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Correlated Island Nucleation in Layer-by-Layer Growth

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Abstract. — Temporal correlations of nucleation events in layer-by-layer growth are studied by simulating a one dimensional model. In order to focus on fluctuations due to the nucleation process, deposition and diffusion are treated deterministically. The correlations decay algebraically with effective exponents depending on the order of the nucleations in a layer. The autocorrelation function of all nucleation events decays as \( t^{-1.35} \) with time \( t \). In spite of these correlations, the corresponding randomness in the growth process does not lead to a roughening of the surface. The reason is that stochastic nucleation implies only conserved current fluctuations.

1. Introduction

Recently there has been increasing interest in nanostructured thin films produced e.g. by molecular beam epitaxy (MBE) [1–4]. In this context a better theoretical understanding of layer-by-layer growth is highly desirable. This type of growth proceeds by nucleation of two-dimensional islands, their growth and coalescence leading to the completion of a new atomic layer of the film. Whereas spatial correlations among these islands within a layer are essentially understood (see Ref. [5] and references therein), we address here the question to what extent the nucleation of subsequent layers remains correlated with the first one. This allows to estimate how many layers one can grow without washing out the initial nucleation pattern.

In MBE atoms are deposited onto a substrate. In layer-by-layer growth they diffuse until they either stick to the edge of a stable twodimensional island or meet with other adatoms to form a new nucleus of such an island. The existence of temporal correlations is very plausible for high symmetry surfaces: the first nucleation event of a new layer is likely to happen close to the one in the previous layer, because the corresponding island grows ahead of the other ones and provides the largest area without traps for adatoms in the next layer. The aim of this paper is to study the character of these correlations.

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Fig. 1. — Cells A and B are sinks for adatoms, i.e. on the time scale $\Delta t$ all adatoms within them will condense to the island edges which exist somewhere in them. Here $m$ has the meaning of condensed material (shaded) on top of completed layers, and the adatom density $\bar{m} = 0$. In all other cells $m = \bar{m}$. Cells A were invaded by the edges of a growing island, in B a nucleation event has happened. All cells, indicated by the dashed lines, are of width $\Delta x$ and height 1 (in units of the atomic lattice constant).

It is an experimental fact that layer-by-layer growth can be sustained for hundreds of layers, if the growth conditions are carefully chosen [6]. In particular a Schwoebel instability [7] should not occur. We show that randomness associated with the nucleation process does not destroy layer-by-layer growth, as long as shot noise and diffusion noise can be neglected. Whereas the latter two are well understood [8], very little is known about the nature of fluctuations due to the nucleation process. In this paper we focus on these properties by introducing a model where all other sources of noise are intentionally eliminated in order to see the bare contribution of the nucleation events. This is achieved by simulating deposition and diffusion on a coarse scale in a deterministic continuum approximation.

2. The Model

We assume that desorption as well as defects in the film can be neglected and that the adatoms are bound irreversibly when they meet a step or another adatom. Then the growth is controlled by the ratio of the surface diffusion constant $D$ to the deposition rate $F$. For simplicity we consider the one dimensional case. Then the typical distance between the island centers is $l \sim (D/F)^{1/4}$ [9].

We divide the substrate into cells of $\Delta x$ lattice constants. This coarsening length must of course be chosen smaller than the typical distance $l$ between nucleation events, otherwise one could not resolve them. Each cell contains an integer number $h(x,t)$ of completed layers and a partial filling $0 \leq m(x,t) \leq \Delta x$ on top of them (Fig. 1). In order to suppress shot noise and fluctuations in the hopping of adatoms, $m(x,t)$ has to vary continuously rather than atom by atom. For example, during one simulation update corresponding to a time increment $\Delta t$ it increases by $F\Delta x\Delta t$ due to deposition. It also changes according to the diffusion equation

$$m(x, t + \Delta t) = m(x, t) + D \Delta t (\Delta x)^{-2} \times$$
$$\left( \bar{m}(x + \Delta x, t) - 2\bar{m}(x, t) + \bar{m}(x - \Delta x, t) \right),$$

(1)

where $\bar{m}(x, t)/\Delta x$ is the adatom density. For cells in which there is no island edge, $\bar{m}$ coincides with $m$.

Cells in which there is an island edge act as sinks for adatoms, $\bar{m} = 0$, thereby filling up irreversibly until $m(x,t)$ exceeds $\Delta x$. Then a new layer is completed in that cell: The island edge invades a neighboring cell or annihilates with another edge (coalescence), $h(x,t)$ is incremented by 1, and $m(x,t)$ is reduced by $\Delta x$. 

394 JOURNAL DE PHYSIQUE I N°3
New islands nucleate only in cells without an island edge. The nucleation rate has two terms

\[ R_{\text{nuc}}(x, t) = \tilde{m}(x, t)[F \Delta x + cD(\Delta x)^{-2}(\tilde{m}(x + \Delta x, t) + \tilde{m}(x - \Delta x, t))]. \]  

(2)

They describe the two main processes leading to a nucleation event: if already one adatom is in the cell at \( x \) (probability \( \tilde{m} \)), a second one may i) be deposited into the same cell or may ii) diffuse into it from its neighborhood. During time \( \Delta t \) nucleation happens with a probability \( 1 - \exp(-R_{\text{nuc}}\Delta t) \). The constant \( c \) in (2) is of order 1. We did simulations for \( c = 2.5 \) (results below) and for 0.6. The exponents reported below did not depend on this.

3. Simulation Results

For the simulations we chose \( \Delta t = 0.2(\Delta x)^2/D \). This shows that coarse graining makes the algorithm very efficient \cite{10}; the number of updates needed for growing a film of given thickness is reduced by a factor \((\Delta x)^2\). As each update involves \( L/\Delta x \) cells, computation time is reduced by another factor \( \Delta x \). In most of the simulations we used \( D/F = 10^4 \), for which \( l \approx 12 \) was found. Coarse graining over \( \Delta x = 3 \) did not change the results but reduced the computation time by a factor \( \approx (\Delta x)^2 \approx 27 \) compared to a simulation without coarse graining.

We recorded all positions \( x_n(h) \) of the \( n \)-th nucleation event (\( n = 1, 2, 3, .. n(h) \)) in the layer at height \( h \). Figure 2 shows the positions \( x_1(h) \) and \( x_2(h) \) of the first two nucleation events during the growth of 1000 layers. Strong temporal correlations along the \( h \)-direction are obvious. In all our simulations no more than two incomplete layers were observed at any time, so that the nucleation events at height \( h \) happen at a time (in units of monolayers) \( h - 1 < t < h + 1 \).

We evaluated the spatial correlation function among all nucleation events,

\[ C(r) = \langle \nu(x, h)\nu(x + r, h) \rangle - \langle \nu \rangle^2, \]

(3)

the autocorrelation function among all nucleation events,

\[ A(\tau) = \langle \nu(x, h)\nu(x, h + \tau) \rangle - \langle \nu \rangle^2, \]

(4)
and the autocorrelation functions among the $n$-th nucleation events,

$$A_n(\tau) = \langle \nu_n(x, h) \nu_n(x, h + \tau) \rangle - \langle \nu_n \rangle^2$$

(5)

where $\nu_n(x, h)$ is 1 or 0 depending on whether or not the $n$-th nucleation event at height $h$ happened in cell $x$, and $\nu(x, h) = \sum_n \nu_n(x, h)$. The averaging $\langle \ldots \rangle$ was done over $x$, $h$ (discarding the 100 initial layers) and over 20 runs. $\langle \nu \rangle$ is the average number of nucleation events in a layer divided by the number of cells, $\langle \nu \rangle = \Delta x / l$. Similarly, $\langle \nu_n \rangle = \Delta x / L$ apart for $n \approx L / l$, where an $n$-th nucleation event does not occur in every layer.

The spatial correlation function $C(\tau)$ shows the expected anticorrelation at short distances: within a distance $l$ of a nucleation site further nucleation events are suppressed. On larger distances no correlation can be observed among the nucleation events.

Figure 3 shows that the temporal correlation among all the nucleation events decays with a power law

$$A(\tau) \sim \tau^{-\alpha}, \quad \text{with} \quad \alpha = 1.35 \pm 0.1.$$  

(6)

Although 10000 layers were grown, $A(\tau)$ could only be evaluated over two decades in $\tau$ because of the bad statistics for large height differences $\tau$.

The correlation among the first nucleation events in every layer decays much more slowly, Figure 4,

$$A_1(\tau) \sim a_1 \tau^{-\alpha_1}, \quad \text{with} \quad \alpha_1 = 0.48 \pm 0.08.$$  

(7)

Similarly, the correlations among the $n$-th nucleation events were fitted with a power law between $\tau = 5$ and $\tau = 1000$, giving effective exponents $\alpha_n$ (Fig. 5). They vary between low values $\sim 0.4$ for the first and the last nucleation events to values between 0.7 and 0.8, indicating that the intermediate nucleation events decorrelate faster.

Apart from a decrease by about one decade for the first 5 nucleation events the prefactors $a_n$ of the power laws remain essentially constant (Fig. 6). The sharp drop of the amplitudes for $n > 22$ is due to the fact that the number of nucleation events in one layer was about 22 for our system size and exceeded this value only very seldomly.

Obviously, there must be cancellation of the $A_n$ terms by the cross correlations $\langle \nu_n \nu_m \rangle - \langle \nu_n \rangle \langle \nu_m \rangle$ with $n \neq m$ since $A(\tau)$ decays faster than the $A_n(\tau)$-s. The anticorrelations are
Fig. 4. — Correlation between first nucleation events in two layers versus their height separation. Fit has slope $-0.48$.

Fig. 5. — Temporal correlations among $n$’th nucleation events decay with exponents $\alpha_n$.

plausible as it is very unlikely that two nucleation events with $|n - m| \approx L/2l$ appear in the same cell. Our results show that there is an approximate cancellation in the leading $\tau$-dependence of the positive and negative terms in $A(\tau)$ and the fast decay (large exponent) is due to the non-vanishing corrections. We cannot be sure, of course, that we see the asymptotic behavior: If the cancellation is not perfect, finally the smallest exponent should overtake. However, we have not detected any crossover of this kind.

4. A Simplified Analytical Approach

In order to understand the results on the correlations $A, A_n$ let us discuss a somewhat simplified picture: we assume that the total number of nucleation events in a layer is a constant, $N$. As indicated by the rapid decay of the spatial correlations, there is a characteristic “exclusion zone” of size $l$ around each nucleation event. Therefore we imagine the whole system divided
into $N$ cells of length $l$.

The cells in each layer are now numbered in the order of the nucleation events (one per cell) from 1 to $N$. From layer to layer one will find only small changes in the sequence: most likely the serial number attributed to a cell will not change. As mentioned in the beginning, the first nucleation event in a layer happens close to the one in the previous layer (i.e. in the same cell). The reason is that adatom sinks (island edges) vanish there first, when the layer fills up, so that the adatom density will be higher there than elsewhere on the surface. Hence the nucleation probability is largest there. Similarly the second nucleation event in the new layer will most likely happen in the same cell as in the previous layer: after the first nucleation created a first sink for adatoms in the new layer, one has to look for the region of maximal adatom density outside its depletion zone. This will be the region which has been devoid of island edges for the next-to-longest period. By induction one expects that also the $n$-th nucleation event has a higher probability to happen, where it took place before, than anywhere else.

Nevertheless, due to the stochastic nature of the nucleation process, the first and the second nucleation events may be interchanged occasionally, for example. More complicated permutations like $(1 \rightarrow 2, 2 \rightarrow 3, 3 \rightarrow 1)$ or $(1 \rightarrow 3, 3 \rightarrow 1)$ will be less probable and the probability will rapidly decay with the range of the jump in the space of serial numbers. Thus any fixed cell performs a random walk in this space. The correlation function $A_n(\tau)$ corresponds then to the probability that a cell starts with the serial number $n$ and returns to it after $\tau$ steps. This is a plausible model for the observed temporal correlations in the absence of spatial ones, since the mechanism leading to the power law time decay is the wandering of nucleation events not in real space but in the space of their serial numbers. It predicts a decay like $A_n(\tau) \sim \tau^{-1/2}$ with exponents $\alpha_n = 1/2$ which fits very well to $n = 1$ but not to medium values of $n$.

In this simplified model $A(\tau)$ vanishes identically: there is a nucleation event in every cell in every layer. In this case the cancellation of the positively and negatively correlated terms is exact. This gives some confidence that indeed there will be an almost perfect cancellation in our simulation, too. The remaining weak correlation may be due to the fluctuation of the nucleation site within a cell of size $l$ or due to the fluctuation of $N$, both of which have been ignored in the simplified model.

The measured values of $\alpha_n$ vary between 0.3 and 0.8 with error bars of at least 0.1. Presumably this variation can be accounted for by making the random walk of the cells over the

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Fig. 6. — Amplitudes $a_n$ of temporal correlation function for $n$'th nucleation events.
serial numbers more realistic. For example the nucleation events with serial numbers close to \( N/2 \) will exchange their positions much easier than the first or last ones. From one layer to the next several cells may change their serial numbers simultaneously, and larger step sizes will be allowed. This makes the combinational problem more difficult and probably decorrelates the nucleation events on different layers.

5. Continuum Description of Stochastic Nucleation

The simulations showed no indication for kinetic roughening [11]: even for values of \( D/F \) as small as 10, system sizes up to \( L = 10000 \) and up to 10000 deposited layers no more than two layers were incomplete at any time. This agrees with the finding for another MBE-model without shot- and diffusion noise [12]. The randomness of the nucleation events does not lead to significant surface roughness, in spite of its temporal correlations. In the following we propose a continuum description of the growth we simulated and explain why stochastic nucleation did not lead to the roughening of the surface, while random diffusion and even more so the shot noise will make the surface rough, ultimately.

Our starting point is a coarse grained description of the growth,

\[
\partial_t h = F - \nabla j + \eta, \tag{8}
\]

where \( j \) is the surface current and \( \eta \) is a noise term. In our simulations the only contribution to \( \eta \) is nucleation noise and \( j \) is a functional of the adatom density that is determined by the distribution of islands and the values of \( h \). Two questions have to be answered: what is the constitutive law specifying the surface current as a functional of \( h \), and what is the appropriate description of the noise due to stochastic nucleation?

The first question is easily answered by observing that the present model does not allow tilt induced currents. No matter what the global tilt of the substrate is, the average current vanishes due to the periodic boundary conditions. Hence the generic form of the constitutive law is

\[
j = \nabla (K \nabla^2 h + \lambda (\nabla h)^2), \tag{9}
\]

where the linear term only matters if \( \lambda = 0 \) [7,13,14].

The fluctuations of the surface current are governed by those of the adatom density, \( \rho(x,t) \),

\[
\delta j = \nabla \rho. \tag{10}
\]

The only source of density fluctuations on the surface are nucleation events, since random hopping has been eliminated. These density fluctuations are correlated only on short scales of order \( l \). On a coarse grained scale which is implied by the continuum description these density fluctuations can be viewed as spatially uncorrelated noise, \( \langle \rho(x,t)\rho(x',t) \rangle \sim \delta(x-x') \).

On the other hand, the temporal correlation among all the nucleation events directly translates into a temporal correlation among the adatom density fluctuations, \( \langle \rho(x,t)\rho(x,t') \rangle \sim |t - t'|^{-\alpha} \), where \( \alpha \) is the same exponent as in (6).

Hence we find that nucleation events lead to conserved, temporally correlated current fluctuations

\[
\langle \delta j(x,t)\delta j(x',t') \rangle \sim \nabla^2 \delta(x-x') |t - t'|^{-\alpha} \tag{11}
\]

The fluctuations \( \eta \) of the growth velocity in (8) are then the divergence of the current fluctuations, i.e.

\[
\langle \eta(x,t)\eta(x',t') \rangle \sim \nabla^4 \delta(x-x') |t - t'|^{-\alpha} \tag{12}
\]
Now we show that the spatial anticorrelation of the current fluctuations on short scales (i.e. on scale $l$) as expressed by the regularized $\nabla^2 \delta(x - x')$ counteracts the temporal correlation and renders it ineffective for kinetic roughening. As usual, roughening is characterized by dynamical scaling of the height-height correlation function, 
\[(h(x, t) - h(x + r, t))^2 \sim r^{2\zeta} \rho(r/t^{1/z})\] with the roughness exponent $\zeta$ and the dynamical exponent $z$ [11].

Comparing the scaling dimensions of the deterministic terms in (8) yields the exponent identities [7,8,14]
\[z = 4 \quad \text{for} \quad \lambda = 0, \quad z = 4 - \zeta \quad \text{for} \quad \lambda \neq 0.\] (13)

Comparing the scaling dimension of the noise term with that of the left hand side of (8) one obtains the scaling relation [8,13,14]
\[2\zeta - z(2 - \alpha) = -4 - \hat{d}',\] (14)
where $\hat{d}'$ is the dimension of the surface, $\hat{d}' = 1$ in the present case. Together with (13) this fixes the exponents. We only need to specify the linear case ($\lambda = 0$),
\[\zeta_0 = (4 - \hat{d}' - 4\alpha)/2,\] (15)

as the roughness exponent for the nonlinear equation is given by $\zeta = 2\zeta_0/(4 - \alpha)$. Equation (15) implies that for sufficiently fast decaying temporal correlations, $\alpha > 1 - \hat{d}'/4$, random nucleation does not roughen the surface. In the present case, for the times simulated, the effective value of $\alpha$ is larger than 3/4, which explains the absence of roughening.

6. Summary

In summary we have shown in a one-dimensional model that long time correlations in nucleation events occur during layer-by-layer growth. These are not accompanied by spatial correlations since the mechanism leading to the power law time decay is the wandering of nucleation events not in real space but in the space of their serial numbers. In spite of the correlations the nucleation noise does not roughen the surface since the conservation of current fluctuations leading to short range anticorrelations compensates the slow decay.

There are several interesting questions related to the problems investigated here. First, it is clear that in real systems nucleation noise coexists with the shot noise and the diffusion noise. It should be important to understand how the latter affect the temporal correlations described above. Another open problem is the behavior in higher dimensions. Our simplified random walk argument is independent of the dimensionality and therefore we expect slow decay of correlations there as well. However, the random walk picture uses a coarsening scale which is equivalent to the island size. For smaller scales the migration of nucleation events on the island (see the wigging of connected patches in Fig. 1) becomes important and the different dimensionality may play a role and could lead to a faster decay of correlations. Thus we do not expect that roughening due to nucleation noise would occur since $\alpha > 1/2$ already leads to smooth surfaces for 2+1 dimensions.

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