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Continuum models of crystal growth from atomic beams with and without desorption

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Abstract. — Continuum equations appropriate to describe crystal growth from atom beams are derived in various cases. When desorption is important, the growth is described on very long lengthscales by the Kardar-Parisi-Zhang equation, but should be corrected for shorter lengthscales where surface diffusion is the dominant mechanism. In the absence of desorption, an important effect at sufficiently low temperature comes from the fact that diffusion of incoming atoms on the surface is anisotropic on long lengthscales because it is biased by reflexions against terrace edges. As a result, the growth is described by a pseudo-diffusion equation. In the case of a high symmetry surface, (001) or (111), an instability arises. Finally, in the absence of diffusion bias, the growth is described by a nonlinear equation of fourth order with respect to $\partial/\partial x$ and $\partial/\partial y$. The exponents are calculated in a Flory-type approximation. In particular the roughness exponent is found to be $\chi = (5 - d)/3$ in d dimensions.

1. Introduction.

1.1 MECHANISMS OF CRYSTAL GROWTH. — This article is intended to give a simple theoretical description of the macroscopic or mesoscopic aspects (beyond, say, 50 atomic distances) of crystal growth by atom beams. It is of interest to recall first an essential difference with growth from the fluid phase: the destabilising effect of heat or impurity diffusion through the fluid phase is absent, so that the corresponding instabilities (Mullins and Sekerka 1963, Langer 1980, Pelcé 1988, Viscek 1989) do not appear. The interface usually remains macroscopically planar if it is initially planar. However it becomes rough in the usual sense of statistical mechanics, to be recalled below. Let Z be the coordinate normal to the average surface (possibly different from the beam direction z), let X and Y be two rectangular coordinates parallel to the average surface, and let $\mathbf{R} = (X, Y)$ be a two-dimensional vector, or a $(d - 1)$ -dimensional vector in the general case of a d -dimensional space. For an initially planar surface, the height $Z(\mathbf{R}, t)$ will be assumed to be a uniform function of \mathbf{R} and t , but it has fluctuations due to the fluctuations of the beam. The roughness is characterized by the correlation function

$$G(\mathbf{R}, t) = \langle (Z(\mathbf{R}', t) - Z(\mathbf{R}' + \mathbf{R}, t))^2 \rangle . \quad (1.1)$$

The surface will be said to be rough if $G(\mathbf{R}, t)$ diverges when \mathbf{R} and t go to infinity. The main goal of the present work is to study this roughness.

Instead of (1.1), it would be possible to define a correlation function $\Gamma(\mathbf{R}, \tau; t)$ between the heights at distance \mathbf{R} in space and τ in time after an irradiation time t . This complication, however, is not extremely useful.

It is of interest to remark that instabilities which are not of the Mullins-Sekerka type can occur in growth by atomic beams, as seen in subsection 4.4.

1.2 VARIOUS MODES OF GROWTH BY ATOMIC BEAMS.

1.2.1 The oscillatory mode. — The best semiconducting devices or metallic multilayers are grown by molecular beam epitaxy (MBE) at fairly high temperatures, where diffusion is fast. In that case, when the surface is parallel to a high symmetry orientation, the roughness of the surface oscillates and exhibits minima (corresponding to the completion of the successive layers) separated by maxima (Fig. 1a). These oscillations are observed in reflection high energy electron diffraction (RHEED) or by other spectroscopic methods.

This growth mode will not be treated in the present work. — The models presented in the next sections are continuous, macroscopic models which are of no use to describe oscillatory growth. Presumably, these models would be in principle applicable on very large lengthscales, which would be unphysical.

We are conscious that most of experimentalists will be disappointed to see that growth in the oscillatory mode is not much studied in the present work. Our main excuse is that it is reasonable to study the simplest problems first. On the other hand, the other types of growth, addressed below, have also been experimentally investigated, and this suggests that their interest is not purely theoretical.

1.2.2 Stepped surface (« Step flow »). — From the theoretical point of view, the simplest case is that of a stepped (or vicinal) surface (Fig. 1b). Such a surface is prepared as a set of large terraces of high symmetry orientation, (001) or (111) in the case of a cubic crystal. These terraces are separated by straight, equidistant, parallel steps. When the beam is switched on, the steps go forward with an average velocity v , and consequently the crystal grows. The beam direction z will be assumed normal to terraces. If ℓ is the distance between steps, the rate of growth in the direction z is

$$\dot{z} = v/\ell. \quad (1.2)$$

1.2.3 High symmetry surface at moderate temperature. — The oscillating growth described in section 1.2.1 is observed only at high enough temperatures. On the other hand, at very low temperature the atoms have no possibility to move and to look for the lowest energy configuration, so that the resulting object is amorphous rather than crystalline. The present work will be focused on moderate temperatures, where surface diffusion is fast enough to allow the growth of a crystal, but the surface will remain appreciably rough (Fig. 1c) on lengthscales larger than a temperature-dependent limit which becomes microscopic at low temperatures. This growth mode might be of interest in the case of certain multilayers in order to avoid volume diffusion. Then a continuum description is reasonable on a timescale larger than the time τ_0 necessary to complete a layer.

The cases of interest in the present article are those of figure 1b and 1c, described in sections 1.2.2 and 1.2.3.

1.3 RANDOM EFFECTS, INHOMOGENEITY EFFECTS AND RECOVERY MECHANISMS. — The main purpose of this work is the investigation of the effects of random or systematic

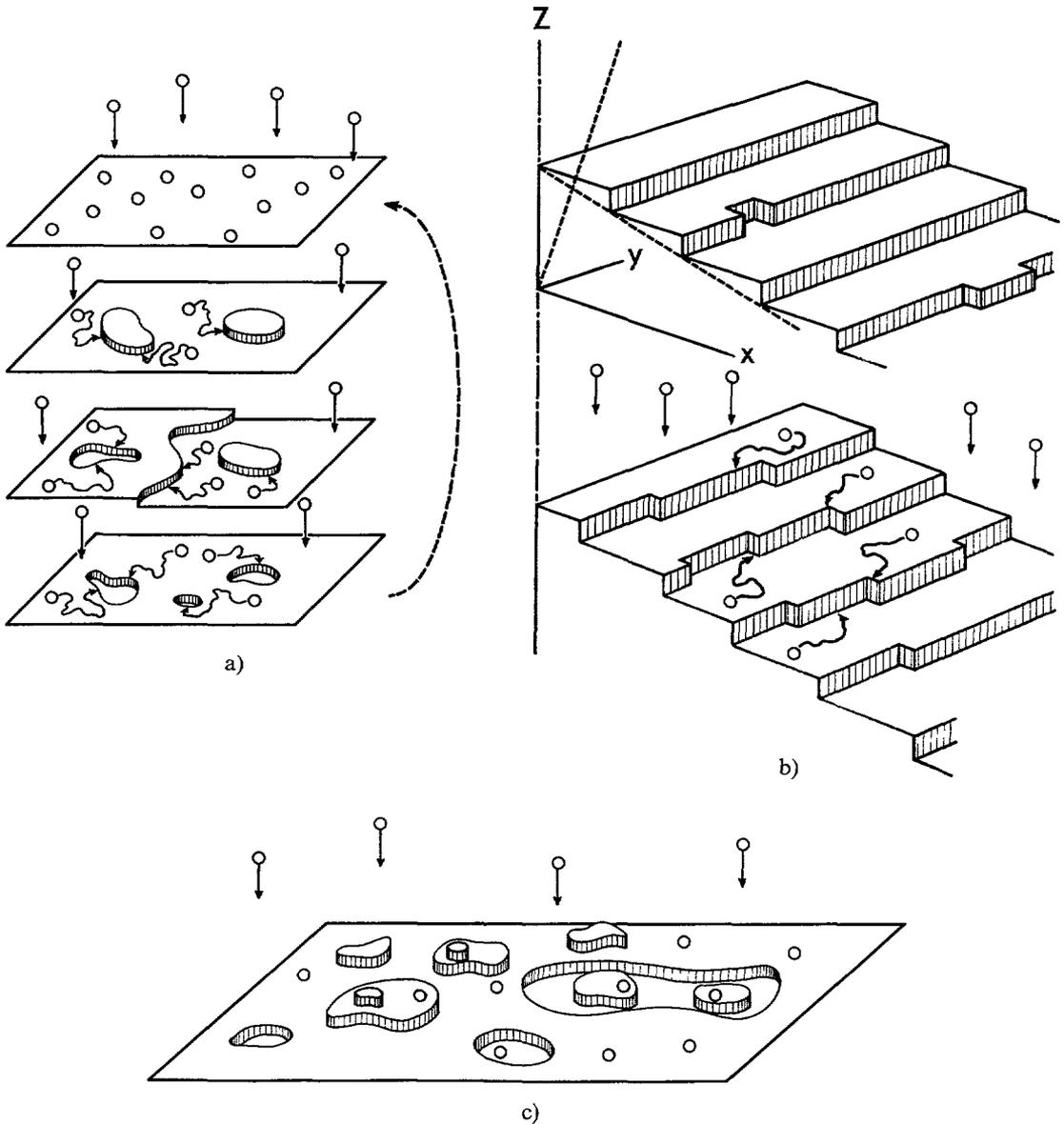


Fig. 1. — a) High symmetry surface of a crystal growing in an oscillatory way, giving rise to RHEED oscillations. This is the usual MBE procedure. b) A vicinal face of a crystal. α) At equilibrium. β) During growth. c) A high symmetry surface growing in a stationary regime. This regime can be reached when the surface diffusion constant is fast enough to ensure the formation of a crystal rather than an amorphous material, but sufficiently slow to avoid complete disappearance of small terraces when bigger ones begin to form.

fluctuations of the beam intensity $f(\mathbf{r}, t)$. It will generally be assumed that the beam is homogeneous apart from random fluctuations $\delta f(\mathbf{r}, t)$ which are uncorrelated in space and time. Thus, if $\mathbf{r} = (x, y)$ denotes the coordinates perpendicular to the beam direction z ,

$$\langle \delta f(\mathbf{r}, t) \delta f(\mathbf{r}', t') \rangle = \tau_0 \delta f^2 \delta_{\mathbf{r}, \mathbf{r}'} \delta(t - t') \quad (1.3)$$

where
$$\delta f \approx 1/\tau_0 = \bar{f} \quad (1.4)$$

is the instantaneous beam fluctuation, which on local scales approximately coincides with the average beam intensity \bar{f} . Indeed, in a time τ_0 , at a given site, an atom can land or not, with the same probability.

In the absence of atomic motion, the surface would become extremely rough and this would result, as noticed above, in amorphisation of the material. The recovery mechanisms which allow for the formation of a good crystal are a) surface diffusion, b) desorption (or « evaporation »), c) volume diffusion, d) formation of vacancies in the superficial layers, which are then incorporated in the bulk. The last two effects will be neglected. The effect (d) is the essential one in a model of Meakin *et al.* (1987) and would have an effect similar to desorption. Surface diffusion is the dominant effect except on long lengthscales at high enough temperature, as it is in the absence of a beam (Mullins 1963).

A short calculation will give the order of magnitude of the effect of fluctuations in the beam intensity for a layer of thickness h atoms and size $R \times R$, where R is measured in atomic distances. The average number of atoms deposited (on an initially flat substrate) is $R^2 h$, and its fluctuation is $\sqrt{R^2 h}$. The resulting fluctuation in h is therefore

$$\delta h \approx \sqrt{R^2 h}/R^2 = \sqrt{h}/R. \quad (1.5)$$

Let us assume that at some particular temperature, diffusion is able to heal the surface on a distance $R \approx 100$ interatomic distances. Then, for $h = 100$, the height fluctuation is $\delta h \approx 0.1$ atomic distances. This is not dramatically big, but it is not extremely small either. Therefore it is of interest to check whether diffusion and desorption are able to heal the surface. The present work is a step in this direction.

Most of the new results will be derived in section 4, where recovery is assumed to be due only to surface diffusion, and where novel models will be derived and solved. In section 3, recovery due to desorption will be studied and the system will be found to be described by the classical equation of Kardar, Parisi and Zhang (1986). Since the properties of this equation are well known, the only problem is to determine explicitly the coefficients. We have been able to do that only in the case of a stepped surface. In section 2 a linearized form of the kinetic equation is studied. It is an exactly soluble generalisation of a model of Edwards and Wilkinson (1982). The reader familiar with the topic might go directly to section 4. However, for tutorial purposes we find it appropriate to give, in the next two sections, some details on problems which are not novel, but which may require a great bibliographical effort from the non-specialist.

2. The linear approximation.

In certain cases the equations which describe growth turn out to be linear and can be solved. Such cases will be studied in subsections 4.4 and 4.6. It is of interest to recall first the equations which rule the smoothening dynamics of a surface in the absence of a beam, since these classical equations (which are linear in the case of weak fluctuations) are suggestive of the kind of equations we would like to have in the presence of a beam.

2.1 RECOVERY OF A MACROSCOPIC PROFILE IN THE ABSENCE OF A BEAM. — The problem to be addressed in this section is the following. The surface of a material is, on the average, planar, but has some macroscopic (e.g. sinusoidal) profile, resulting for instance from grooves having been digged on it — a classical experiment (Mullins 1957, 1959, Bonzel *et al.* 1984).

How will that surface go back to equilibrium? The average surface will be assumed to be orthogonal to a high symmetry axis z , also chosen as the beam direction.

2.1.1 *Evaporation dynamics* (Mullins 1959). — The chemical potential $\bar{\mu}$ of the vapor will be assumed uniform, so that the evaporation term in the kinetic equation has the form

$$\dot{z}_{\text{ev}}(\mathbf{r}, t) = B(\mu(\mathbf{r}, t) - \bar{\mu}) \quad (2.1)$$

where the local chemical potential $\mu(\mathbf{r}, t)$ on the surface is a function of the shape of the surface at time t in the neighbourhood of \mathbf{r} . The simplest assumption is that it is a function of the partial derivatives of z with respect to x and y , which can be expanded as a power series if the roughness is weak. This assumption can be argued to be correct, except in the case of a crystal surface below its roughening transition, but that case is not relevant in the frame of this article, the other sections of which deal with growing surfaces. A growing surface is, we believe, unavoidably rough. The expansion of μ cannot contain powers of the first derivatives, $(\partial_x z)^p (\partial_y z)^q$, because then (2.1) would imply that a crystal limited by a plane surface can grow or not according to the orientation of the plane. In reality growth or evaporation depends only on the sign of the difference between the chemical potentials. For the sake of simplicity, only the case of an isotropic surface will be considered, and then the leading term of the expansion is

$$\mu(\mathbf{r}, t) = \text{Const.} \times (z''_{xx} + z''_{yy}) = \text{Const.} \times \nabla^2 z(\mathbf{r}, t). \quad (2.2)$$

This is a particular form of the Gibbs-Thomson formula, appropriate for variations of weak amplitude. Insertion into (2.1) yields

$$\dot{z}_{\text{ev}}(\mathbf{r}, t) = \nu \nabla^2 z + \text{Const.} \quad (2.3)$$

with a positive value of ν .

2.1.2 *Surface diffusion* (Mullins 1959). — The part of \dot{z} which is due to surface diffusion obeys a continuity equation

$$\dot{z}_{\text{dif}} = - (j'_x + j'_y) = - \text{div } \mathbf{j}(\mathbf{r}, t) \quad (2.4)$$

where the current density $\mathbf{j} = (j_x, j_y)$ is a vector parallel to the average surface direction (not to the local surface). According to irreversible thermodynamics (Landau and Lifshitz, 1967).

$$\mathbf{j}(\mathbf{r}, t) = \text{Const.} \times \nabla \mu(\mathbf{r}, t). \quad (2.5)$$

Relations (2.2), (2.4), (2.5) yield

$$\dot{z}_{\text{dif}}(\mathbf{r}, t) = -K \nabla^2 (\nabla^2 z). \quad (2.6)$$

If both surface diffusion and desorption are present, both contributions (2.3) and (2.6) should be added. The result is a simple equation, which is universal in that sense that the microscopic details of desorption and diffusion are contained in the constants ν and K . Equations (2.3) and (2.6) suggest the kind of equations that most of theorists, fond of simple and universal models would like to have in the case of crystal growth.

2.2 CRYSTAL GROWTH IN THE LINEAR APPROXIMATION. — If the beam is switched on, the naivest guess is that the local rate of growth of the surface is the sum of (2.3), (2.6) and the

beam intensity $f(\mathbf{r}, t)$. It will be seen in section 3 that this is not generally true. However the resulting equation

$$\dot{z}(\mathbf{r}, t) = f(\mathbf{r}, t) + \nu \nabla^2 z - K \nabla^2 (\nabla^2 z) \quad (2.7)$$

turns out to be correct in certain cases (Sects. 4.3 and 4.6 below). In addition, the solution, to be given below, has certain qualitative features which are also present in the general case.

The solution of (2.7) is a slight generalisation of the treatment of Edwards and Wilkinson (1982) who treated the case $K = 0$. However, their article is rather difficult to read, and it is of interest to outline the method. This is done in appendix A. Here, only the results will be given. The beam direction z is assumed to coincide with the normal Z to the average surface, which is assumed to be initially flat, so that $z(\mathbf{r}, 0) = 0$.

2.2.1 *The limit $t \rightarrow \infty$.* — If $0 < \nu \ll K$, in 3 dimensions and for long distances,

$$\delta z(r)^2 = \langle [z(\mathbf{r}', t) - z(\mathbf{r}' + \mathbf{r}, t)]^2 \rangle \sim 2 \pi \tau_0 \frac{\delta f^2}{\nu} \ln (r \sqrt{\nu/K}). \quad (2.8)$$

K appears only in the logarithm and does not play an important part. The logarithmic dependence in r is the same as in the original Edwards-Wilkinson model ($K = 0$), and also as in a surface at equilibrium at some effective temperature. Thus the noise δf may be viewed as simulating thermal disorder. This way of thinking can, however, be misleading since the noise does not satisfy any detailed balance formula. Indeed, if $\nu = 0$, the K term is unable to evacuate all the energy and formula (2.8) should be replaced by

$$\delta z(r)^2 = \langle [z(\mathbf{r}', t) - z(\mathbf{r}' + \mathbf{r}, t)]^2 \rangle \approx \tau_0 \frac{\delta f^2}{K} r^2. \quad (2.9)$$

This formula is valid in 3 dimensions for $\nu = 0$, or for small ν and short lengths r . It predicts a roughness which is much stronger than thermal roughness. A numerical evaluation is appropriate to get convinced that this result is not in contradiction with the experimental realisation of good layers by MBE. According to (1.3), $\delta f \approx 1/\tau_0$, where τ_0 is the time necessary to complete one atomic layer. The current is given by $\mathbf{j} = D\rho \nabla\mu / T$, where ρ is the density of moving atoms, D their diffusion constant and μ the chemical potential. This yields $K \approx D\rho g / T$, where g is the surface tension. At high enough temperature, D should be of order $1/\tau_1$, where τ_1 is a typical phonon time. The order of magnitude of ρ is the maximum concentration possible without nucleation of terraces. This threshold does not depend very much on the growth rate (except if it becomes very slow) and is between 0.01 and 0.1. Thus, taking $g/T \approx 10$, a reasonable evaluation is $K \approx 10^{-10}$ and $\tau_0 \approx 1$ second. Then the height variation predicted by (2.9) is one atomic distance for two points of the surface distant of 10^5 atomic distances. In such a case the continuous approximation is not applicable and formula (2.9) is not reliable. However, the above numerical calculation shows that this unreliable formula is not in disagreement with the very smooth surfaces grown by MBE. When the temperature decreases, τ_1 increases drastically and the roughness becomes much stronger. Then the present article becomes of physical interest.

The case of a dimension $d \neq 3$ is also of interest. For $\nu \neq 0$ and $d < 3$ the result of the Edwards-Wilkinson model is recovered, namely

$$\delta z(r) \sim r^{-(3-d)/2} \quad (d < 3). \quad (2.10)$$

For $\nu = 0$ one finds

$$\delta z(r) \sim r^{-(5-d)/2} \quad (d < 5). \quad (2.11)$$

It follows from these formulae that, in all physically relevant cases ($d = 3$ and $d = 2$) the continuum, linear model (2.7) is consistent with our prejudice that a growing surface should be rough. This property will be found to hold even when nonlinear terms are taken into account.

2.2.2 *The limit $r = \infty$.* — For $0 < \nu \ll K$ it is shown in appendix A that

$$\langle [z(\mathbf{r}', t) - z(\mathbf{r}' + \infty, t)]^2 \rangle \sim \frac{\pi}{\nu} \tau_0 \delta f^2 \ln(\nu^2 t / K) \tag{2.12}$$

and for $\nu = 0$

$$\langle [z(\mathbf{r}', t) - z(\mathbf{r}' + \infty, t)]^2 \rangle \sim \tau_0 \delta f^2 (t/K)^{1/2} \tag{2.13}$$

The crossover from (2.8) to (2.12) takes place for $r \approx \xi$, with

$$\xi \approx \sqrt{\nu t} \quad (\nu \neq 0) \tag{2.14}$$

and the crossover from (2.9) to (2.13) occurs at

$$\xi \approx (K^3 t)^{1/4} \tag{2.15}$$

Following the notations of Kardar *et al.* (1986), we define two exponents z and χ by $\xi \sim t^{1/2}$ and $\delta z(r) \sim r^\chi$. Unfortunately χ is sometimes called α (Vicsek and Family 1985, Jullien and Botet 1985, Viscek 1989) or ζ . Comparison with formulae (2.12) to (2.15) yields the following values in $3 = 2 + 1$ dimensions.

	$\nu \neq 0$ (Edwards-Wilkinson model)	$\nu = 0$ (no desorption)
z	2	4
χ	0	1

Below 3 dimensions χ is seen from (2.11) to be larger than 1 in the absence of desorption. This implies that, below 3 dimensions, the model is not physically acceptable. However, mathematically, it is still consistent.

3. Growth with desorption.

In this section it will be seen that, in the presence of a beam, the linear equation (2.7) is generally not acceptable. The nonlinear terms can be explicitly calculated in the case of a stepped surface (Fig. 1b).

3.1 CASE OF A STEPPED SURFACE. — The following model will be assumed. An atom which has just landed on the surface diffuses until it finds a step or desorbs. If it reaches a step it does not play any role. The desorption probability obviously depends on the step density $|\nabla z| = 1/\ell$, where ℓ is the local distance between steps, and z is the coordinate perpendicular to terraces (while Z denotes the direction perpendicular to the average surface). The beam direction is assumed to be the high symmetry orientation z . For short ℓ , the atoms have no time to desorb and the growth rate is

$$\dot{z}(\mathbf{r}, t) = f(\mathbf{r}, t) + \text{diffusion terms} . \tag{3.1a}$$

On the other hand, if ℓ is large, all atoms desorb except those which land very close to a

step. Therefore, each step has a velocity $v = Cf$, where C is a constant. Relation (1.2) then yields

$$\dot{z}(\mathbf{r}, t) = (C/\ell) f(\mathbf{r}, t) + \text{diffusion terms} . \quad (3.1b)$$

It is easy to find the appropriate interpolation formula between (3.1a) and (3.1b). It is convenient to introduce a function of ℓ^{-2} rather than ℓ or $1/\ell$. The interpolation formula is

$$\dot{z}(\mathbf{r}, t) = f(\mathbf{r}, t) \varphi(\ell^{-2}) + \text{diffusion terms} . \quad (3.1c)$$

For the function φ , the form calculated by Burton, Cabrera and Frank (1951) in the absence of fluctuations can be used, namely

$$\varphi(1/\ell^2) = 2\ell^{-1} \sqrt{A/\eta} \tanh\left(\frac{\ell}{2} \sqrt{\eta/A}\right) , \quad (3.2)$$

where η is the evaporation rate and A the diffusion coefficient of freshly landed atoms (« adatoms »). The derivation of (3.2) will be briefly recalled. The density $\rho(x)$ of adatoms satisfies $\dot{\rho} = \bar{f} - \eta\rho + A\rho''$, and this is zero if the system is almost in a steady state. The solution is, with an appropriately chosen origin, $\rho(x) = \bar{f}/\eta - A \cosh \kappa x$, where $2\kappa = \sqrt{\eta/A}$. If the capture of adatoms by steps occurs with probability 1, A is determined by $\rho(\pm \ell/2) = 0$. The rate of capture by steps is $2A\bar{f}|\rho'|$. If this is identified with the step velocity, (3.2) results from the above formulae and (1.2).

Expanding φ in (3.1c) in a power series around the average orientation yields

$$\begin{aligned} \dot{z}(\mathbf{r}, t) &= f(\mathbf{r}, t) \varphi(z_x'^2 + z_y'^2) + \text{diffusion terms} \\ &= f(\mathbf{r}, t) \varphi_0 + f(\mathbf{r}, t) (z_x'^2 - \bar{z}_x'^2 + z_y'^2) \varphi_0' + \frac{1}{2} f(\mathbf{r}, t) (z_x'^2 - \bar{z}_x'^2 + z_y'^2)^2 \varphi_0'' . \end{aligned}$$

In this formula, y is the average step direction, $\bar{z}_x' = 1/\bar{\ell}$ is the average slope, and φ_0 , φ_0' and φ_0'' denote the values of φ and its derivatives for $z_x' = \bar{z}_x'$ which can easily be calculated from (3.2). Introducing the coordinate Z perpendicular to the average surface and neglecting the fluctuations of f except in the first term, the above equation yields

$$\dot{Z}(\mathbf{r}, t) = f(\mathbf{r}, t) \varphi_0 + cZ_x' + \frac{\lambda}{2} Z_x'^2 + \frac{\lambda'}{2} Z_y'^2 + \text{diffusion terms} ,$$

where

$$\lambda = 2\bar{f}(\varphi_0' + 2\bar{z}_x'^2 \varphi_0'') , \quad (3.3a)$$

$$\lambda' = 2\bar{f}\varphi_0' , \quad (3.3b)$$

and

$$c = 2\bar{f}\bar{z}_x' .$$

In the equation for \dot{Z} one can get rid of the constant part of the first term by a translation $Z \rightarrow Z - \varphi_0 t$, and of the second term by a Galilean transformation $x \rightarrow x - ct$. Finally

$$\dot{Z}(\mathbf{r}, t) = \delta f(\mathbf{r}, t) \varphi_0 + \frac{\lambda}{2} Z_x'^2 + \frac{\lambda'}{2} Z_y'^2 + \text{diffusion terms} . \quad (3.4a)$$

The actual beam intensity is renormalized by the factor φ_0 . This factor will be omitted in the following. This is possible if (as generally done) the beam intensity is not directly measured, but deduced from the layer completion time τ_0 through (1.4). On long

lengthscales, diffusion terms can be neglected. The above calculation is too crude because it assumes that atoms coming to a step do not play any role. In reality, they can still escape from the step and even from the surface, but these events follow the laws of thermodynamics and are described by equation (2.3). Combining (2.3) and (3.3) leads to an anisotropic form of the well-known « KPZ » equation

$$\dot{Z}(\mathbf{r}, t) = \delta f(\mathbf{r}, t) + \frac{\lambda}{2} (\nabla Z)^2 + \nu \nabla^2 Z \quad (3.4b)$$

introduced by Kardar, Parisi and Zhang (1986) to describe the long distance behaviour of the Eden (1958) model. The anisotropy in (3.3) will be neglected for simplicity from now on. Certain models different from the present one are also represented by (3.4) on long lengthscales. Certain of these models do exhibit desorption, since particles which land at certain places are not accepted. It is so in the model of Kim and Kosterlitz (1989), Amar and Family (1990) and in an exactly soluble, two-dimensional model of Meakin *et al.* (1986). In the Eden model the justification of (3.4) is completely different (Kardar *et al.* 1986). In the Edwards-Wilkinson model, $\lambda = 0$ and the ν -term is due to gravity. In other models described by (3.4), desorption is not allowed, but vacancy formation is allowed (Meakin *et al.* 1987, Pellegrini and Jullien 1990). When neither vacancy formation nor evaporation are possible, then λ should vanish as discussed in the next section.

Let the values (3.3) of λ and λ' be reconsidered. The derivatives of φ are taken with respect to $\xi = z_x'^2$. From (3.2) one deduces

$$\begin{aligned} \varphi(\xi) &= \sqrt{\xi/\kappa^2} \tanh \sqrt{\kappa^2/\kappa}, & \varphi'(\xi) &= \frac{\varphi(\xi)}{2\xi} - \frac{1}{2\xi} \frac{1}{\cosh^2 \kappa/\sqrt{\xi}} \\ \varphi'(\xi) + 2\xi\varphi''(\xi) &= -\frac{d}{d\xi} \frac{1}{\cosh^2 \kappa/\sqrt{\xi}} = -\frac{\kappa}{\xi\sqrt{\xi}} \frac{\sinh \kappa/\sqrt{\xi}}{\cosh^3 \kappa/\sqrt{\xi}}. \end{aligned}$$

Comparison with (3.3) shows that λ' vanishes for very large and very small ℓ . This can be directly checked from the asymptotic behaviour. More important, λ and λ' are seen to have different signs! The possible consequences of this effect is left for future work.

3.2 HIGH SYMMETRY SURFACE. — A growing crystal surface of high symmetry at moderate temperature (Fig. 1c) is expected to be rough. Similarly to the case of a surface at equilibrium at high temperature, we expect all properties to be analytic functions of the derivatives of the height z . If one writes that \dot{z} is an analytic function, equation (3.4) is obtained at lowest order. However, we have not been able to calculate explicitly λ .

A simple-minded argument would be the following: a non-vanishing value of ∇z indicates that steps are present. A step produces a disturbance of finite extension b . Therefore a non-vanishing value of ∇z should be reflected by a term of \dot{z} proportional to $b\sqrt{\ell}$. If this is (incorrectly) identified with $b|\nabla z|$, the result is in contradiction with (3.4). The flaw in the above argument is the following: equation (3.4) should hold only in the limit $\nabla z \rightarrow 0$ (this means: on very long lengthscales). Thus, $|\nabla z|$ should not be equal, but much smaller than $1/\ell$, where ℓ is the average distance between steps resulting from fluctuations. Since the numbers of up and down steps are on the average equal, a finite ℓ does not imply a nonvanishing $|\nabla z|$.

To summarize, the dynamics of a growing surface with a high symmetry orientation is presumably given by the KPZ equation for very long lengthscales, but the practical use of this equation might well be very limited. Further investigation would be necessary to precise this point.

3.3 PROPERTIES OF THE KARDAR-PARISI-ZHANG EQUATION (3.4). — They are reviewed in detail elsewhere by more competent specialists (Medina *et al.* 1989, Wolf 1990). However the non-specialist may be glad to find the summary which will be given here. \bar{f} will be assumed to vanish, since this condition can be fulfilled with the help of a translation $z \rightarrow z - \bar{f}t$.

a) The sign of λ is irrelevant since it can be changed by the transformation $z \rightarrow -z$, $\lambda \rightarrow -\lambda$ which leaves (3.4) invariant.

b) The nonlinear term in (3.4) is relevant, at least in 3 dimensions and below 3 dimensions. Indeed the ratio of the nonlinear to the linear (ν -) term in (3.4) on a lengthscale R scales as

$$\frac{(\nabla z)^2}{\nabla^2 z} \approx \frac{(\delta z(R)/R)^2}{\delta z(R)/R^2} = \delta z(R),$$

where $\delta z(R) = \sqrt{G(R, \infty)}$ (see formulae (1.1) and (2.9)). Since the surface is rough, $\delta z(R)$ goes to infinity with R . This argument suggests, in fact, that not only the nonlinear term is relevant, but the linear term is irrelevant.

c) A similar argument would show that all analytic terms which might be added to (3.4) and are consistent with translation invariance are irrelevant at long lengthscales. Translation invariance implies that Z can only appear through its derivatives.

d) The relaxation time τ of a bump or a hole of radius ξ and height h can indeed be calculated from the λ -term alone in the absence of beam, and is given by $h/\tau \approx h \approx \lambda (\dot{h}/\xi)^2$. It follows

$$\tau \approx \xi^2 / \lambda h. \quad (3.5)$$

e) Some insight into the relevance of the ν -term in (3.4) may be gained if one considers the decay (Fig. 2) of a sine profile $Z(x, y, 0) = a \cos qx$ under the effect of the deterministic version ($f = 0$) of (3.4). Assuming first $\nu = 0$, the surface becomes flat almost everywhere after a time given by (3.5), but there are still sharp peaks at the places where $\cos qx = 1$ (if $\lambda < 0$) or $\cos qx = -1$ (if $\lambda > 0$). These spikes disappear when the ν -term is switched on, but presumably a term of the form (2.6) would have the same effect. To summarize, the ν -term or a similar term is necessary (at least for $f = 0$) but the relaxation time (3.5) does not depend on it.

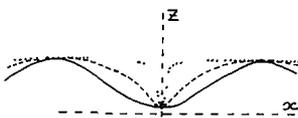


Fig. 2. — Relaxation of an initially sinusoidal profile (full curve) subject to equation (3.4) with $f = \nu = 0$. The sharp spike obtained at long time (dotted curve) would disappear for nonvanishing values of ν .

f) The exponents χ and z defined in section 2 should be such that the characteristic length ξ and height h associated to a time τ satisfy

$$\xi \sim \tau^{1/2} \quad (3.6a)$$

and

$$h \sim \tau^{\chi/z} \quad (3.6b)$$

Insertion into (3.5) yields

$$\chi + z = 2. \quad (3.7)$$

The use of (3.5) implies the non-obvious assumption that λ is not renormalized by the noise. The justification will be found in the literature, e.g. Medina *et al.* (1989).

g) The exponent χ in 3 dimensions (Forrest and Tang 1990) turns out to be

$$\chi \approx 0.385 . \quad (3.8)$$

Surprisingly, $\chi \neq 0$ although the argument (b) suggests the upper critical dimension to be 3. A discussion by Halpin-Healy (1989) casts some light on this point.

Let the physical relevance of the KPZ equation (3.4) in crystal growth be briefly discussed. Firstly, it clearly neglects higher order terms, in particular the K -term of (2.7). As said above (point c) these terms are negligible in the long lengthscale limit. If one wants to describe short lengthscales, it is necessary to reintroduce the K -term (2.6).

4. Growth without desorption.

At temperature where MBE is usually performed, desorption is generally negligible at equilibrium. Although this is not necessarily true in the presence of a beam, the limit of vanishing desorption is of physical interest.

4.1 ABSENCE OF λ -TERM IN THE GROWTH EQUATION. — In this section it will be assumed that all atoms coming from the beam are incorporated in the surface, and that the concentration of vacancies in the bulk is not influenced by the growth process and is just the equilibrium concentration at the temperature of the material. Under these circumstances, the rate of growth \dot{z} of a planar surface (if z is the beam direction, assumed to be also a high symmetry axis) should be independent of the orientation of the surface, therefore independent of ∇z . It follows, as noticed by Kariotis (1989) that the coefficient λ should vanish in (3.4). Another way to obtain this result is to write that the growth rate should satisfy a continuity equation (corrected by the beam intensity), namely

$$\dot{z}(\mathbf{r}, t) = f(\mathbf{r}, t) - \text{div } \mathbf{j}(\mathbf{r}, t) , \quad (4.1)$$

where the current density \mathbf{j} is perpendicular to z . The λ -term in (3.4) is not a divergence, and therefore $\lambda = 0$.

4.2 DIFFUSION BIAS. — The next question is whether the linear term of (3.4) also vanishes. In the absence of a beam, it does, and the dynamics are described by equation (2.6) where only higher order derivatives appear. This is a consequence of the detailed balance principle, which obliges the average current to vanish if $\nabla z = 0$. A non-vanishing current would indeed imply, through the detailed balance principle, that the particle energy depends on the height z , and this is excluded if gravity is neglected. However, in the presence of a beam, the average current does not vanish. The reason is the following. Assume $\nabla z \neq 0$, so that the surface has steps (Fig. 3). A number of experimental facts (Fink and Ehrlich 1986, Tsushiya *et al.* 1989) show that freshly landed atoms (which are not thermalized and therefore ignore the detailed balance principle) prefer to be incorporated to the « upper » step than to the lower one. The reason is that incorporation to the lower step would imply that the atom first jumps down to the lower terrace. To do that, the atom should jump over a potential barrier (Fig. 3). This can be understood from the fact that the number of neighbours is particularly small when the atom passes through the terrace edge. Experimentally, the effect is strong at room temperature in usual metals and semiconductors, but does not prevent the formation of smooth surfaces at around 600 K, so that the order of magnitude of the potential barrier can

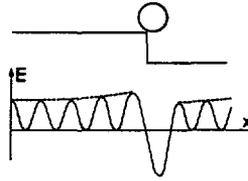


Fig. 3. — Potential seen (lower part) near a step (upper part) by an atom diffusing on a stepped surface. The potential has a maximum near the terrace edge because an atom at this position (circle) is at maximum distance from its remote neighbours. As a consequence, atoms go more easily to the upper step than to the lower step.

be estimated to 300 K. We propose to call the effect defined in this section ($j \neq 0$ if $\nabla z \neq 0$) « diffusion bias ». Its consequences are studied in the next two subsections.

The effect of diffusion bias can be tested by the following exercise. Consider (Fig. 4) a stepped surface with parallel steps separating *alternatively broad and narrow* terraces of two different lengths ℓ_1 and ℓ_2 . If there is diffusion bias, the upper edge of broad terraces proceed faster because they collect most of atoms falling into the broader terraces (see appendix B, Eq. (B1)). Thus the terrace widths equalize. This equalization does not occur in the absence of diffusion bias, as seen from equation (B1) with $\eta = 1/2$. Thus, diffusion bias accelerates the relaxation of a stepped surface to equilibrium. A more general study is presented below.

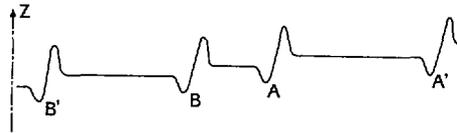


Fig. 4. — If diffusing atoms are strongly repelled by the lower step, this can stabilize a stepped surface. Indeed, if a narrow terrace AB is between two broader terraces AA' and BB', the steps B and A' collect most of the atoms landing on the broad terraces, and therefore go faster than A. Therefore the narrow terrace grows at the expense of the broad ones, and a flat surface is stabilized. The curve is a combination of the geometrical profile (Fig. 3a) and of the potential (dotted curve of Fig. 3b) which has no physical meaning.

4.3 CASE OF A STEPPED SURFACE. — In the case of stepped surface, the equation which describes growth on long lengthscales (longer than the step distance ℓ) can easily be derived in the case of infinitely high barrier. This approximation can be expected to be good at low enough temperature and if ℓ is not too large, otherwise the atoms would try a large number of times to jump the barrier, and would ultimately succeed with a high probability.

What is needed is an evaluation of the current density, to be inserted into (4.1). All landing atoms should go to the upper ledge, and it will be assumed that they do that in a time τ sufficiently short, so that the ledge does not move very much in that time. If the atom has landed at a distance x from the upper edge, its average velocity is x/τ . If the probability for an atom to have certain values of x and τ is $p(x, \tau)$, the average current density is

$$|\mathbf{j}| = \int_0^{\ell} dx \int_0^{\infty} d\tau p(x, \tau) (\bar{v}\tau) x/\tau .$$

The factor ($\bar{f}\tau$) is the average density of particles with a given τ . The beam intensity fluctuations are neglected in the second term of (4.1) which is now being calculated, although they should be taken into account in the *first* term. The integration over τ is readily done and one obtains

$$|\mathbf{j}| = \int_0^{\ell} dx p(x) \bar{f}x,$$

where $p(x) = 1/\ell$ is the probability that an atom has a given value of x . The final result is

$$|\mathbf{j}| = \bar{f}\ell/2 = \bar{f}/2 |\partial z/\partial X|,$$

where X denotes the coordinate perpendicular to the local orientation of the steps. Coming back to the coordinate x perpendicular to the average step orientation, the above formula yields

$$j_{\alpha} = \frac{z'_{\alpha} \bar{f}}{2(z'_x{}^2 + z'_y{}^2)} \quad (\alpha = x, y).$$

Insertion into (4.1) yields after linearization about the average slope ($\bar{z}'_x = 1/\ell$, $\bar{z}'_y = 0$).

$$\dot{z}(\mathbf{r}, t) = f(\mathbf{r}, t) + \nu \nabla^2 z(\mathbf{r}, t), \quad (4.2)$$

with

$$\nu = \ell^2 \bar{f}/2. \quad (4.3)$$

Thus, the linear model of section 2 is applicable and ν can be explicitly calculated. For short lengthscales, the term (2.6) should presumably be taken into account. The result is equation (2.7). In the absence of diffusion bias, there is no linear coupling between \mathbf{j} and ∇z , so that $\nu = 0$. This is in agreement with the finding of the last subsection that diffusion bias accelerates relaxation on a stepped surface.

We have discussed this section with various distinguished specialists, and we had some difficulty to convince them. The reason is presumably the following. The growth of a stepped surface corresponds to an forward motion of the steps. On the other hand the current density \mathbf{j} considered in this section is defined between steps and has no intuitive relation with steps. For that reason an alternative derivation of (4.2) is given in appendix C (in the one-dimensional case). That derivation considers step motion directly.

Deterministic version of (4.2). — If the surface is initially rough and if the beam fluctuations δf are negligible with respect to the initial disorder, f can be replaced by the constant \bar{f} in (4.2). Equation (4.2) now becomes deterministic, and can be used to study the evolution of the surface toward equilibrium.

As an application, a result of Gossmann *et al.* (1990) will be rederived. These authors consider an array of parallel steps, the distance of which has random fluctuations which are initially uncorrelated. They find that after a time t the probability distribution of the distances has a width proportional to $t^{-1/4}$ (or to the power $-1/4$ of the coverage, in their language). This will now be rederived from (4.2). A consequence of (4.2) is that the Fourier transform of $z(x)$, which can be defined with some precaution as explained in appendix B, satisfies, for $q \neq 0$,

$$z_q(t) = z_q(0) \exp(-\nu q^2 t).$$

The Fourier transform $\ell_q \simeq iqz_q$ of $\ell(x)$ satisfies the same equation and therefore

$$\langle |\ell_q(t)|^2 \rangle = \langle |\ell_q(0)|^2 \rangle \exp(-2\nu q^2 t).$$

Since the distances $\ell + \delta\ell_n$ between consecutive steps are uncorrelated at $t = 0$, it follows

$$\langle |\ell_q(0)|^2 \rangle \sim \sum_{n,n'} \langle \delta\ell_n \delta\ell_{n'} \rangle \exp[iq(x_n - x_{n'})] = \text{Const.}$$

In this formula we have ignored the fluctuations of x_n in the exponential. This approximation might be justified at the cost of some complications. The last two equations yield

$$\langle \delta\ell^2 \rangle \sim \sum_q \langle |\ell_q(t)|^2 \rangle = \sum_q \langle |\ell_q(0)|^2 \rangle \exp(-2\nu q^2 t) \sim \sum_q \exp(-2\nu q^2 t) \sim 1/\sqrt{t}.$$

The width of the distribution is therefore proportional to $t^{-1/4}$

4.4 HIGH SYMMETRY SURFACE. — In the case of a high symmetry surface, it is not so easy to take diffusion bias into account. Formula (4.2) would suggest an infinite current. This is in fact not true. The current density was found in subsection (4.3) to be proportional to the step distance ℓ because the atoms were supposed to diffuse to the upper ledge. This is not true if ℓ is too large because the density of free, diffusing atoms becomes so large that they form their own terraces as in figure 1c. It is reasonable to assume, as in section 3, that on a growing surface, all properties are analytic functions of the derivatives of z , as they are for a rough surface near equilibrium (Mullins 1963). In particular

$$\mathbf{j}(\mathbf{r}, t) = \alpha \bar{f} \nabla z(r, t)$$

where α is a constant and the factor \bar{f} recalls that the coefficient should vanish in the absence of beam. The point is that, according to the discussion of subsection 4.2, α should be positive. Then insertion into (4.1) yields equation (4.3) again, but with a negative value of ν ! A microscopic justification is given in appendix C, together with a rough evaluation of ν .

The negative value of ν implies an instability. Equations (2.7) and (3.4), derived in the previous sections, were consistent with a macroscopically planar (since $\chi < 1$), though microscopically rough surface. If $\nu < 0$, the surface cannot be macroscopically planar. To our knowledge no such instability has been reported, presumably because experimentalists just try to avoid it.

Since the surface of the growing crystal cannot be macroscopically planar, what is the actual surface shape? The answer requires further specification of the model. In a particular model treated in appendix D, a sawtooth profile is generated (Fig. 5).

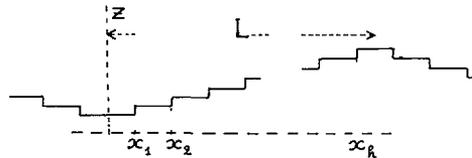


Fig. 5. — Steady state profile resulting of the growth of a surface having initially a periodic profile of weak amplitude.

4.5 THE CASE $\nu = 0$. — In practice, although diffusion bias is a well-known effect, it seems to be mainly present at rather low temperatures. The fact that it does not prevent the growth of very smooth surfaces at higher temperatures suggests that diffusion bias becomes weak, so that ν can be neglected.

The simplest idea (Bruinsma 1990, Bales *et al.* 1990) would be to use equation (2.7) with $\nu = 0$. However, since a nonlinear term is necessary in the case where desorption is possible, nonlinear terms can also be expected to be present in the absence of desorption. We have not been able to produce any real derivation, but the following argument may have some value. One would like to use equation (4.2) and an appropriate equation to replace (10) when no chemical potential can be defined. A reasonable Ersatz of (10) is $\mathbf{j}(\mathbf{r}, t) \sim \nabla \rho(\mathbf{r}, t)$, where ρ is the density of adatoms. Then it is reasonable to assume that ρ depends on the surface slope. For a highly symmetric orientation the lowest order expansion should be $\rho = \rho_0 + \rho_1(\nabla z)^2$. Combining both relations, one arrives at

$$\dot{z}(\mathbf{r}, t) = f(\mathbf{r}, t) - K \nabla^2(\nabla^2 z(\mathbf{r}, t)) + \sigma \nabla^2(\nabla z(\mathbf{r}, t))^2. \quad (4.4)$$

It turns out that, in this model, all exponents can be calculated from an argument à la Flory. Firstly a bump or a hole of radius R and height h is healed by the last two terms of (4.4) after a time τ which satisfies the relation

$$h/\tau \approx \dot{h} \approx (Kh + \sigma h^2)/R^4.$$

Hence

$$\tau \approx R^4/(K + \sigma h). \quad (4.5)$$

Another relation can be obtained by looking for the time τ necessary for the random noise f to make a bump or a hole of radius R and height h . The number of atoms landing into the area R^{d-1} (if d is the space dimension, usually 3) in the time τ is $\bar{f}R^{d-1}\tau/\tau_0$, and the fluctuation of this number, responsible for the formation of the bump or hole, is equal to its square root. It should also be equal to the volume $R^{d-1}h$ divided by the atomic volume (assumed to be one). This yields

$$\tau \approx (\tau_0/\bar{f}) R^{d-1} h^2 \quad (4.6)$$

Equating (4.5) and (4.6) yields

$$h^2(K + \sigma h) \approx (\bar{f}/\tau_0) R^{5-d} \quad (4.7)$$

It follows that the surface should be rough below 5 dimensions, since $\lim_{R \rightarrow \infty} h = \infty$. It follows also that the relaxation time does not depend on K for large lengthscales.

Inserting $h \sim R^\chi$ into (4.7) yields, if $\sigma \neq 0$,

$$\chi = (5 - d)/3. \quad (4.8)$$

In particular, for $d = 3$, $\chi = 2/3$. For $\sigma = 0$, (4.7) yields $\chi = (5 - d)/2$ in agreement with (2.11). Insertion of $h \sim t^{\chi/z}$ and $R \sim t^{1/z}$ into (4.5) yields

$$z = 4 - \chi = (7 + d)/3.$$

The above relations result from an argument à la Flory or à la Imry and Ma, which would fail in the case of the KPZ equation. More precisely, relation (4.6) cannot be applied to the

KPZ equation (3.4). It can be understood from the physical interpretation given in the present work, that (4.6) is correct in the case of surface diffusion dynamics and not in the case of desorption dynamics. Indeed, the correct way to handle the problem would be a coarse-graining procedure, in which an initial cell of linear size R would be divided in cells of size bR , then in cells of size $b^2 R$, etc. (with $b < 1$). The fluctuation of the atoms having landed in the initial cell and having remained in that cell is normally expected to depend on all scales. Equation (4.6) states that they do not. This is (presumably) correct in the case of surface diffusion because the atoms which land in a small cell of size $b^p R$ have no great probability to leave the big, initial cell. In the case of desorption dynamics, on the contrary, desorption is dominated by small cells, because the average value of $|\nabla Z|$ in (3.4) is bigger in small cells. Even though a more precise justification of (4.6) seems necessary, the present argument probably yields the correct answer.

On the other hand, equation (4.5) is not so safe as (3.5) was in the KPZ case (Nattermann 1990). There are two reasons for that. The first one is that the relative weight of short wavelength fluctuations in (4.4) is higher than in (3.4) because q^4 increases faster than q^2 . Therefore renormalisation due to large q values should be more effective. The second reason is that there is an exact proof of (3.5) in the KPZ case, based on Galilean invariance (Medina *et al.* 1989). If (4.6) is correct, but not (4.5), the following exact relation follows (Wolf and Villain 1990).

$$z - 2 \chi = d - 1 . \tag{4.9}$$

The argument à la Flory which leads to (4.8) is similar to the one which yields the equilibrium roughness of a domain wall (Villain 1982, Grinstein and Ma 1982) in the random field Ising model (RFIM). The result is the same. The analogy between the RFIM and growth problem was already remarked by Zhang (1988). The analogy between the Eden or KPZ growth and the random *exchange* Ising model is well known (Kardar *et al.* 1986). In the random field problem, formula (4.8) is believed to be exact. Whether it is exact in the present case is an open question.

An interesting consequence of the above Flory-like calculation in the case of equation (4.4) is the following. The effect of the nonlinear term is a *decrease* of the roughness, as seen from the comparison between (4.8) and (2.11). This is in contrast with the KPZ case, where comparison between (2.8) and (3.8) shows that roughness is increased.

4.6 SIMULATIONS IN TWO-DIMENSIONAL MODELS. — Although surface diffusion seems to be the dominant healing mechanism, there are not many numerical studies. Wolf and Villain (1990) studied a one dimensional model in which a freshly landing atom moves by one step (in the direction x of the average surface) if it increases the number of its neighbours, and then does not move any more (Fig. 6). A similar model has been devised by Das Sarma and Tamborenea (1990).

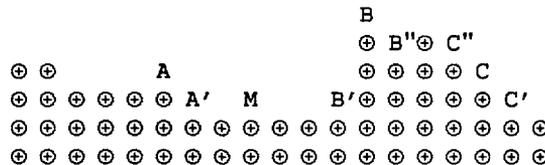


Fig. 6. — A one-dimensional model with no evaporation and no vacancy formation. Atoms landing at A', B', B'', C' or M do not move. Atoms landing at A should go to A'. An atom landing at B can choose between B' and B''.

The numerical results are in both models consistent with equation (35) with $\sigma = 0$. It is not quite clear why σ should vanish. Moreover there is no clear derivation of equation (35) in this case. The coefficient K cannot be obtained from the thermodynamical argument of section 2, since there is no Hamiltonian, no free energy and no chemical potential in the problem. It is interesting to see that thermodynamic-looking equations can occur in non-thermodynamical problems.

5. Conclusion.

The most important new results reported in this paper are related to the case when the smoothening dynamics is due to surface diffusion only. In that case the most striking results are i) a non-vanishing ν in the presence of diffusion bias (Sect. 4.3). ii) The instability which arises from diffusion bias for a high symmetry surface (Sect. 4.4). iii) The calculation of the roughness exponents in the absence of diffusion bias (Sect. 4.5) in an approximation à la Flory.

Another contribution, which we believe to be useful, is the actual calculation, for a stepped surface, of the coefficients λ and λ' (of opposite signs!) in equation (3.4), and of the coefficient ν of equation (4.2). The coefficient λ gives access to the knowledge of the surface roughness in the case of evaporation-dominated smoothening, while the coefficient ν contains the same information in the case of surface diffusion. Our understanding of high symmetry surfaces is poorer, in spite of the attempts reported in appendix C.

Since the coefficient ν can be positive or negative in the case of surface diffusion dynamics (Eq. (4.2)) there is a particular case where ν is exactly zero. This can occur for a stepped surface with a weak slope when temperature is varied: at lower temperature ν can be seen to become negative. This corresponds to the onset of an instability, rather than to a kind of roughening transition as in the case where λ is not generically zero, but vanishes accidentally (Yan *et al.* 1989, Amar and Family 1990, Pellegrini and Jullien 1990, Krug and Spohn 1990, Kim *et al.* 1990).

In the case of surfaces with a high symmetry orientation we have not been able to calculate the coefficients of the continuum equation (except for the rough evaluation of ν in appendix C). Moreover, the value of continuum equations in the case of highly symmetric orientations may be limited to unphysically large lengthscales. A general and realistic description of the growth of a surface of highly symmetric orientation is still an open problem.

Most of the results of the present paper (for instance exponent values) describe the effect of random beam fluctuations. However the kinetic equations (2.7), (3.4), (4.2), (4.4) can also describe, in the respective cases, systematic fluctuations in space, thus competing with the alternative continuum theory of Ohtsuka and Miyazawa (1988).

Acknowledgments.

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Appendix A.**Solution of the linear model (2.7).**

The Fourier transform of (2.7) is

$$\dot{z}_q(t) = f_q(t) - \alpha(q) z_q(t) \quad (\text{A1})$$

where

$$\alpha(q) = \nu q^2 + Kq^4 \quad (\text{A2})$$

Introducing the variable

$$\zeta_q(t) = z_q(t) e^{\alpha(q)t}, \quad (\text{A3a})$$

so that

$$z_q(t) = \zeta_q(t) e^{-\alpha(q)t}, \quad (\text{A3b})$$

equation (A1) becomes

$$\dot{\zeta}_q(t) = f_q(t) e^{\alpha(q)t}$$

This is readily integrated, yielding

$$\zeta_q(t) = \zeta_q(0) + \int_0^t dt' f_q(t') e^{\alpha(q)t'}$$

The correlation function is therefore

$$\langle \zeta_q(t) \zeta_{-q}(t') \rangle = \int_0^{t'} d\tau' \int_0^t d\tau \langle f_q(\tau') f_{-q}(\tau) \rangle e^{\alpha(q)(\tau'+\tau)} + \langle z_q(0) z_{-q}(0) \rangle.$$

Assuming $t' > t$ and using (1.3), the integral over τ' is readily performed :

$$\langle \zeta_q(t) \zeta_{-q}(t') \rangle = \tau_0 \int_0^t d\tau \delta f^2 e^{2\alpha(q)\tau} + \langle z_q(0) z_{-q}(0) \rangle.$$

An initially flat surface normal to the beam direction z will be assumed, so that the last term vanishes for $q \neq 0$. Coming back to the z variable, one finds

$$\langle z_q(t) z_{-q}(t') \rangle = \tau_0 e^{-\alpha(q)(t'-t)} \int_0^t d\tau \delta f^2 e^{-2\alpha(q)(t-\tau)} \quad (t' > t). \quad (\text{A4})$$

For large times, $t, t' \gg 1/\alpha(q)$, one finds for both signs of $(t' - t)$

$$\langle z_q(t) z_{-q}(t') \rangle = \tau_0 \frac{\langle \delta f^2 \rangle}{2(\nu q^2 + Kq^4)} e^{-(\nu q^2 + Kq^4)|t'-t|} \quad (\text{A5})$$

EQUAL TIME CORRELATIONS. — The case $t = t'$ is of special interest. Then (A4) reads

$$\langle z_q(t) z_{-q}(t) \rangle = \tau_0 \int_0^t d\tau \delta f^2 e^{-2\alpha(q)(t-\tau)} = \tau_0 \delta f^2 \frac{1 - \exp(-2(\nu q^2 + Kq^4)t)}{(\nu q^2 + Kq^4)} \quad (\text{A6})$$

The Fourier transformation can be inverted, yielding the results (2.8) to (2.11) when the limit $t = \infty$ is taken, and (2.12) and (2.13) in the limit $r = \infty$.

Appendix B.**Derivation of (4.2) in terms of step flow.**

In this appendix the one-dimensional version of (4.2) will be derived in an alternative way. It will be assumed that the velocity of the n -th step is

$$\dot{x}_n = -\bar{f}\eta(x_{n+1} - x_n) - \bar{f}(1 - \eta)(x_n - x_{n-1}). \quad (\text{B1})$$

The case $\eta = 0$ corresponds to maximum diffusion bias. The case $\eta = 1/2$ corresponds to no diffusion bias. The present approach is less general than that of section 4, because (B1) neglects retardation effects. It is convenient to introduce the Fourier component z_q of the height

$$z_q = \sum_{x=n_0}^{n_0+L} z(x) \exp(iqx). \quad (\text{B2})$$

This Fourier transformation is not very useful unless periodic conditions are artificially introduced, namely

$$x_{n+h} = x_n + L \quad (\text{B3})$$

where n and L are constants. The sum in (B2) depends on n_0 , but the Fourier transforms of $z'(x)$ and $\dot{z}(x)$ are independent of n_0 , provided q satisfies

$$q = 2\pi Q/L \quad (\text{B4})$$

where Q is an integer.

The derivative of (B2) is

$$\dot{z}_q = -\sum_n \dot{x}_n \exp(iqx_n). \quad (\text{B5})$$

Note that, in this equation, z has not the same meaning as in section 4. In equation (4.1), all atoms on the surface contribute to z . In (B2), the atoms which have not yet been incorporated in a step are ignored. Of course the continuum equation (4.1) makes sense only for long lengthscales.

Inserting (B1) into (B5) and making a Taylor expansion yields, to second order

$$\begin{aligned} \dot{z}_q &= \sum_n \left\{ \bar{f}\eta \left(x'_n + \frac{1}{2} x''_n \right) + \bar{f}(1 - \eta) \left(x'_n - \frac{1}{2} x''_n \right) \right\} \exp(iqx_n) \\ &= \bar{f} \sum_{n=1}^h x'_n \exp(iqx_n) - \bar{f} \left(\frac{1}{2} - \eta \right) \sum_{n=1}^h x''_n \exp(iqx_n), \end{aligned} \quad (\text{B6})$$

where x' and x'' denote the derivatives of x with respect to n or z . If the sum are replaced by integrals, the first term vanishes for $q \neq 0$ because

$$\int_0^h x' \exp(iqx) dz = \int_0^L dx \exp(iqx) = 0 \quad \text{if (B4) is satisfied.} \quad (\text{B7})$$

Thus (B6) reads for $q \neq 0$

$$\begin{aligned} \dot{z}_q &= -\bar{f} \left(\frac{1}{2} - \eta \right) \int_0^h x'' \exp(iqx) dz \\ &= -\bar{f} \left(\frac{1}{2} - \eta \right) \int_0^L x'' z' \exp(iqx) dx \\ &= -\bar{f} \left(\frac{1}{2} - \eta \right) \int_0^L \frac{\partial(1/z')}{\partial z} z' \exp(iqx) dx = -\bar{f} \left(\frac{1}{2} - \eta \right) \int_0^L \frac{\partial(1/z')}{\partial x} \exp(iqx) dx \end{aligned}$$

or

$$\dot{z}_q = \bar{f} \left(\frac{1}{2} - \eta \right) \int_0^L \frac{z''}{z'^2} \exp(iqx) dx .$$

Linearizing around the average value \bar{z}' yields

$$\dot{z}_q = \bar{f} \left(\frac{1}{2} - \eta \right) \bar{z}'^{-2} \int_0^L z''(x) \exp(iqx) dx \quad (\text{B8})$$

or

$$\dot{z}(x) = \bar{f} \left(\frac{1}{2} - \eta \right) \bar{z}'^{-2} z''(x) + \text{Const.}$$

This is the required equation. The constant (obviously equal to \bar{f}) is necessary because (B8) is not correct for $q = 0$. When the beam intensity fluctuation δf is added, the one-dimensional version of (4.2) is obtained, with

$$\nu = \bar{f}(1 - 2\eta) / \bar{z}'^2$$

We have taken the shortest way to the result. A few remarks are appropriate. Firstly, one can wonder whether the replacement of the first term of (B6) by an integral does not introduce an approximation which would be of the same order as the second term. To prove that it is not so, it is appropriate to consider the integral

$$\begin{aligned} \int_0^h x' \exp(iqx) dz &= \sum_{n=1}^h \int_{-1/2}^{1/2} d\xi x'(n + \xi) \exp[iqx(n + \xi)] \\ &\simeq \sum_{n=1}^h \int_{-1/2}^{1/2} d\xi [x'_n + \xi x''_n] \exp[iqx(n + \xi)] \\ &\simeq \sum_{n=1}^h \int_{-1/2}^{1/2} d\xi x'_n \exp[iq(x_n + x'_n \xi)] + \sum_{n=1}^h \int_{-1/2}^{1/2} d\xi \xi x''_n \exp iqx_n . \end{aligned}$$

This does reduce to the first term of (B6), which therefore vanishes according to (B7). Higher order terms in q have been neglected.

Another remark is that it is easy to make a mistake by inserting (B1) into (B2) and relabelling the x_n 's, if one forgets that $x_n \neq x_{n+h}$ is not periodic, in contrast with \dot{x}_n .

Finally, it can be remarked that the use Fourier transformation in the above argument was merely to provide a precise definition of the continuum approximation.

Appendix C.

Calculation of the negative ν for a high symmetry surface.

We consider a growing surface with an orientation of high symmetry, where steps are however present because of growth-induced fluctuations. Figure 7 gives an idea of this situation. For simplicity, the steps have been sketched as straight in order to distinguish them from smaller fluctuations which do not contribute to ∇z at the scale of interest. Moreover all steps present in the field of the figure have been assumed to be of the same sign — a somewhat misleading simplification. Thus, the surface is formed of terraces separated by ledges. On those terraces there are smaller, roughly circular terraces and holes. An atom which has just landed starts diffusing with zero average velocity (if long range, e.g. elastic forces are neglected) until it finds either a small terrace or a straight step. The only process which gives rise to a finite current is when the atom hits a straight step, and this happens only for atoms landing at a distance of a step which is not too much bigger than the distance a to small terraces. The proportion of such atoms is of order a/ℓ , where ℓ is the average distance between steps. In this fraction of the total area the local current density $j = \bar{f}a/2$, as seen from section 4.3 if ℓ is replaced by a . However this current is either in one direction or in the other. For a given value of ∇z , the probability that the local current is parallel to ∇z is of order $\ell |\nabla z|$. Finally the current density averaged in a region where ∇z has a given value is $\mathbf{j} \approx (a/\ell)(\bar{f}a/2)(\ell |\nabla z|) = \bar{f}a^2 \nabla z / 2$. Insertion into (4.1) yields (4.2) with

$$\nu \approx -a^2 \bar{f}.$$

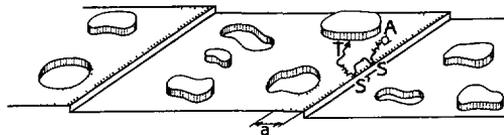


Fig. 7. — Schematic representation of a growing surface with a weak, but non-vanishing value of $|\nabla z| = 1/\ell$, where Z is orthogonal to a high symmetry orientation. To make the argument clearer, the surface is arbitrarily represented as an array of large terraces separated by straight steps at distance ℓ , with small terraces and small valleys inside large terraces. Most of the atoms which land on the surface go to small terraces and do not contribute to the current, except those which land on the dotted stripes of width a . Such an atom landing in A diffuses with zero average velocity until it reaches a step at S . At low temperature, it is then reflected with a high probability and diffuses again until it is caught by a smaller terrace at T .

This relation yields ν if a is known. In the calculation, barriers have been assumed completely opaque. At high temperature, where their transperance is high and a is large, each atom will try a large number of times to jump over the barrier, and the probability of success will be large. Thus, ν will decrease rapidly with increasing temperature, more rapidly than would be predicted by the Arrhenius exponential.

Appendix D.

The instability arising from diffusion bias.

The diffusion bias arising from the effect of figure 3 has been seen, in section 4.4, to give rise to an instability. When this instability takes place, the growing crystal is no longer limited by a planar surface. What happens then? Presumably, fluctuations play a very important role and the situation is unpredictable in most cases. However, we describe in this section a deterministic model in which a periodic profile is possible. This would be similar to cellular growth from the liquid phase (Pelcé 1988).

The surface (Fig. 5) will be assumed to be made of terraces separated by ledges parallel to the y direction. The initial conditions are assumed periodic with period $2L$. It will be assumed that no additional instability destroys this periodicity. The initial, periodic profile can be microscopic. We will argue that a macroscopic profile results.

The ledges present in the interval $(0, L)$ will be assumed to follow the equation (B1) with $\eta = 0$ (maximum diffusion bias).

$$\dot{x}_m = -\varepsilon(x_m - x_{m+1}) \quad (2 \leq m \leq h), \quad (\text{D1})$$

where h is the height. The appropriate form for $m = 1$ is obviously

$$\dot{x}_1 = -\varepsilon x_1 \quad (x_1 > 1/2). \quad (\text{D2})$$

The lowest terrace width can of course not be narrower than one atomic distance. It can be assumed, for instance, that x_1 vanishes abruptly when it reaches the value $1/2$.

Formula (D1) and (D2) are most reasonable. The difficulty is to describe the formation of new terraces at the top of the upper ones. Random beam fluctuations can presumably have a dramatic effect at this stage. We shall assume the following, deterministic process: a new terrace is formed whenever $(L - x_h)$ takes a certain value ℓ_0 . For the sake of simplification, it will be assumed that terrace formation at the top coincides with terrace death at the bottom, so that h is constant. This can only be true for certain, quantized values of h (fixed by ℓ_0 in the stationary regime) but this is no serious restriction if h is large enough.

It may look intuitive that the profile is rather linear. We are going to prove that. The solution of (D1) between two discontinuities may be written as

$$x_m(t) = \sum_{p=0}^{m-1} \frac{1}{p!} A_{m-p} \varepsilon^p t^p e^{-\varepsilon t} \quad (\text{D3})$$

Now we look for a solution which is stationary in this sense that it satisfies

$$x_m(\theta) = x_{m-1}(0) = A_{m-1} \quad (\text{D4})$$

where θ is the time which is necessary to fill a layer, so that

$$\theta = 1/\varepsilon. \quad (\text{D5})$$

Insertion of (D3) into (D4) yields

$$A_m = A_{m-1} e^{\varepsilon\theta} - \sum_{p=1}^{m-1} \frac{1}{p!} A_{m-p} \varepsilon^p t^p \quad (\text{D6})$$

This equation can be solved by introducing the Laplace transform

$$\varphi(q) = \sum_{m=1}^{\infty} A_m e^{-(m-1)q} \quad (\text{D7})$$

Insertion of (D6) yields

$$\begin{aligned} \varphi(q) &= A_1 + e^{\varepsilon\theta} \sum_{m=2}^{\infty} A_{m-1} e^{-(m-1)q} - \sum_{p=1}^{\infty} \frac{1}{p!} \sum_{m=p+1}^{\infty} A_{m-p} \varepsilon^p t^p \\ &= A_1 + \varphi(q) e^{\varepsilon\theta - q} - \sum_{p=1}^{\infty} \frac{1}{p!} e^{-pq} \varepsilon^p \theta^p \varphi(q) \\ &= A_1 + \varphi(q) e^{\varepsilon\theta - q} - \varphi(q) \exp(\varepsilon\theta e^{-q}) + \varphi(q). \end{aligned}$$

Thus

$$\varphi(q) = \frac{A_1}{\exp(\varepsilon\theta e^{-q}) - e^{\varepsilon\theta - q}} \quad (\text{D8})$$

This defines φ as a function of A_1 since θ is given by (D5). Inverting the Laplace transform yields the various A_m 's as functions of A_1 . Then the relation $L = x_h(0) = A_h$ yields h as a function of A_1 . Finally the relation $L - x_{h-1}(0) = L - A_{h-1} = \ell_0$ determines A_1 .

For small q , (D8) reduces to

$$\varphi(q) = \frac{2 A_1}{e q^2},$$

and this turns out to be Laplace transform of

$$x_m(0) = A_m = 2 A_1 m e^{-1} + \text{Const.} \quad (m \geq 1).$$

The profile is seen to have a sawtooth shape.

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