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Growth with Surface Diffusion.

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Abstract. – A simple growth model is investigated where particles are deposited onto a substrate randomly and subsequently relax into a position nearby where the binding is strongest. In space dimension $d = 2$ the surface roughness exponent and the dynamical exponent are $\zeta = 1.4 \pm 0.1$ and $z = 3.8 \pm 0.5$. These values are larger than for previous models of sedimentation or ballistic deposition and are surprisingly close to the ones obtained from a linear generalized Langevin equation for growth with surface diffusion. A scaling relation $2\zeta = z - d + 1$ is proposed to be valid for a large class of growth models relevant for molecular beam epitaxy.

Kinetic roughening [1, 2] has attracted a lot of attention over the last few years not only because of its practical importance for the growth of solid films, but also as an example for a dynamical mechanism that drives a system into a spatially and temporally scale invariant state («self-organized criticality» [3]). Most of the models of kinetic roughening studied so far can be described by the Kardar-Parisi-Zhang (KPZ) equation [4]

$$\partial h/\partial t = \lambda_0 + \lambda_1(\nabla h)^2 + \nu \nabla^2 h + \eta\,, \tag{1}$$

where $h(\boldsymbol{x}, t)$ is the height of the surface at time t above the substrate site specified by the d'-dimensional vector $\boldsymbol{x}$. $d' = d - 1$ denotes the surface dimension. The two λ-terms determine the average growth velocity which may depend on the tilt of the surface. ν has a smoothening effect on the surface, while the white noise $\eta(\boldsymbol{x}, t)$ due to fluctuations in the growth rate makes the surface rough.

In this note growth processes which *cannot* be described by (1) will be studied. Indeed (1) is not adequate for describing growth by molecular beam deposition if desorption and the formation of defects in the film may be ignored [5-7]. Then one should have

$$\partial h/\partial t = \lambda_0 + \eta - \nabla \boldsymbol{j}\,, \tag{2}$$

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where $\nabla \boldsymbol{j}$ is the divergence of the d'-dimensional surface current parallel to the substrate. This rules out the λ_1-term in (1).

From $\lambda_1 = 0$, eq. (1) reduces to the sedimentation model first studied by Edwards and Wilkinson [8]. In this model, λ_0 is the deposition rate of particles that fall down at constant $\boldsymbol{x}$ until they meet the surface atom at height $h(\boldsymbol{x}, t)$. If there is no neighbouring column with smaller h the incoming particle sticks, *i.e.* $h(\boldsymbol{x})$ is increased by 1. Otherwise it relaxes to the lowest site in a given neighbourhood thereby lowering its potential energy in a gravitational field. In this case the Laplacian term in (1) has a simple physical meaning. It describes mass conservation during the relaxation process, since it may be written as $-\nabla \boldsymbol{j}$ with the downhill current

$$\boldsymbol{j} = -\nu \nabla h . \tag{3}$$

The driving force for this current is the potential difference in the gravitational field.

For vapour deposition of solid films gravitational forces are so minute compared to the binding at the surface that they can be neglected. Nevertheless the Laplacian term may be present, *e.g.*, due to desorption [9]. Even if no particles can leave the surface, a current of kind (3) would be expected, if the momentum component parallel to the surface is not immediately thermalized when the particles touch the surface. Other mechanisms have been conceived which also lead to a Laplacian term in (1), however with a negative ν [7, 10]. (Formally this would imply that any periodic perturbation of an initially horizontal surface grows exponentially if higher-order terms are not taken into account.) If by contrast one assumes that the surface current is driven by differences in the surface chemical potential μ (as in the absence of deposition), $\boldsymbol{j} \propto -\nabla \mu$, and that μ is proportional to the surface curvature $\nabla^2 h$, one has to replace (3) by [5, 7, 9]

$$\boldsymbol{j} = K \nabla (\nabla^2 h) . \tag{4}$$

In the following we investigate a model without desorption (which is therefore described by (2) in the continuum approximation) and in which the current of freshly landed atoms is 0 on a planar surface independently of its slope, so that ν should be 0 in (3). For simplicity let us consider a square lattice: the height variable h and the substrate coordinate x are integers. All sites with $h \leqslant 0$ are occupied and form the substrate. The growth proceeds in a strip of infinite length and finite width, $1 \leqslant x \leqslant L$, with periodic boundary conditions. In every time step a particle is added on top of a column of occupied sites with a randomly chosen substrate coordinate x. Then it looks around among the surface sites at the nearest-neighbour coordinates, $x \pm 1$ and x, which one offers the strongest binding, *i.e.* the most occupied neighbours. The particle moves to this site and sticks there (see fig. 1*b*)). This is intended to simulate surface diffusion at not too high temperatures where the particles move only a short distance to find a favourable growth site before other particles are deposited on top of them. If there are as many bonds at x as next to x, the particle stays at x. If two sites next to x are equally preferable, one of them is chosen at random. The growth rule can be generalized straightforwardly for higher dimensions.

This model is very similar to the one by Edwards and Wilkinson [8, 11], where the particle moves to the lowest surface site in the neighbourhood (fig. 1*a*)) instead of the one of maximum number of bonds. However the two models produce drastically difference surface morphologies (fig. 2). Whereas in the Edwards-Wilkinson model the surface looks basically flat from the distance, it develops deep valleys with high steps perpendicular to the substrate in our model.

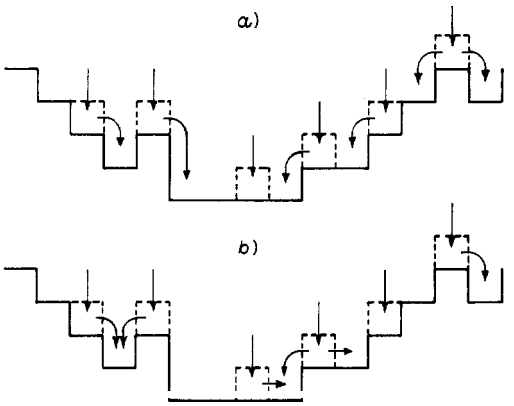

Fig. 1. – Six possible moves of a freshly landed particle on the surface. In the Edwards-Wilkinson model [8, 11] *a*) a particle that arrived on top of the column at x sticks at the lowest column among the nearest neighbours, x and $x \pm 1$. In our model *b*) it chooses the column on top of which it touches the most occupied sites. In case of a tie the particle stays at x if this column is among the favourable ones, or otherwise moves to either of the neighbour columns with equal probability.

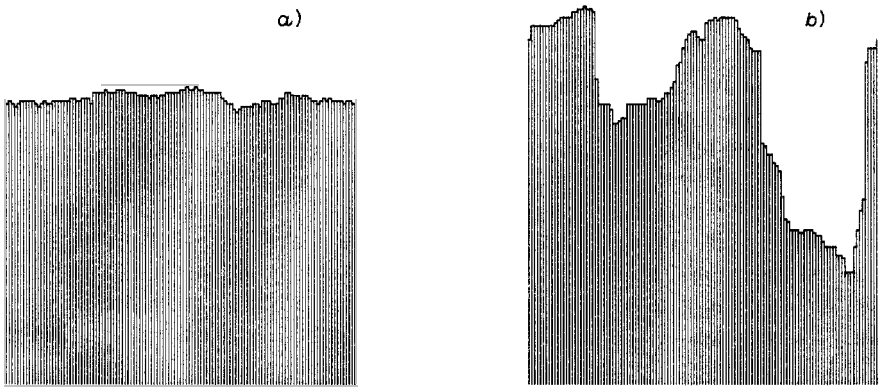

Fig. 2. – Surface configuration in the steady state *a*) of the Edwards-Wilkinson model [8, 11], *b*) of our model. The deposits grew in a strip of width $L = 120$ with periodic boundary conditions starting from a flat substrate. The average height of the surfaces is $\bar{h} = t = 2^{20}$. Only the region close to the surface is shown.

We have performed simulations by which we have measured the surface width

$$w = \langle \sqrt{\overline{h^2} - \overline{h}^2} \rangle , \tag{5}$$

where the bar denotes an average over x and $\langle \ldots \rangle$ that over many independent runs (between 1300 runs for the smallest L-values and 50 for $L = 480$). As in other growth models [1] the width increases initially with time according to a power law $w \sim t^{\beta}$ and saturates after a time $T \sim L^{z}$ (fig. 3). Time is identified with the average height. These power laws imply that the stationary value of the width scales with L according to $w_{\infty} \sim L^{\zeta}$, with roughness exponent $\zeta = \beta z$. We obtain the effective exponents (fig. 3)

$$\beta_{\text{eff}} = 0.365 \pm 0.015 , \qquad \zeta_{\text{eff}} = 1.4 \pm 0.1 , \tag{6}$$

and thus $z_{\text{eff}} = 3.8 \pm 0.5$. As $\beta_{\text{eff}} < 1$, the valley structure of the surface is confined to a

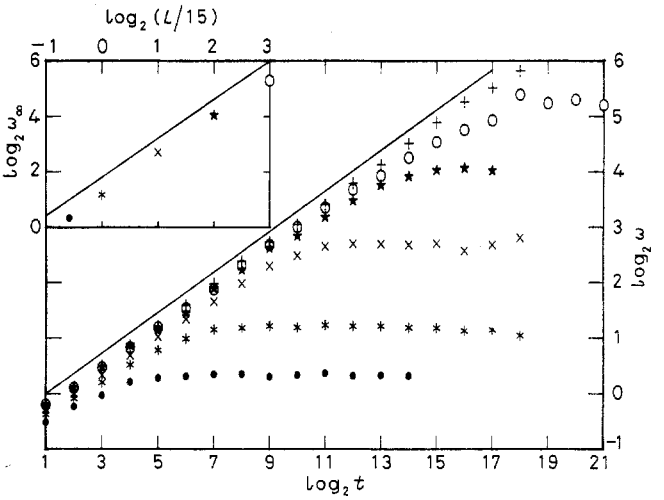

Fig. 3. – Surface width w *vs.* time t for $L = 10$ (●), 15 (*), 30 (×), 60 (★), 120 (○) and 450 (+). Statistical error bars are about the size of the symbols. The solid line has slope $\beta_{\text{eff}} = 0.365$. The inset shows the stationary values w_∞ *vs.* L. The solid line has slope $\zeta_{\text{eff}} = 1.4$.

relatively narrow growth zone compared to the film thickness. The dynamics is extremely slow compared to previous growth models where $z \leqslant 2$. This makes it difficult to investigate the stationary regime for large systems. On the IBM 3090 we were able to measure w_∞ with reasonable accuracy only up to $L = 120$. The effective roughness exponent given above is valid in this range. An exponent ζ larger than 1 means that the surface develops high steps which are responsible for a fluctuation amplitude increasing faster than the typical wavelength. Notice that in spite of $\zeta > 1$ the surface has long-range orientational order. This is enforced by the deposition process which explicitly breaks rotational invariance.

Obviously in our model the absence of shadowing of the deep valleys is essential for getting $\zeta > 1$. If particles were allowed to form overhangs at the sides of high steps the surface part underneath would stop growing. The width of the active growth zone should then scale with a roughness exponent $\zeta = 1/2$ as in two-dimensional ballistic deposition [12]. In any real growth process ζ should be $\leqslant 1$ for large system sizes and times. However $\zeta > 1$ may well describe the transient behaviour before the system reaches the asymptotic scaling regime.

The above exponents are close to the ones obtained analytically [7, 13] assuming that the time evolution of the surface is described by (2), using the expression (4) for the current along the surface. It is easy to show that this equation yields

$$z = 4 \quad \text{and} \quad \zeta = (5 - d)/2 \tag{7}$$

for space dimensions $d \leqslant 5$ and nonconserved white noise. This agreement actually comes as a surprise. The linear equation can only be expected to hold if the surface does not develop arbitrarily high steps. Therefore $\zeta = 3/2$ already disqualifies eqs. (2) and (4) for describing the long-wavelength behaviour of any generic growth model. Nonlinear terms must be taken into account. As in our model the deposition rate λ_0 determines the average growth velocity (there are no holes in the deposit), the space integral of all possible nonlinear terms added to (2) must vanish. This rules out a nonlinearity of the type occurring in the KPZ-equation. The simplest nonlinear term consistent also with the symmetry $x \to -x$ is

$\nabla^2(\nabla h)^2$. Simple power counting shows that this term is a relevant perturbation in any dimension $d < 5$ which is also true for other nonlinear terms. Therefore the exponents should change. Why do we then observe effective exponents so close to the ones obtained from (4)?

In our model the amplitude A in $w_\infty \approx AL^{\zeta_{\text{eff}}}$ is rather small: $A \approx 0.04$. Hence in order to get a surface width w of the order of L one has to study substrate sizes larger than $L \approx 600$ which would take a prohibitive amount of computation time. For our L-values it may therefore be that one sees effective exponents determined by the linear theory, and that the crossover to the asymptotic scaling can only be observed for much larger systems.

The following argument suggests that for any model described by the continuum equation (2) the exponents ζ and z should obey the hyperscaling relation

$$2\zeta = z - d', \tag{8}$$

provided $d = d' + 1$ is not larger than the upper critical dimension d_c. This is in marked contrast to the scaling relation $\zeta + z = 2$ found in models described by the KPZ-equation [14].

The values of the individual exponents as well as d_c will depend on the form of the current. For instance, if $\boldsymbol{j}$ is given by (3) one has [8, 11] $z = 2$ and $\zeta = (3 - d)/2$ with an upper critical dimension $d_c = 3$, while the current (4) leads to the exponents (7) with $d_c = 5$. In both cases (8) is fulfilled, and our simulation results (6) also agree.

The physical picture behind the scaling relation (8) is very simple: starting from a flat substrate it takes a time

$$t \propto \xi^z \tag{9}$$

until the surface has become rough up to horizontal distances of order ξ. Within the area $\xi^{d'}$ the surface has already been shaped and fluctuates over a vertical distance $w(t)$, whereas fluctuations of the height averaged over this area are yet to be developed. In every time step $L^{d'}$ particles are deposited. For models described by an equation of type (2) this means that the average height increase v_ξ in the area $\xi^{d'}$ is 1 with a variance

$$\Delta v_\xi \propto \xi^{-d'/2}. \tag{10}$$

This fluctuation in the local growth velocity is statistically independent of the pre-existing surface fluctuations so that one expects

$$w^2(t+1) = w^2(t) + \Delta v_\xi^2 \tag{11a}$$

or with (9), (10)

$$\frac{\mathrm{d}w^2}{\mathrm{d}t} \propto t^{-d'/z}. \tag{11b}$$

The scaling relation (8) is obtained by inserting $w \sim t^\beta$ with $\beta = \zeta/z$ into (11*b*). For times much larger than L^z the correlation length ξ becomes equal to L, and w does no longer increase since $\Delta v_L = 0$.

In the case of the KPZ-equation this argument breaks down because the growth velocity fluctuations (10) are also influenced by the gradients in the surface. Hence the statistical independence required for (11) is not guaranteed.

In conclusion we have demonstrated in a simple growth model that one can get effective

roughness exponents larger than 1. The surface fluctuations seem to be fairly well described by a linear theory for surface diffusion at least for small samples. In two dimensions roughness exponents smaller than 1 are always due to other effects like nonlinear contributions to the current. No matter what the current looks like, we expect the scaling relation (8) to hold for growth processes without desorption and formation of defects in the film.

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