is white noise with second moment

\[
\langle \eta(x,t)\eta(y,s) \rangle = F\delta^d(x-y)\delta(t-s), \tag{5}
\]

which describes the fluctuations in the deposition rate. It was proposed by Villain \[8\] that in growth processes far from equilibrium where local chemical potentials along the surface are ill defined, diffusion currents should be driven by gradients in the growth-induced, nonequilibrium adatom density \( n \),

\[
j = -D\nabla n. \tag{6}
\]

On a singular surface, the balance between deposition and capture of adatoms at steps leads to a stationary adatom density \( n = n_0 \) of the order of \[15\] \( n_0 \approx (F/D)\ell^2 \).

On a vicinal surface, the adatom density is reduced due to the presence of additional steps. However, this effect is felt only if the miscut \( m = |\nabla h| \) exceeds \( 1/\ell \), in which case \( n \approx (F/D)m^{-2} \). In terms of a coarse-grained description of the surface this implies that the local adatom density depends on the local miscut or surface tilt. A useful interpolation formula which connects the regimes \( m \ll 1/\ell \) and \( m \gg 1/\ell \) is \[16\]

\[
n(\nabla h) = \frac{n_0}{1 + (\ell\nabla h)^2} \approx (F/D)\ell^2 - (F/D)\ell^4(\nabla h)^2 + \ldots \tag{7}
\]

Inserting the leading quadratic term of this gradient expansion into \[8\], which is appropriate for describing long-wavelength fluctuations around the singular orientation, one obtains

\[
\eta = \nabla \lambda(\nabla h)^2 \tag{9}
\]

with

\[
\lambda = F\ell^4. \tag{10}
\]

Combining Eq. \[1\] and Eq. \[9\] one sees that the physical dimension of \( \lambda \) is \((\text{length})^4/(\text{time}\cdot\text{height})\). Within the continuum description, the only characteristic length and time scales are the layer coherence length and the damping time, whereas the lattice constant \( a \) has been chosen as a unit of height. Therefore

\[
\lambda \propto \ell^4/\tilde{\ell} \tag{11}
\]
on dimensional grounds.

Finally, the number of particles deposited during the time \( \tilde{\ell} \) onto an area \( \ell^d \) is \( F\ell^d(\tilde{\ell}^d)^{1/2} \). Thus the fluctuation of the film thickness over the distance \( \tilde{\ell} \) is \( w(\tilde{\ell}) \approx \sqrt{F}\ell^d \ell^d/\tilde{\ell}^d \). At \( \tilde{\ell} \) this should be the thickness of about one atomic layer, \( w(\tilde{\ell}) \approx 1 \), which results in

\[
F\tilde{\ell} = \tilde{\ell}^d. \tag{12}
\]

Combining \[10\], \[11\] and \[12\] one obtains Eq. \[1\], or

\[
F\tilde{\ell} \approx (D/F)^\delta \quad \text{and} \quad \tilde{\ell} \approx (D/F)^{\delta/d} \tag{13}
\]

with the exponent

\[
\delta = \frac{4d}{4-d}\gamma. \tag{14}
\]

Notice that the layer coherence length \( \tilde{\ell} \) is substantially larger than the characteristic distance \( \ell \) between islands (\( \tilde{\ell} \approx \ell^2 \) at \( d = 2 \)).

III. 3 MODEL

In our model, atoms are deposited onto the (100) surface of a simple cubic lattice with the rate of \( F \) atoms per unit time and area. The surface size is \( L \times L = 128 \times 128 \) \[17\]. Atoms with no lateral neighbors are allowed to diffuse with diffusion constant \( D \). Atoms with lateral neighbors are assumed to be immobile so that, e.g., dimers are immobile and stable. Growth commences on a flat substrate, \( h(x,0) = 0 \) for all sites \( x \). On deposition at \( x \), \( h(x,t) \) is increased by one. We neglect barriers to interlayer transport (Ehrlich–Schwoebel barriers \[18\]) so that the only parameter of the model is the ratio \( D/F \).
IV. 4 DAMPING TIME

First we present the results for the damping time extracted from kinematic intensity data,

\[ I \equiv \langle (N_{\text{even}} - N_{\text{odd}})^2 / L^2 \rangle \]  

(see Fig. 1; \( N_{\text{even}} \) \( (N_{\text{odd}}) \) denotes the number of atoms in even (odd) layers). The brackets \( \langle \ldots \rangle \) denote averaging over different runs. The same analysis was done for the surface width with equivalent results for the damping time.

The kinematic intensity oscillates between zero and maxima which decrease until the oscillations vanish. We measure \( \tilde{t} \) as the time where the maxima of the kinematic intensity drop below \( I = 0.05 \) [19]. The results are shown in Fig. 2.

Obviously there are strong corrections to scaling which can be attributed to an offset \( \tilde{t}_0 > 0 \):

\[ \tilde{t} = A_1 \left( \frac{D}{F} \right)^{\delta} - \tilde{t}_0. \]  

(16)

\( \tilde{t}_0 \) plays the role of a cutoff for the validity of our scaling theory. \( (D/F)_0 \equiv (\tilde{t}_0 / A_1)^{1/\delta} \) can be interpreted as that value of \( D/F \) below which the oscillations are not observable anymore [20].

A three-parameter fit to the data shown in Fig. 2 gives an exponent

\[ \delta = 0.69 \pm 0.05, \]  

(17)

where the error bar reflects the variations obtained when the evaluation method is modified [19,20] or when the data for the surface width are evaluated in the same way. This value is in good agreement with the theoretical prediction of \( \delta = 2/3 \) (see Eq. (14)) for compact islands.

In our simulation, neither detachment of adatoms from islands nor edge diffusion are considered. Therefore, the islands are fractal for large diffusion lengths [21–23] with the fractal dimension \( d_f \approx 1.72 \) of two-dimensional diffusion limited aggregation [24]. Then \( \gamma \) changes from \( \gamma = 1/6 \) to \( \gamma \approx 0.175 \). This leads to theoretical prediction \( \delta \approx 0.70 \) which is also within the error bars of
Eq. (17). However, in the present case the values of $D/F$ are sufficiently small, so that this complication may be ignored.

The scaling plot of the kinematic intensity with the time divided by the damping time according to Eq. (16) (see Fig. 3) confirms the validity of the approach used.

V. 5 LAYER COHERENCE LENGTH

The measurement of the height difference correlation function \[ G(x, t) \equiv \langle [h(x_0, t) - h(x_0 + x, t)]^2 \rangle, \] (18)
evaluated at $t = \tilde{t}$ shows that $G$ has a maximum. This can be explained as follows. At $t = \tilde{t}$ the probability to find the surface at the same height as at a reference point $x_0$ is minimal at a distance $x - x_0$ corresponding to the layer coherence length. For larger distances, deviations from the average height are essentially uncorrelated. At very small distances, their correlation is positive, while around $\tilde{t}$ they are anticorrelated. This is how the data denoted by the squares in Fig. 4 were obtained. The result is in good agreement with the predicted exponent, cf. Eq. (13). (Note that this method could be also used for experimental determination of $\tilde{\ell}$.)

\[ A^2(t) \equiv \langle w^2(t) \rangle_{[t,t+\tau]} - \langle w(t) \rangle_{[t,t+\tau]}^2 \] (19)
of the surface width during the layer completion time $\tau \equiv 1/F$, where $\langle \ldots \rangle_{[t,t+\tau]}$ denotes the time average over the interval $[t, t + \tau]$. Its stationary value decreases with increasing system size and ultimately becomes equal to the statistical fluctuations of $w$ when the system size is big enough so that the oscillations can die out completely. The values of $\tilde{\ell}$, denoted by the circles in Fig. 4 represent the linear system size $L$ at which the stationary value of $A(t)$ drops below 0.37. Both methods of measuring $\tilde{\ell}$ are in excellent agreement with each other and with the theoretical prediction [25].

VI. 6 CONCLUSIONS AND OUTLOOK

We have presented a theory for the damping of growth oscillations caused by kinetic roughening. We have shown that the results of numerical simulations of a minimal model compare very favorably with the theory, and directly determined two key quantities, the damping time and the layer coherence length. The instability associated with barriers to interlayer transport [8] may compete with the kinetic roughening mechanism as a source of oscillation damping [26]. This, as well as the transition to step-flow growth on vicinal surfaces will lead to different power laws for the damping time and the layer coherence length. This remains for future research.

The results of this paper can be directly verified by diffraction or real-space surface sensitive techniques. The determination of the damping time and, in particular, of the layer coherence length as a function of growth conditions should be possible using the methods outlined above.

VII. ACKNOWLEDGEMENTS

Useful conversations with Martin Rost are gratefully acknowledged. D. E. W. acknowledges support by DFG within SFB 166 Strukturelle und magnetische Phasenübergänge in Übergangsmetall-Legierungen und Verbindungen. J. K. acknowledges support by DFG within SFB 237 Unordnung und grosse Fluktuationen. P. S. acknowledges the financial support of Alexander von Humboldt Foundation and Volkswagen Stiftung. H. K. acknowledges support by the German Academic Exchange Service within the Hochschulsonderprogramm III.

[1] E. Bauer, Z. Kristallogr. 110, 372 (1958).
[2] D. E. Wolf, in: *Dynamics of Fluctuating Interfaces and Related Phenomena*, eds. D. Kim, H. Park and B. Kahng (World Scientific, Singapore, 1997) pp. 173 – 205

[3] H. Kallabis, P. L. Krapivsky, and D. E. Wolf, Eur. Phys. J. B 5, 801 (1998).

[4] J. Sudijono, M. D. Johnson, C. W. Snyder, M. B. Elowitz, and B. G. Orr, Phys. Rev. Lett. 69, 2811 (1992); M. D. Johnson, J. Sudijono, A. W. Hunt, and B. G. Orr, Appl. Phys. Lett. 64, 484 (1994).

[5] K. Theis-Bröhl, I. Zoller, P. Bödeker, H. Zabel, L. Brendel, M. Belzer and D. E. Wolf, Phys. Rev. B 57, 4747 (1998)

[6] H. C. Kang and J. W. Evans, Surf. Sci. 271, 321 (1992); M. C. Bartelt and J. W. Evans, Phys. Rev. Lett. 75, 4250 (1995).

[7] R. Kunkel, B. Poehsema, L. K. Verheij and G. Comsa, Phys. Rev. Lett. 65, 733 (1990)

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

[9] W. W. Mullins, J. Appl. Phys. 28, 333 (1957); L.-H. Tang, in: *Dynamics of crystal surfaces and interfaces*, eds. P. M. Duxbury and T. Pence, (Plenum, NY, 1997), pp. 169-184.

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

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

[12] G. Zinsmeister, Thin Solid Films 2, 497 (1968); ibid. 4, 363 (1969); ibid. 7, 51 (1971).

[13] F. Family and T. Vicsek (eds.): *Dynamics of Fractal Surfaces* (World Scientific, Singapore 1991).

[14] The consideration of the linear contribution to the surface current leads to the same result, see Refs. [11, 10]

[15] J. Villain, A. Pimpinelli, and D. Wolf, Comments Cond. Mat. Phys. 16, 1 (1992).

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

[17] At high $D/F$ ratios, simulations were performed using systems of linear size $L = 200$ to ensure that finite size effects are negligible: Within the statistical fluctuations there was no observable change in the measured quantities.

[18] G. Ehrlich and F. G. Hudda, J. Chem. Phys. 44, 1039 (1966); R. L. Schwoebel and E. J. Shipsey, J. Appl. Phys. 37, 3682 (1966).

[19] In order to check the robustness of our results, we repeated the same analysis for $I = 0.1$ and obtained the same exponent $\delta$ within the error bar.

[20] We checked that another possible Ansatz $\tilde{i} = A_i[|D/F – const.|]^\delta$ only changes the exponent within the error bar.

[21] J. Villain, A. Pimpinelli, and D. E. Wolf, J. Phys. I (France) 3, 447 (1993).

[22] J. G. Amar, F. Family, and P.-M. Lam, Phys. Rev. B 50, 8781 (1994).

[23] P. Jensen, A.-L. Barabasi, H. Larralde, S. Havlin, and H. E. Stanley, Fractals 4, 321 (1996).

[24] T. A. Witten and L. M. Sander, Phys. Rev. Lett. 47, 1400 (1981).

[25] Details of the data analysis will be published elsewhere (H. Kallabis and D. E. Wolf, in preparation).

[26] L. Brendel, Ph. D. dissertation, Gerhard–Mercator–Universität Duisburg, in preparation.