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Non-equilibrium roughening transitions

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Abstract. Non-equilibrium roughening transitions are described and shown to take place as a function of deposition rate and temperature. Discussion is given of the physical mechanism behind this effect, relating its appearance to the relative magnitudes of the vertical and horizontal length scales of surface configurations. The roughening length is defined, and several models are considered, one of which gives a finite transition temperature. Reference to recent experimental results is made and a phase diagram is discussed.

1. Introduction

In this paper we wish to discuss a class of what might be called non-equilibrium roughening transitions, which are of importance in the field of molecular beam epitaxy (MBE) (for a review see Madhukar (1983); for more recent work, Clarke and Vvedensky (1987), Das Sarma *et al* (1987) and Schneider *et al* (1987)). These transitions are to be distinguished from the more familiar equilibrium roughening transition as is discussed below (Weeks and Gilmer 1979). Some aspects of this problem have been considered previously by other authors (Kardar *et al* 1986, Nozieres and Gallet 1987). However, in view of recent experimental evidence that non-equilibrium roughening depends in a non-universal way on external parameters (for example on the deposition rate) we present here a different approach to the problem. To describe this phenomenon, consider a *two-dimensional* substrate with an external source depositing adatoms at a given rate R and let us define the interface width as being the number of incomplete layers at a given instant. This we will denote as Z . In the case (usually associated with zero temperature) that the adatoms arrive on the surface and stick to where they land it is easy to show (see below) that in the long-time limit

$$Z \sim t^x \tag{1}$$

(here we are following previous nomenclature of Rockett (1988) and Barnett and Rockett (1988)) where the $T = 0$ value is $x = \frac{1}{2}$. Generally deposition can be monitored in the laboratory by means of diffracted intensity oscillations (Ghaisas and Madhukar 1985, Singh *et al* 1986), and a simple interpretation of the observed results requires that the oscillations die out as the surface grows increasingly rough. The diffracted intensity in the out-of-phase condition is given in the kinematic approximation by

$$I(s_z) = \int d^2r_1 d^2r_2 \langle \exp[i s_z (\phi(r_1) - \phi(r_2))] \rangle. \tag{2}$$

$\phi(r)$ is the column height at lattice site r and s_z is evaluated at the out-of-phase condition ($s_z = \pi/d$, d is the lattice spacing). In many laboratory experiments, the observed behaviour is such that as $t \rightarrow \infty$, $I \rightarrow 0$. This will be our working definition of a *rough* interface.

As we will discuss later, there are cases where this condition is not met, but the surface is still regarded as rough; in some situations this distinction is one of experimental limitations (Lagally and Kariotis 1989). However, from a theoretical point of view the loss of diffracted intensity due to surface disorder is a result of competing length scales, laterally and vertically. In the literature (for example, Weeks *et al* 1976) the quantity that is of theoretical interest characterising the surface is the column-column height correlation function

$$G(|r_1 - r_2|) \equiv \langle [\phi(r_1) - \phi(r_2)]^2 \rangle. \quad (3)$$

which is the theorist's means of defining Z . For Gaussian noise (used to describe the random deposition of adatoms) the scattered intensity is a simple functional of $G(r)$. In special circumstances, it has been found that the diffracted oscillations can persist for rather long times and thus speculation can be made whether genuine steady-state oscillations are possible. In figure 1 we show a proposed phase diagram based on our rate equation calculations (Kariotis and Lagally 1989), suggesting the existence of two special critical temperatures. Below T_1 and above T_2 , $\alpha > 0$ and the interface width grows without bound, hence oscillations must die out. For $T_1 < T < T_2$, $\alpha = 0$ and the interface width is finite, and it is likely that oscillations persist indefinitely.

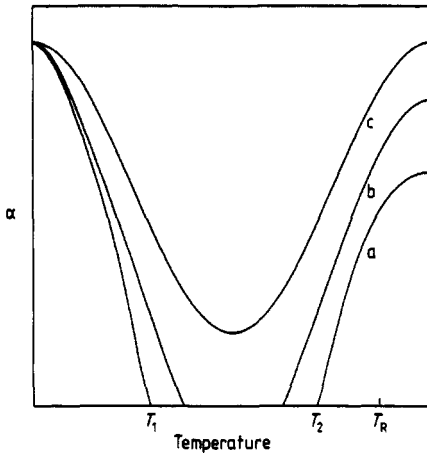


Figure 1. The growth exponent α for various rates of deposition. The curves marked a, b, c refer to increasing rates respectively. T_R is the thermodynamic roughening temperature.

In the following paragraphs we will describe a series of heuristic arguments and models, including reference to Monte Carlo simulation and recent experimental data, which indicate that true steady-state behaviour may be a realisable effect, at least in principle. In the next section we will discuss the physical criterion in order that there be a *smooth* interface, and then in §3 we will describe two limiting cases, one model which is never smooth and one which is never rough by our criterion. Finally in §4 we

will give evidence that the *binary model* can be either rough or smooth, depending on the values of the internal parameters.

An important conclusion of this paper is that it is not sufficient to state whether a surface is rough or smooth without giving a lateral length scale over which this property is supposed to appear. This 'roughening length' may be zero, finite or infinite. In the equilibrium case the interface is infinite above the roughening temperature T_R , that is, the interface width diverges logarithmically with column-column separation. This is *not* a sufficient criterion to result in total loss in diffracted intensity. In the kinematic approximation, the diffracted intensity has two parts, the *incoherent* part (of order N for a surface of N columns) and a *coherent* part (of order N^2 for a flat surface). The *incoherent* part is always present. It is the strength of the *coherent* part which is a measure of the surface roughness. It is apparent that there is a hierarchy of degrees of *roughness*; the simplest and most intuitive is that the surface derivative diverges (resulting in a complete absence of *coherent* intensity). This is the condition with which we will work in §4. However, this is a stronger condition than actually occurs in practice. Rather, there should be cases where $G(r)$ diverges over a finite lateral scale of the surface resulting in a partial loss of diffracted intensity. This statement of course is made with the supposition that the diffraction experiment be performed with an arbitrarily large coherence length. In practice, the coherence length is finite, and hence these effects may be observed due to the fact that the lateral scale is an experimental parameter, rather than a physical one. We have not been able to obtain anything other than an infinite or vanishing roughening length from the simple models here; however, the belief that it must also take on finite values is based on the observation that there is a lateral length scale in the problem which determines the diffusion distance over which atoms must be able to migrate in order that the interface be bounded. This is the property which we will now discuss.

2. Finite interface width criterion

In order to justify the following sections we will reproduce and expand upon an argument presented earlier (Kariotis and Lagally 1989). This involves a heuristic argument, followed by a quantitative example, describing the conditions under which bounded interface growth can occur. For sufficiently small deposition rate, there should generally exist an intermediate temperature range in which the interface width is finite. Below the roughening temperature any surface fluctuation must have a finite lifetime, and so if the deposition rate is lower than this decay rate, it should be possible to deposit without creating a rough surface. In order to see this, we propose the following construction. In figure 2 we show a typical interface configuration in which the vertical and horizontal length scales are Z and k respectively. In equilibrium, k is finite below T_R and we want to use this knowledge to determine the steady-state conditions under which Z is finite also. Now consider a particle at the top of the fluctuation, marked A in the figure. The criterion for a finite interface can be expressed in the following statement: the interface width will stop growing when the condition is met that a particle at A can diffuse to B in the time that it takes for an additional atom to be deposited at A.

To provide a quantitative basis for this statement, we will write a master equation

$$\dot{P}_l(t) = \sum_{\beta} W_{l\beta} P_{\beta} \quad (4)$$

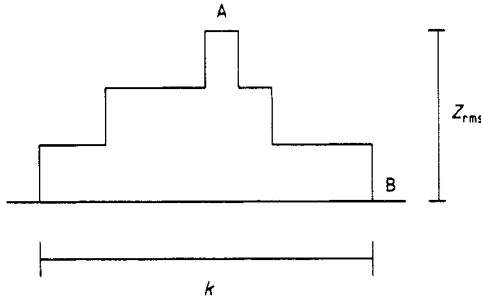


Figure 2. A typical configuration on the surface showing the vertical and horizontal fluctuation scales which characterise the resulting structure due to finite deposition.

describing a particle hopping on a lattice with a vertical and horizontal structure determined by the height and breadth of the surface fluctuation in figure 2. (In this problem the transition rates to the left and the right are different.) In particular, the transition rates along the surface, up the edge barrier, and down the edge barrier, respectively, are

$$W_s = \nu T \exp\left(-\frac{E_d}{T}\right) \quad W_u = \nu T \exp\left(-\frac{E_a}{T}\right) \quad W_d = \nu T \exp\left(-\frac{E_b}{T}\right) \quad (5)$$

where E_d , E_a and E_b are the hopping energies along the surface, onto an island, and off an island, respectively. Because there are $n \equiv k/Z$ sites over which the particle moves along the top of an island, for every jump that involves a change in height, the average transition rates up and down the incline are

$$\frac{1}{W_+} = \frac{1}{W_s} + \frac{1}{nW_u} \quad \frac{1}{W_-} = \frac{1}{W_s} + \frac{1}{nW_d} \quad (6)$$

since transition rates add inversely. We can solve this simply by taking the continuum limit, yielding a one-sided diffusion equation with an effective *diffusion* and *drift* coefficient respectively

$$D \equiv \frac{1}{2}(W_+ + W_-) \quad \Delta \equiv W_- - W_+ \quad (7)$$

The statement for the interface width to be finite now takes the form $k = \Delta/R$. That is, the particle must drift a distance k during the period before another atom is added. Rearranging the factors in this result, we have an expression for Z in terms of the horizontal correlation length, k , the binding energies of the adatoms, and the deposition rate

$$\frac{ZR}{W_s} = \left(\frac{k}{Z} + \frac{W_s}{W_d}\right)^{-1} - \left(\frac{k}{Z} + \frac{W_s}{W_u}\right)^{-1} \quad (8)$$

To complete this formulation, we will replace k with an expression for the thermodynamic correlation length, which below T_R is given by $k(T) \simeq \xi = A \exp[c(T_R - T)^{-1/2}]$. (Any function $k(T)$ will give the same qualitative result, provided $k = 0$ at $T = 0$ and $k > 0$ for $T > 0$.) Using this expression we can find, for fixed rate and fixed binding energies, two temperatures which bound regimes where the interface is rough at low T ,

smooth at intermediate T and rough again at high T . These conclusions are expressed graphically in figure 1. Above T_2 and below T_1 there is no solution to the equation, which we interpret to mean an interface growing arbitrarily rough. For $T_1 < T < T_2$ the equation provides a value of the steady-state interface width.

3. Two limiting models

3.1. Poisson growth

Now we wish to study specific models which might be used to check the results obtained in the previous section. Consider the case of $T = 0$ where the atoms are deposited at random, but at a fixed average R , and in each case, the atom sticks where it lands and does not move at all. This is the case of *Poisson statistics*. Each column grows independently of all others, and we can simply look at the rate of growth of a representative column. Call the height of the column above the surface ϕ , then the time rate of change of this height is

$$\frac{\partial \phi}{\partial t} \equiv \phi_t(x, t) = f(x, t) \tag{9}$$

where $f = 0, 1$ is a random function that is completely uncorrelated in time and space. The result is that the variable ϕ is a Poisson process, and is distributed such that at time t the probability of $\phi = n$ is

$$P(n, t) = \frac{(Rt)^n e^{-tR}}{n!} \tag{10}$$

where $\langle f \rangle = R$. The diffracted intensity at time t in the out-of-phase condition is

$$I(s_z = \pi/a) = \sum_{n=0} P(2n, t) - P((2n + 1), t) = [\cosh(Rt) - \sinh(Rt)]e^{-tR} \tag{11}$$

There are no oscillations and the diffracted intensity decays exponentially with time to zero.

3.2. Diffusive model

Next, consider a somewhat more physical, and also more familiar case (Edwards and Wilkinson 1982). Suppose that the deposition is given as a Gaussian process with zero mean (which implies that the column heights are given with respect to the moving interface) and atoms which land at a given column have a certain probability of dropping down to neighbouring columns if the neighbours are less tall. In the continuum limit this can be approximated by adding a diffusive term to the equation of motion given above for the column heights

$$\phi_t(x, t) = J \nabla^2 \phi(x, t) + f(x, t). \tag{12}$$

This equation describes the evolution of a surface above the x, y plane, and is easily solved in terms of the external driving force

$$\phi(x, t) = \int d^2x' dt' G_0(x - x', t - t') f(x', t') \tag{13}$$

where the Green function in two dimensions is

$$G_0(x - x', t) = \frac{1}{2tJ} \exp\left(-\frac{1}{2Jt}(x - x')^2\right) \theta(t). \quad (14)$$

The disorder is described by the Gaussian random variable f distributed by

$$P[f(x, t)] = \exp\left(-\frac{1}{2\sigma} \int d^2x dt f^2(x, t)\right). \quad (15)$$

Although this has somewhat different properties than the distribution used in the Poisson case, it suffices for our purposes here with the reminder that there is a constant term to the equation which represents the average deposition rate. The diffracted intensity is

$$I(s) = \int d^2r \langle \exp(is_z [\phi(r, t) - \phi(0, t)]) \rangle. \quad (16)$$

Performing the implied integration we have

$$I(s) = \int d^2r \exp\left(-\frac{s_z^2}{J} Q(r, t)\right) \quad (17)$$

where

$$Q(r, t) \equiv \int dx^2 dt [G_0(r, t) - G_0(0, t)]^2 \sim \log|r|. \quad (18)$$

as $t \rightarrow \infty$. Thus, in the long-time limit this leads to:

$$I(s) = L^2 L^{2(1-\eta)} \exp(a_1/t) \quad (19)$$

where $\eta = J^2\sigma$, $a_1 > 0$ is a constant and L is the linear dimension of the substrate. It is possible to show that I decays monotonically for all t to its final value; there are no intensity oscillations and I does not decay to zero.

3.3. Discussion

Quite a lot of work has been done previously on this problem and so before we go on to discuss the main contribution of this paper, we want to summarise the main points of the previous paragraphs. (See Weeks and Gilmer (1979) for a complete review up to that time; for more recent work, see Meakin *et al* (1986) and Nozieres and Gallet (1987).)

In general, we must make the distinction between an interface which can get rough over a finite lateral length scale and one which cannot. This is implicit in the arguments of §2: 'roughness' is a characteristic which implies a lateral length scale, depending on the physical properties of interest in a given experiment. In other words, there are different degrees of roughness, depending on the lateral scale over which the interface width diverges, or over which it is observed (i.e. the coherence length of the diffracted beam relative to the roughening length). In §3.1 we saw that in the *Poisson* model all columns are independent (there is no lateral motion of the deposited atoms) and the (coherent part of the) diffracted intensity is zero in the long-time limit. In the

diffusive model, the interface width is finite for all finite column separations on the surface, and the diffracted intensity is finite for all time (except in the special case of very large deposition). These models act as the two extreme cases: the *Poisson* model is always rough with zero roughening length (zero since even closest neighbours are uncorrelated); the *diffusive* model is never rough (by our definition) since correlation is never lost except for columns separated by an infinite distance. Note that the *diffusive* case has the form of the equilibrium behaviour, $\langle(\phi(r_1) - \phi(r_2))^2\rangle \sim \log(|r_1 - r_2|)$ which, by the usual definition, is referred to as an infinite interface. However, the diffracted intensity is not zero, regardless of the size of the substrate or coherence length.

In some previous work, equation (12) has been modified by the addition of a non-linear term $V = \lambda(\nabla\phi)^2$ in order to make the surface rougher. However, this term is not conservative; that is, it acts as a *source* term since it does not provide a mechanism for balanced transfer of particles between columns (gains of one column must be the losses of other neighbouring columns). This is easily seen by considering the one-dimensional equation

$$\frac{\partial h}{\partial t} = D \frac{\partial^2 h}{\partial x^2} + \left(\frac{\partial h}{\partial x}\right)^2 + \eta$$

where $h = h(x)$ is the column height at x , and η is the noise term. To see that the squared term generates particles on the surface even if the deposition is turned off, integrate over the surface

$$\frac{\partial}{\partial t} \int dx h = D \int dx \frac{\partial}{\partial x} \frac{\partial h}{\partial x} + \int dx \left(\frac{\partial h}{\partial x}\right)^2 + \int dx \eta.$$

The quantity $\int dx h(x)$ is the total amount deposited. If the rate is off, the time rate of change of this integral is zero. Integration of the ∂^2 term contributes only at the boundary, which is negligibly small for large substrates. The noise term averages to zero, and since the remaining term on the right is positive everywhere, then $\dot{h} \neq 0$.

In order to correct this we will consider a different model in the next section that has the desired conservative property, and that also is apparently rough or smooth depending on its internal parameters relative to the strength of the fluctuations in the deposition.

4. Binary model

The diffusive model does not exhibit a transition from finite to infinite interface width since lateral motion is always large enough to restrain the growth of neighbouring columns. We now consider a model which does appear to have this property. We start with a generalised version of equation (12)

$$\phi_t(x, t) = V[\phi(x, t)] + f(x, t) \tag{20}$$

and instead of the diffusive term used above, choose for the interaction a modified version called the *binary model*

$$V[\phi(r)] \equiv J_0 \sum_x \frac{1 - \exp(H_x)}{1 + \exp(H_x)} \tag{21}$$

where

$$H_x \equiv D_1(\phi(r) - \phi(r + x)) \tag{22}$$

or in the continuum limit

$$H_x \equiv D_1 \frac{\partial \phi(r)}{\partial x_x}.$$

The derivatives are summed over the different nearest neighbours indexed by α . In this model the columns exchange atoms at a *fixed* threshold rate as long as they are not of equal height (provided D_1 is sufficiently large, which for the moment we will assume to be the case). As required, motion of the adatoms depends only on column height differences, not on absolute heights. In this model, D_1 plays the role of the hopping rate, equivalent to those used in §2. This model is particularly appropriate to chemisorbed systems where, as shown in figures 3(a) and 3(b), an important mechanism of exchange between columns is that of diffusion down the side of the column. Since this proceeds at a finite and *constant* rate as long as the columns differ in height, the *binary* model should be qualitatively accurate. (It can be seen from this why the *diffusive* model is incorrect since the exchange rate between columns is proportional to the column height difference, and thus grows large if the height difference is large. This is enough to guarantee that the interface width is always bounded in the sense defined previously.) Also, as discussed at the end of the previous section, this model conserves particle number during the exchange between columns (Bruinsma and Aeppli 1984, Kardar *et al* 1986).

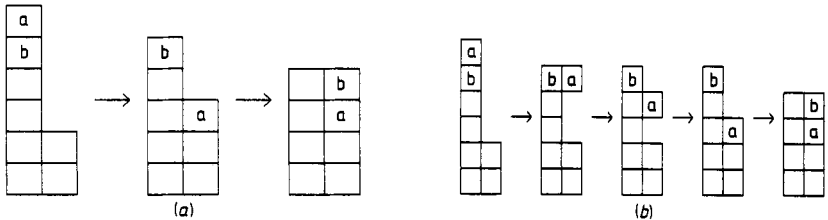


Figure 3. (a) Schematic of the *binary* model and comparison with a more physical picture. Both have a constant rate of exchange between columns as long as there is a difference in column height. The *diffusive* model has an exchange current, J , that is proportional to the column height difference, $J = -D_0(\phi_1 - \phi_0)$, and so is smooth by the criterion stated in the text. (b) Schematic of physical diffusion between neighbouring columns. In this case, if two columns differ in height, the mechanism which drives the change includes motion down the column side. The *binary* model is qualitatively the same.

The outline of this section is as follows. In §4.1 we show that a simple effective field argument at fixed deposition rate gives a transition temperature, T_1 of figure 1, at which a finite interface changes over into an infinite one. In §4.2, which is the main part of this paper, we obtain this result a second time using a more careful and systematic approach.

4.1. Effective field

The column growth equation is

$$\phi(r, t) = \int_0^t d\tau \{f(r, \tau) + V[\phi(r, \tau)]\}. \tag{23}$$

Ideally one would like to do perturbation theory with this in order to obtain the diffracted intensity $I(s_z)$. This would begin by expressing I in terms of the cumulants of the disorder

$$\langle \exp[is_z(\phi(r_1) - \phi(r_2))] \rangle = \exp(\Phi) \tag{24}$$

$$\Phi = is_z \langle \Delta \rangle - \frac{1}{2} s_z^2 [\langle \Delta^2 \rangle - \langle \Delta \rangle^2] + \dots \tag{25}$$

where we can write the general self-consistent solution to the random dynamics problem as

$$\phi(r, t) = \int_0^t d\tau (f(r, \tau) + ZV[\phi(r, \tau)]) \tag{26}$$

$$\phi(r, t) \equiv \phi_0 + \phi_1.$$

Z is the coordination number, and defining $\Delta\phi \equiv \phi(r_1) - \phi(r_2)$ the first few terms in the series are

$$\Phi = \langle (\Delta\phi_0)^2 \rangle + \langle (\Delta\phi_1)^2 \rangle + 2\langle \Delta\phi_0\Delta\phi_1 \rangle - \langle \Delta\phi_1 \rangle^2. \tag{27}$$

Perturbation theory is most easily performed when the non-quadratic part of the ‘Hamiltonian’ is a polynomial. Since we do not want to lose the saturation property of this model, some other approach is called for.

As an aside we note that the simple case of a two-site system, with column heights $\phi(x_1) \equiv \phi_1$ and ϕ_2 , and difference $\log(w) \equiv \phi_2 - \phi_1$ can be solved to give

$$w(t) = w_0(t) \left(1 + \gamma \int_0^t ds \frac{w(s)}{w_0(s)} \frac{1 - w(s)}{1 + w(s)} \right) \tag{28}$$

where $\gamma = J_0 D$ and $w_0(t) = \exp(\int_0^t ds f(s))$. To lowest order in γ this gives

$$w(t) = \exp(D^2\sigma t - \gamma t) + \mathcal{O}(\gamma^2) \tag{29}$$

which we will see gives the same transition as the effective field method.

Having obtained a qualitative idea of the two extreme modes of behaviour of the surface, we now wish to make explicit calculations, if possible. To this end, the first attempt here is to look at the average behaviour of the interface. The second and last terms in equation (27) cancel to lowest order in $\exp(-\sigma t D^2)$, since in this approximation

$$\langle V^2(r_1) \rangle = \langle V^2(r_2) \rangle = \langle V(r_1) \rangle^2 \tag{30}$$

leaving only the cross term. We can attempt to investigate the general behaviour of the remaining term with an effective field approach. In this context we can write the solution as

$$\int dt \langle fV[\phi(r)] \rangle = J_0 \int dt \frac{1 - \exp(\gamma t + \int dt' \langle fV \rangle)}{1 + \exp(\gamma t + \int dt' \langle fV \rangle)} \tag{31}$$

$\gamma = D_1^2\sigma$, or assuming saturation

$$\langle fV[\phi(r)] \rangle = J_0 \frac{1 - \exp(\gamma t - Zt \langle fV \rangle)}{1 + \exp(\gamma t - Zt \langle fV \rangle)}. \tag{32}$$

So when $ZJ_0 = \gamma$ the interface no longer grows indefinitely. This provides a ‘zeroth’-order solution and implies a transition temperature defined by

$$D_1^2 \sigma = ZJ_0 \equiv ZT_1 v \exp(-E_a/T_1). \tag{33}$$

Thus for low temperature, the surface grows with an unbounded interface width since the mechanism for moving particle from tall structures cannot keep up with the deposition. At higher temperatures, where the particle motion on the surface (both down a column and along lateral distances) is speeded up due to thermal motion, the interface width is inhibited from growing beyond a finite amount. Equation (33) defines T_1 of figure 1 in terms of the deposition rate, and has recently been observed for Ge deposited on Ge(110) (Chason *et al* 1989).

This presentation provides a semi-microscopic justification for the arguments given in §2. To proceed from here we must perform more careful calculations.

4.2. Saddle point

Because we assume that the distribution of $f(x,t)$ is known and that the relation between f and ϕ is given by equation (20), it is possible to study the nature of the resulting system by looking at its average behaviour. The change of variables can be done in the continuum limit without too much trouble

$$Df(x,t)P[f] = D\phi(x,t)\mathcal{J}(f;\phi)P[\phi] = D\phi(x,t)e^{-\mathcal{H}} \tag{34}$$

where the Jacobian of the transformation is written

$$\mathcal{J}(f;\phi) \equiv \text{Det} \left| \frac{\delta f(x,t)}{\delta \phi(y,\tau)} \right| \tag{35}$$

and the effective ‘action’ is given by

$$\mathcal{H} = \frac{1}{2\sigma} \int d^2x dt (\phi_t - V)^2 - \int d^2x dt \log(\mathcal{J}(f;\phi)) \equiv \frac{1}{2\sigma} H_1 - H_2. \tag{36}$$

To investigate the average behaviour of the system, we apply the method of steepest descent, or the ‘saddle point’ method. The saddle point equation is found by minimising the effective action with respect to the field variable ϕ and its derivatives, which in this case is

$$\frac{\delta \mathcal{H}}{\delta \phi} = \frac{\partial}{\partial t} \frac{\delta \mathcal{H}}{\delta \phi_t} + \frac{\partial}{\partial x} \frac{\delta \mathcal{H}}{\delta \phi_x} - \frac{\partial^2}{\partial x^2} \frac{\delta \mathcal{H}}{\delta \phi_{xx}} \tag{37}$$

where ϕ_x and ϕ_t respectively are the partial derivatives of $\phi(r,t)$ with respect to x and t . The full equation can be expressed in terms of functional derivatives of $\phi(x_1) \equiv \phi_1$ if H_1 and H_2 are properly symmetrised. Thus the full non-linear equation of motion for the saddle point can be written, before taking the continuum limit

$$\phi_{tt} = \frac{\delta H_1}{\delta \phi_1} - \sigma \frac{\delta H_2}{\delta \phi_1}. \tag{38}$$

These functions are easily evaluated as

$$\frac{\delta H_1}{\delta \phi_1} = V_1 [\cosh^{-2}(\varphi_{10}) + \cosh^{-2}(\varphi_{12})] - V_2 \cosh^{-2}(\varphi_{21}) - V_0 \cosh^{-2}(\varphi_{01}) \tag{39}$$

while the second term is

$$\frac{1}{J_0} \frac{\delta H_2}{\delta \varphi_1} = \tanh(\varphi_{10}) \cosh^{-2}(\varphi_{10}) + \tanh(\varphi_{12}) \cosh^{-2}(\varphi_{12}) \quad (40)$$

where $\varphi_{10} \equiv \frac{1}{2}D_1(\phi_1 - \phi_0)$. We have also abbreviated the discrete form of the column-column interaction as

$$V_j = J_0[\tanh(\varphi_{j,j+1}) + \tanh(\varphi_{j,j-1})]. \quad (41)$$

The full equation can be integrated numerically, but before we discuss the result of that effort, consider first the equation of motion in the linear limit, with the idea in mind of obtaining a more intuitive description of what to expect. H_2 can be approximated in the limit of small gradients which is essentially the non-linear version of the Onsager-Machlup theory (Graham 1973). After rearranging things, the quadratic part of the resulting 'action' has the form

$$H_1 - \sigma H_2 \simeq \int d^2x dt (\phi_t^2 + J_0^2 \phi_{xx}^2) - \sigma D_1^2 \frac{2}{3} \int d^2x dt (\phi_x^2 + \phi_{xx}^2). \quad (42)$$

In this approximation the interface obeys a linear equation of motion, the fluctuations about the mean are described by a plane wave

$$\phi \sim \exp(ikx - i\omega t). \quad (43)$$

If the interface is growing arbitrarily, then we expect this to be reflected in an instability of the dispersion relation $\omega(k)$ as determined by the saddle point equation. Assuming small gradients, the saddle point equation can be written in the form

$$\phi_{tt} = \sigma \phi_{xx} + D_2 \phi_{xxxx} \quad (44)$$

and where an effective 'diffusion' coefficient has been defined $D_2 \equiv J_0 D_1^2 (J_0 + \frac{2}{3}\sigma)$, and the resulting dispersion relation is

$$\omega^2 = \sigma k^2 - D_2 k^4. \quad (45)$$

Long-wavelength excitations are undamped travelling waves, but the important part of this expression is for large k , since it is the short-wavelength configurations which determine the degree of roughness on the surface. If $D_2 > 0$, large- k excitations take the form $\exp(ikx \pm \sqrt{D_2}k^2 t)$, characteristic of diffusive behaviour (damped excitations), and we conclude that the interface width is 'regular' since any rough configuration imposed by an external source (i.e. the deposition) will become smooth in time. But for k large the dispersion relation leads to $\exp(ikx \pm i\sqrt{|D_2|}k^2 t)$, and we say that the interface width is 'irregular' in the sense that it takes on whatever short-wavelength structures are imposed by the source—which we know to be random and uncorrelated, in other words, rough.

The above arguments for the quadratic 'action' have employed a number of approximations which could make the resulting conclusion quantitatively incorrect. However, we believe that the qualitative content of the calculation follows through. We have tried a numerical solution to the full saddle point equation, and obtained a result that is essentially in agreement with the above conclusion. In the interest of computer resource

conservation, we used only the one-dimensional version with a random set of initial conditions, but the qualitative behaviour was unambiguous: fluctuations are *diffusive* when $\frac{3}{2}\sigma \leq J_0$ and *rough* for $\frac{3}{2}\sigma \geq J_0$. The interpretation of these two properties in terms of the saddle point equation is that when the system is *rough* the structure simply propagates, as would be expected of a dispersive, but non-dissipative medium. When it is *diffusive* the initial conditions decay exponentially (although part of the actual numerical calculation will grow exponentially, characteristic of the two solutions $\omega = \pm i\sqrt{D_2}k^2$). The behaviours are distinct and easily recognised, but the distinction is not sharply defined quantitatively, since we were unable to run the computation long enough to check the transition. Future work in this area is obviously required.

Thus we conclude that for sufficiently small fluctuations in the deposition rate, σ , the surface should grow smoothly. As the temperature is lowered (or the rate is increased), there will appear a transition to a roughly growing surface. The transition temperature is a function of the deposition rate, and increases as the rate increases. In reference to figure 1, we have derived an expression for T_1 only. T_2 remains unaccounted for.

5. Discussion and conclusion

In figure 4(a) we show a Monte Carlo evaluation of the ASOS model (Weeks and Gilmer 1979). In this example we have made several runs which indicate that the intensity oscillations at high temperature are strongly damped (curve k), that as the temperature is lowered the oscillations first become less damped (curves j-b) and then become completely damped again at $T = 0$ (curve a), suggesting behaviour qualitatively similar to that shown in figure 1. It is common knowledge that a surface which grows roughly at low temperature will grow smoothly at higher temperature. Recently, experimental evidence has been found (Flynn *et al* 1988, Chason *et al* 1988) that this change in behaviour may be localised at a sharply defined temperature (though to our knowledge nothing is known yet about the rate dependence of this temperature). In figure 4(b) we show Monte Carlo evidence that the binary model has a finite roughening length. In curves a-g, evaluated at increasing values of J_0 , the amount of scattered intensity (at large t) increases, first from exactly zero, to a finite amount at around $J_0 \simeq 0.5$, and then quickly increases with increasing J_0 .

The main conclusion of this paper is that in the process of interface growth, the surface may grow with a finite interface width, or with an unbounded interface width, depending on the strength of the deposition rate relative to the rate at which atoms can move laterally on the surface. Essentially what we have shown is that there are two length scales, one lateral, determined by the equilibrium properties of the surface, and one vertical determined by the external deposition rate. Whether or not diffracted intensity is lost depends on the relative magnitudes of these two scales. A different way of expressing this condition is to say that there are two timescales involved, one determined by the rate of decay of fluctuations on the surface (an equilibrium property), and the second fixed by the deposition rate (a dynamic property). Related to this is the observation that it is necessary to state both a lateral, as well as a vertical, length scale when describing a surface as 'rough' (Lagally and Kariotis 1989). The models discussed were used to demonstrate these conclusions. In particular, the *binary* model was shown to have the property of describing either a rough or smooth interface, depending on the magnitude of the parameter J_0 relative to the size of the fluctuations

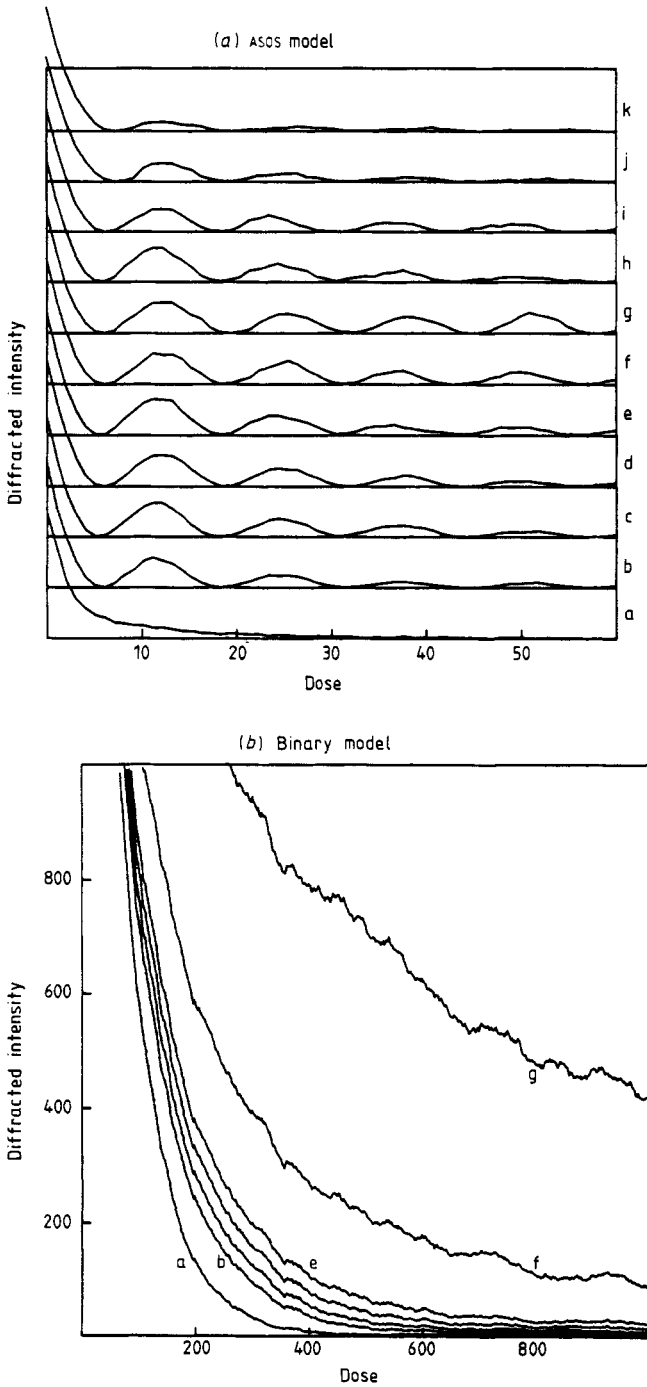


Figure 4. (a) Monte Carlo data for the ASOS model showing the diffracted intensity oscillations for eleven different temperatures. Curves are shown for various values of T : a: $T = 0.0$, b: $T = 0.2$, c: $T = 0.4$, d: $T = 0.6$, e: $T = 1.0$, f: $T = 1.2$, g: $T = 1.4$, h: $T = 1.6$, i: $T = 2.0$, j: $T = 4.0$, k: $T = 6.0$. (b) Monte Carlo data for the binary model showing the decay of diffracted intensity for seven different values of the column coupling constant, J_0 : a: $J_0 = 0.00$, b: $J_0 = 0.03$, c: $J_0 = 0.04$, d: $J_0 = 0.05$, e: $J_0 = 0.06$, f: $J_0 = 0.10$, g: $J_0 = 0.20$.

in the deposition. As far as we can tell, this model does not yield a finite roughening length.

While this work was in progress, numerical work appeared in print presenting some of the material which we have analysed in §4 (Karunasiri *et al* 1989). The interaction term used by these authors is qualitatively the same as that in the binary model, and produces the expected change over from rough to smooth growth, depending on temperature and deposition rate.

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