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LETTER TO THE EDITOR

Chapman-Kolmogorov equation for Markov models of epitaxial growth

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Abstract. We outline a probabilistic approach to modelling epitaxial growth, whereby we solve for the complete probability distribution of the surface using the Chapman-Kolmogorov equation. We describe a simple Markov growth model which is analytically solvable using this approach and exhibits damped oscillations of the step density. The model specifies that perfect layer growth occurs within subsections of an adjustable size on an atomically smooth surface. Increasing the size of a subsection increases the amplitude of the oscillations in the step density and decreases the rate of decay of the oscillations. The mechanism for the damping of the oscillations is shown to be phase incoherence between growth occurring in different subsections.

The preparation of sophisticated semiconductor devices using molecular-beam epitaxy (MBE) demands a thorough understanding of the growth processes occurring at the surface. Experimentally, the most widely used probe of the morphology of the growth front during epitaxy is reflection high-energy electron diffraction (RHEED) (Neave *et al* 1983, Van Hove *et al* 1983). Using this technique, the time evolution of the surface structure may be followed by monitoring the intensity of the reflected beam. Utilisation of MBE to grow better devices would be greatly facilitated by theoretical models which could elucidate the important growth mechanisms and allow unambiguous interpretation of RHEED measurements. However, due to the complexity of the growth process, most existing theoretical models are based on mean-field theories in which kinetic rate equations are solved for average quantities such as the coverage of a layer (Weeks and Gilmer 1979, Venables *et al* 1984, Stoyanov and Michailov 1988, Cohen *et al* 1989).

In this letter we propose a new approach to modelling of epitaxial growth: the use of the Chapman-Kolmogorov equation to find the probability of a configuration of the surface atoms. There are several advantages of this approach over the mean-field approaches: (i) the complete probability distribution of the surface is obtained rather than an average quantity; thus it is possible to calculate quantities related to the microscopic arrangement of atoms, such as the surface step density; (ii) since the Chapman-Kolmogorov equation makes no assumption concerning the equilibrium nature of the system, it is ideally suited to the far-from-equilibrium growth conditions typically encountered in MBE; (iii) a formally exact description of the microscopic kinetics is an appropriate starting point for the systematic derivation of modified diffusion equations, especially using techniques developed for birth-death processes (Gardiner 1985), of which MBE is an example (Cohen *et al* 1989). Description of the approach and its application to several growth models is given below.

The substrate is composed of sites arranged in a simple cubic lattice oriented in a $\langle 100 \rangle$ direction. The total number of sites on the substrate is N ; the lattice is square with \sqrt{N} sites along a side. Periodic boundary conditions are imposed parallel to the substrate. Prior to growth the substrate is flat with all sites unoccupied. During growth, atoms impinge on random substrate sites at regular time intervals of τ_a ; no evaporation or diffusion occurs. Growth is therefore modelled as a series of discrete events at times $m\tau_a$ ($m = 1, 2, \dots$), each event being the deposition of one atom on the surface. The time for deposition of a monolayer is τ , where $\tau = N\tau_a$. Attachment of atoms is assumed to follow the solid-on-solid (sos) model (Weeks and Gilmer 1979) where vacancies and overhangs are forbidden.

The state of the lattice during growth is characterised by a matrix \mathbf{H} , where matrix element $h_{i,j}$ is the height of the site at $[i, j]$ (also the number of atoms deposited on that site if $\mathbf{H} = \mathbf{0}$ at the commencement of growth). We introduce the probability of a particular configuration \mathbf{H} after a total of m atoms have been deposited:

$$P(\mathbf{H}; m) = P(h_{i,j}, \hat{\mathbf{H}}; m) \quad (1)$$

where $\hat{\mathbf{H}}$ denotes the matrix of all h not explicitly written.

For the above model, a configuration at time $m\tau_a$ differs from one at $(m-1)\tau_a$ only by the height of one lattice site, since in one time step, one atom is deposited. The probability of a configuration at time $m\tau_a$ is dependent only on the probabilities of configurations at $(m-1)\tau_a$ which differ from that at $m\tau_a$ by the height of one lattice site (decreased by one). The growth of the lattice is therefore a Markov process and one may write the Chapman-Kolmogorov equation (Gardiner 1985) for the probability of a given configuration \mathbf{H} :

$$P(\mathbf{H}; m) = \sum_{\mathbf{H}_i} W(\mathbf{H}|\mathbf{H}_i; m-1)P(\mathbf{H}_i; m-1). \quad (2)$$

In the above equation, $W(\mathbf{H}|\mathbf{H}_i; m-1)$ is the transition probability between configuration \mathbf{H}_i and configuration \mathbf{H} in one time step. The sum in (2) is over all possible configurations \mathbf{H}_i that may give rise to \mathbf{H} .

From the complete probability distribution of the surface it is possible to derive several useful quantities in terms of reduced probabilities. One of these is the one-site probability $p(h_{i,j}; m)$ that a site at $[i, j]$ will have height $h_{i,j}$ after m atoms have been deposited:

$$p(h_{i,j}; m) = \sum_{k=1}^{\sqrt{N}} \sum_{l=1}^{\sqrt{N}} \sum_{h_{k,l}=0}^{\infty} (1 - \delta_{k,i}\delta_{l,j})P(h_{k,l}, \hat{\mathbf{H}}; m). \quad (3)$$

When the probability of a site having height h is independent of position, the fractional coverage of layer h is given by $\sum_{n=h}^{\infty} p(n; m)$.

The two-site probability that two neighbouring sites at $[i, j]$ and $[i+1, j]$ will have heights $h_{i,j}$ and $h_{i+1,j}$ after m atoms have been deposited is:

$$p(h_{i,j}, h_{i+1,j}; m) = \sum_{k=1}^{\sqrt{N}} \sum_{l=1}^{\sqrt{N}} \sum_{h_{k,l}=0}^{\infty} (1 - \delta_{k,i}\delta_{l,j})P(h_{k,l}, h_{k+1,l}, \hat{\mathbf{H}}; m). \quad (4)$$

It is desirable to compare the morphology of the growing surface given by the theoretical model with that observed by temporal oscillations in the specular RHEED intensity during epitaxial growth. The step density of the growing surface has been demonstrated to be an excellent qualitative representation of RHEED intensity oscillations (Clarke and Vvedensky 1987a, b, c, 1988). The surface step density is calculated

as the number of steps formed parallel to the surface by neighbouring sites of different heights, and from (4) is given by

$$S(m) = \frac{1}{N} \sum_{i=1}^{\sqrt{N}} \sum_{j=1}^{\sqrt{N}} \sum_{h_{i,j}=0}^{\infty} \left(\sum_{\substack{h_{i+1,j}=0 \\ h_{i+1,j} \neq h_{i,j}}}^{\infty} p(h_{i,j}, h_{i+1,j}; m) + \sum_{\substack{h_{i,j+1}=0 \\ h_{i,j+1} \neq h_{i,j}}}^{\infty} p(h_{i,j}, h_{i,j+1}; m) \right). \quad (5)$$

In the following sections we will propose a simple model, the local perfect layer model, and calculate the probability distribution for the surface, the one- and two-site probabilities, and the surface step density using the above approach. However, first it is instructive to consider two extreme cases of the local perfect layer model, random deposition and perfect layer growth.

Random deposition. In this extreme of growth, atoms impinge randomly on the lattice sites and remain at the point of initial attachment. No diffusion of the deposited atoms to other sites is allowed. Thus, prior to completion of one layer, growth of higher layers may begin. This model corresponds roughly to low-temperature epitaxial growth, in which case newly deposited adatoms do not have sufficient mobility to migrate from their initial positions.

The Chapman-Kolmogorov equation for random deposition is

$$P_N^{(1)}(\mathbf{H}; m) = \frac{1}{N} \sum_{i=1}^{\sqrt{N}} \sum_{j=1}^{\sqrt{N}} P_N^{(1)}(h_{i,j} - 1, \hat{\mathbf{H}}; m - 1) \quad (6)$$

where the superscript (1) refers to random deposition and the subscript N refers to the total number of sites in the lattice. The solution to this equation for the initial condition $P_N^{(1)}(\mathbf{0}; 0) = 1$ is

$$P_N^{(1)}(\mathbf{H}; m) = \frac{m!}{N^m \prod_{i=1}^N (h_i)!} \delta_{m, \sum_{i=1}^N h_i}. \quad (7)$$

Using (3) and (7), we obtain an expression for the one-site probability of a site having height h :

$$p_N^{(1)}(h; m) = \begin{cases} \frac{m!(N-1)^{m-h}}{h!(m-h)!N^m} & \text{if } m \geq h \\ 0 & \text{if } m < h. \end{cases} \quad (8)$$

In order to calculate the surface step density, we need the two-site reduced probability, $p_N^{(1)}(h_{i,j}, h_{i+1,j}; m)$. Since the probability of two sites having heights $h_{i,j}$ and $h_{i+1,j}$ is the same in the x and y direction, we will henceforth reduce the two subscripts on h to one. The two-site probability is

$$p_N^{(1)}(h_i, h_{i+1}; m) = \begin{cases} \frac{m!(N-2)^{m-h_i-h_{i+1}}}{h_i!h_{i+1}!(m-h_i-h_{i+1})!N^m} & \text{if } m \geq h_i + h_{i+1} \\ 0 & \text{if } m < h_i + h_{i+1}. \end{cases} \quad (9)$$

Using (5) for the step density and (9) for the two-site probability we obtain

$$S_N^{(1)}(m) = 2 \left(1 - \sum_{h=0}^{m_2} \frac{m!(N-2)^{m-2h}}{(h!)^2(m-2h)!N^m} \right) \quad (10)$$

where $m_2 = m/2 - (m \bmod 2)/2$ is the largest integer less than or equal to $m/2$.

Perfect layer growth. In this extreme, an impinging atom must go into the highest unfilled layer. Thus growth proceeds as the sequential filling of successive monolayers. Only two heights are possible on the lattice at a given time, the height of the partially filled layer and the height of the completely filled layer exposed directly beneath it. This case corresponds approximately to growth at high temperatures, in which case newly deposited atoms have sufficient mobility that they are able to search for and fill depressions in the surface.

Due to the successive filling of monolayers characteristic of perfect layer growth, only the top layer will be incomplete and will have f filled sites where $f = 1 + [(h^{\text{tot}} - 1) \bmod (N)]$, and $h^{\text{tot}} = \sum_{i,j} h_{i,j}$ is the total number of atoms on the surface. The Chapman-Kolmogorov equation for the probability of a configuration \mathbf{H} is

$$P_N^{(2)}(\mathbf{H}; m) = \sum_{i=1}^{\sqrt{N}} \sum_{j=1}^{\sqrt{N}} \left(\frac{\delta_{h_{i,j}, q+1}}{N-f+1} \right) P_N^{(2)}(h_{i,j}-1, \hat{\mathbf{H}}; m-1) \quad (11)$$

where $q = [(m-1)/N] - (1/N)[(m-1) \bmod N]$. The solution to this equation for $P_N^{(2)}(\mathbf{0}; 0) = 1$ is

$$P_N^{(2)}(\mathbf{H}; m) = \frac{f!(N-f)!}{N!} \delta_{h^{\text{tot}}, m}. \quad (12)$$

For the one-site probability we obtain the simple result

$$p_N^{(2)}(h; m) = [1 - (1/N)(m \bmod N)] \delta_{h,j} + (1/N)(m \bmod N) \delta_{h-1,j} \quad (13)$$

where $j = (m/N) - (1/N)(m \bmod N)$. (Note that j and $j+1$ are the only two heights possible on the surface at time $m\tau_a$.)

The two-site probability for perfect layer growth is found to be

$$\begin{aligned} p_N^{(2)}(h_i, h_{i+1}; m) &= \left(\frac{(m \bmod N)(N - m \bmod N)}{N(N-1)} \right) (\delta_{h_i+1, h_{i+1}} \delta_{h_i, j} + \delta_{h_i-1, h_{i+1}} \delta_{h_i-1, j}) \\ &+ \left(\frac{(N - m \bmod N)(N - 1 - m \bmod N)}{N(N-1)} \right) \delta_{h_i, h_{i+1}} \delta_{h_i, j} \\ &+ \left(\frac{(m \bmod N)(m \bmod N - 1)}{N(N-1)} \right) \delta_{h_i, h_{i+1}} \delta_{h_i-1, j}. \end{aligned} \quad (14)$$

Substitution of the above expression into definition (5) of the surface step density yields

$$S_N^{(2)}(m) = \left(\frac{4(m \bmod N)(N - m \bmod N)}{N(N-1)} \right). \quad (15)$$

Local perfect layer growth. We consider a growth model intermediate between the two extreme cases of random deposition and perfect layer growth. In this model, the surface is divided into square subsections each containing n sites, with sides \sqrt{n} in length. (\sqrt{N} must be an integer multiple of \sqrt{n} .) There are $\alpha = N/n$ of these subsections. We specify that *within each subsection* perfect layer growth occurs. Thus, an atom impinging randomly on a subsection must go into the lowest unfilled layer *in that subsection*, and within each subsection only two heights may coexist at a given time. Although growth within each subsection is perfect, growth in different subsections is uncorrelated and proceeds at different rates. Note that for $n = 1$, $\alpha = N$, the model reduces to that of random deposition, while for $n = N$, $\alpha = 1$, the model becomes perfect layer growth on a lattice of N sites.

Since perfect layer growth occurs in each subsection, only the top layer of each subsection k ($k = 1, 2, \dots, \alpha$) is incomplete and will have f_k filled sites where $f_k = 1 + [(h_k^{\text{tot}} - 1) \bmod(n)]$ and $h_k^{\text{tot}} = \sum_{i=nk-n+1}^{nk} h_i$ is the total number of atoms in the subsection k . (Observe that sites are no longer indexed by i, j but for convenience are numbered by subsection so that $i = 1, \dots, n$ for atoms in subsections $1, \dots, n$ respectively.) The Chapman-Kolmogorov equation for the probability of a configuration \mathbf{H} is

$$P_{N,n}^{(3)}(\mathbf{H}; m) = \sum_{k=1}^{\alpha} \sum_{i=nk-n+1}^{nk} \left(\frac{\delta_{h_{i,j}, q+1}}{n - f_k + 1} \right) P_{N,n}^{(3)}(h_i - 1, \hat{\mathbf{H}}; m - 1) \quad (16)$$

where the first subscript, N , denotes the total number of sites on the substrate and the second subscript, n , is the total number of sites in a subsection, and

$$q_k = [(h_k^{\text{tot}} - 1)/n] - (1/n)[(h_k^{\text{tot}} - 1) \bmod n].$$

The solution to this equation with $P_{N,n}^{(3)}(\mathbf{0}; 0) = 1$ is

$$P_{N,n}^{(3)}(\mathbf{H}; m) = \frac{m!}{\alpha^m \prod_{j=1}^{\alpha} (h_j^{\text{tot}})!} \prod_{j=1}^{\alpha} \frac{(f_j)!(n - f_j)!}{n!} \delta_{m, \sum_{k=1}^{\alpha} h_k^{\text{tot}}}. \quad (17)$$

Recognising that the one-site probability $p_{N,n}^{(3)}(h_{i,j}; m)$ is the same for any location (i, j) on the lattice we derive

$$p_{N,n}^{(3)}(h; m) = \sum_{k=0}^m \frac{m!(\alpha - 1)^{m-k}}{k!(m - k)! \alpha^m} \left[\left(1 - \frac{k \bmod n}{n} \right) \delta_{h,j} + \left(\frac{k \bmod n}{n} \right) \delta_{h-1,j} \right] \quad (18)$$

where j is as defined as for (13).

Unlike the one-site probability which has no dependence on position of the site, the two-site probability $p_{N,n}^{(3)}(h_{i,j}, h_{i+1,j}; m)$ is *not* the same for any two neighbouring sites on the lattice. For two neighbouring sites both located in the *same* section where perfect layer growth occurs, the two-site probability will be different from when the neighbour sites belong to *different* subsections. We will denote the two-site probability for neighbours in the same lattice as $p_{N,n}^{(3a)}(h_{i,j}, h_{i+1,j}; m)$ and for neighbours in different subsections as $p_{N,n}^{(3b)}(h_{i,j}, h_{i+1,j}; m)$.

Neighbour sites at $[i, j]$ and $[i + 1, j]$ in the x direction (or $[i, j]$ and $[i, j + 1]$ in the y direction) are thus divided into two classes: neighbours belonging to the same subsection, and those belonging to two adjacent but different subsections. This is illustrated in figure 1 where sites within the shaded subsection are labelled 1 and those in adjacent subsections are labelled 2. Inspection of the diagram shows that for each subsection, there will be $2(n - \sqrt{n})$ interactions between sites at $[i, j]$ and $[i + 1, j]$ (or $[i, j]$ and $[i, j + 1]$ in the y direction) of the type 1:1, and $2\sqrt{n}$ of the type 1:2. The exception to this case is when $n = N$, in which case due to periodic boundary conditions there are no interactions of type 1:2, and therefore there are N interactions of type 1:1. To obtain the total number of each type of interaction for the entire lattice, it is necessary to multiply the above values for one subsection by $\alpha = N/n$, the number of subsections. For convenience we therefore define two functions giving to the number of interactions of each of the two types (normalised by division by N). Denoting $w_1(n)$ for 1:1 interactions and $w_2(n)$ for 1:2 interactions, then

$$w_1(n) = \begin{cases} 2(n - \sqrt{n})/n & \text{if } 1 \leq n < N \\ 1 & \text{if } n = N \end{cases} \quad (19)$$

$$w_2(n) = \begin{cases} 2\sqrt{n}/n & \text{if } 1 \leq n < N \\ 0 & \text{if } n = N. \end{cases}$$

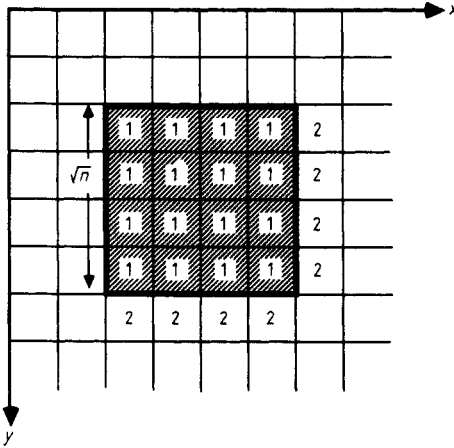


Figure 1. Two-dimensional lattice in local perfect layer growth illustrating interaction between sites in the same section and sites in adjacent but different subsections. The number of sites in the subsection for the above diagram is $n = 16$; the number of 1:1 interactions is $2(n - \sqrt{n}) = 24$; the number of 1:2 interactions is $2\sqrt{n} = 8$.

Thus we may write the expression for the surface step density (5) as

$$S_{N,n}^{(3)}(m) = \sum_{h_i=0}^{\infty} \sum_{\substack{h_{i+1}=0 \\ h_{i+1} \neq h_i}}^{\infty} \left(w_1(n) p^{(3a)}(h_i, h_{i+1}; m) + w_2(n) p^{(3b)}(h_i, h_{i+1}; m) \right) \quad (20)$$

where we have abbreviated the two subscripts on x denoting x, y position to one subscript, since the two-site probabilities are the same in the x or y direction.

The next step is to find the values of the two-site probabilities $p^{(3a)}(h_i, h_{i+1}; m)$ and $p^{(3b)}(h_i, h_{i+1}; m)$. First we focus on $p^{(3a)}(h_i, h_{i+1}; m)$. Since in this case h_i and h_{i+1} are in the same section, the two-site probability for a given number of atoms k falling into a section is simply $p_n^{(2)}(h_i, h_{i+1}; k)$, the solution for perfect layer growth on a lattice of n sites. Since $k = 0, 1, 2, \dots$ or m , we must average over all possible k , weighted by the probability that k atoms will fall into a subsection after a total of m atoms are deposited over the entire lattice:

$$p_n^{(3a)}(h_i, h_{i+1}; k) = \sum_{k=0}^m p_{\alpha}^{(1)}(k; m) p_n^{(2)}(h_i, h_{i+1}; k). \quad (21)$$

Secondly we must find the value of $p^{(3b)}(h_i, h_{i+1}; m)$. This is more complicated since h_i and h_{i+1} refer to sites in adjacent but different sections. After a total of m atoms are deposited, $k = 0, 1, 2, \dots$ or m atoms may fall into a certain subsection, and consequently $l = 0, 1, 2, \dots$ or $m - k$ atoms may fall into an adjacent subsection (given that k have dropped into the first subsection). Since growth in each of the two adjacent subsections is independent, the two-site probability of neighbouring sites having heights h_i and h_{i+1} is given as the *product* of the individual one-site probabilities for perfect layer growth of a site having height h_i (after k atoms have fallen in a section of n atoms) and of a site having height h_{i+1} (after l atoms have fallen in a section of n atoms). This product of perfect layer one-site probabilities must then be averaged over all possible k and l , weighted by the likelihood of k and l atoms falling in adjacent

subsections:

$$p^{(3b)}(h_i, h_{i+1}; m) = \sum_{k=0}^m p_{\alpha}^{(1)}(k; m) p_n^{(2)}(h_i, k) \sum_{l=0}^{m-k} p_{\alpha-1}^{(1)}(l; m-k) p_n^{(2)}(h_{i+1}, l). \quad (22)$$

The step density is now known entirely as a function of known solutions for the cases of random deposition and perfect layer growth. Substituting equations (8), (13), (21) and (22) into (20), we obtain the equation for the surface step density for local perfect layer growth:

$$\begin{aligned} S_n^{(3)}(m) = & w_1(n) \sum_{k=0}^m \frac{m!(\alpha-1)^{m-k}}{k!(m-k)! \alpha^m} \frac{2n(k_n)(1-k_n)}{(n-1)} \\ & + w_2(n) \left(1 - \sum_{k=0}^m \sum_{l=0}^{m-k} \frac{m!(\alpha-2)^{m-k-l}}{k!l!(m-k-l)! \alpha^m} \right. \\ & \left. \times \sum_{h=j}^{j+1} [(1-k_n)\delta_{n,j} + (k_n)\delta_{h-1,j}][1-l_n)\delta_{h,j} + (l_n)\delta_{h-1,j}] \right) \end{aligned} \quad (23)$$

where $k_n = (k \bmod n)/n$, $l_n = (l \bmod n)/n$, $j = k/n - k_n$, and $j' = l/n - l_n$. As n increases and the sections where perfect layer growth occurs become larger, the weighting of the first term in the equation increases relative to the other terms, and the solution increasingly resembles that for perfect layer growth, namely marked oscillations. As n decreases and the subsection become small, the contribution of the first term to the solution becomes progressively less, and the solution approaches the smoothly decaying curve characteristic of random deposition. The step density is plotted in figure 2 as a function of time and n .

Physically, the local perfect layer growth model corresponds approximately to a situation whereby an atom impinging upon the surface is allowed to sample an area of the lattice before coming to rest on the most stable site. In our model, an energetically favourable site is assumed to be one in the lowest unfilled layer, where the atom would tend to have more nearest neighbours and thus be more strongly bound than if it were perched atop a lone cluster. Once an atom is incorporated, however, no further movement is allowed. Homoepitaxial low-temperature growth of silicon and germanium exhibits damped oscillations in the RHEED intensity which show a marked resemblance to the curves in figure 2 (Aarts *et al* 1986). Since growth occurs at temperatures where diffusion is unlikely, it has been proposed (Clarke and Vvedensky 1988) that a newly arrived Si or Ge adatom forms a mobile precursor state which is able to sample a small portion of the surface before becoming incorporated into the lattice. Though greatly simplified, our model incorporates the two essential features of no diffusion and a mobile precursor state characteristic of Si and Ge growth at low temperatures.

In our analytical model, the origin of the damping of the oscillations is readily elucidated. At the beginning of growth, the large majority of the subsections reach monolayer completion at the same time, yielding a maximum in the step density (a *global* quantity) at half-layer completion and a minimum at layer completion. As growth proceeds, however, the rates of completion of monolayers in the different sections fall out of phase. This leads to a damping of the oscillations, with the degree of damping increasing as the size of a subsection, n , decreases. The effect of n on the calculated step density is due to the fact that growth in different subsections falls out of phase more quickly for small subsections than for large ones.

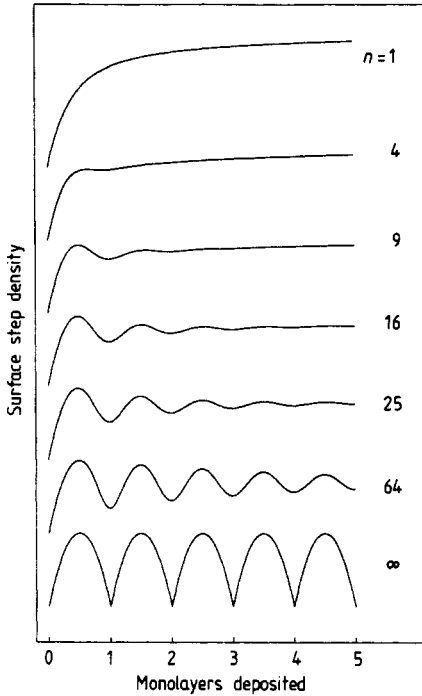


Figure 2. Surface step density for local perfect layer growth as a function of time for various values of n , the number of sites in a subsection. N , the total number of sites, is infinity.

In conclusion, modelling of epitaxial growth using the discrete Chapman-Kolmogorov equation allows us to derive quantities dependent on the microscopic disorder of the surface, such as the surface step density, which are unobtainable from knowledge of the coverage alone. Since we obtain the full height distribution function, such an approach would also be necessary in a first-principles calculation of RHEED intensities from a growing surface, which to date have been attempted only with artificial periodic configurations (Kawamura and Maksym 1985, Kawamura *et al* 1987).

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