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LETTER TO THE EDITOR

Classification of some deposition and growth processes

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Abstract. Stochastic models of surface deposition and growth processes fall into two classes with different scaling properties of the surface fluctuations. The class to which a given model belongs is determined by the variation of the macroscopic growth velocity with surface inclination. A distinction is made between deposition and growth processes, and it is shown that the non-linearity of the surface evolution equation is due to different mechanisms in the two cases.

Thirty years after the pioneering work of Vold [1] and Eden [2] simple stochastic models of deposition and growth processes continue to attract much attention [3]. While the early investigations focused on bulk properties of the deposit, the availability of large computers has made it possible to study more refined aspects of the process, such as the statistical fluctuations of the growing surface [4]. The numerical data are commonly presented in terms of the dependence of the variance ξ of the surface position on the linear substrate size L and the deposit thickness h, which can be written in the scaling form [5, 6]

$$\xi(L, t) = L^{\zeta} f(h/L^{z}). \tag{1}$$

Here ζ and z are characteristic scaling exponents and the scaling function f has the asymptotic behaviour $f(x \to \infty) = \text{constant}$, $f(x \to 0) \propto x^{\zeta/z}$.

A theory of deposition and growth processes should be able to predict the values of ζ and z for a given model. The first step in this direction is due to Edwards and Wilkinson [7]. They derived a linear Langevin equation for the growing surface which yields the exponents [8]

$$\zeta_0 = (2-d)/2 \qquad z_0 = 2 \tag{2}$$

where d is the substrate dimension. The surface is smooth ($\zeta_0 < 0$) for $d > 2$ and logarithmically rough for $d = 2$. This work was later extended by Kardar *et al* [9] who proposed the following generic equation for surface growth;

$$\partial h(x, t)/\partial t = D\nabla^2 h(x, t) + (\lambda/2)(\nabla h(x, t))^2 + \eta(x, t). \tag{3}$$

Here $h(x, t)$ measures the height of the surface relative to the average height above a point x of the substrate at time t, and $\eta(x, t)$ is white noise in space and time. With $\lambda = 0$, (3) reduces to the linear equation of Edwards and Wilkinson [7]. Kardar *et al* demonstrated [9] that the non-linear term in (3) changes the dynamic exponent to $z = \frac{3}{2}$ in $d = 1$, while $\zeta = \zeta_0 = \frac{1}{2}$. In higher dimensions the non-linearity is expected to roughen the surface as compared to (2), i.e. $\zeta > \zeta_0$, but the exact values of ζ and z are

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still unknown for $d \geq 2$. Independent of d the exponents satisfy the scaling relation [10, 11]

$$z = \min(2, 2 - \zeta). \tag{4}$$

In the physically relevant case $d = 2$ a recent theoretical approach [12] suggests that $\zeta \simeq \frac{1}{3}$.

The theory of Kardar *et al* [9] is expected to apply to any deposition or growth process which generates a compact structure with a well defined, continuous surface. The published simulation results fall into two classes which are consistent with the predictions of either the linear ($\lambda = 0$) or the nonlinear ($\lambda \neq 0$) version of (3). It is the purpose of this letter to clarify which features of a given model determine the class to which it belongs. A distinction is made between deposition and growth processes, and it is shown that the non-linearity in (3) is generated through different mechanisms in the two cases. As a consequence, a class of deposition processes which have $\lambda = 0$ is identified. A specific one-dimensional model of this class was briefly discussed in a previous publication [13].

The evolution (3) describes the growing surface on a hydrodynamic scale [14] which is large compared to the size of the individual particles. Implicit in such a description is the assumption of local equilibrium, which states that on scales below the hydrodynamic scale the system is uniform.. Therefore, up to curvature corrections, the local velocity $v = \partial h(x, t)/\partial t$ of the surface is determined by its local inclination $u = |\nabla h(x, t)|$ through a function $v(u)$ defined as the growth velocity of a *macroscopic* surface of constant inclination u. The non-linearity in (3) then arises as the leading non-trivial term in an expansion of $v(u)$ around the average inclination u_0. Hence the coupling constant is

$$\lambda = v''(u_0). \tag{5}$$

An intuitive derivation of this result, along with the scaling relation (4), was given in [15]. For one-dimensional models the calculation of $v(u)$ is considerably simplified by the mapping to driven diffusive systems [13, 16].

A *deposition process* is characterised by a uniform particle flux J towards the surface. Usually the flux is assumed to be sufficiently dilute so that different deposition events do not interfere. The geometry is chosen such that particles fall along vertical trajectories ('rain model'). The surface height is measured in the direction of the flux and the surface has some inclination u relative to the horizontal. The deposit mass per horizontally projected substrate area increases at rate J independent of u. The deposit thickness is related to its mass through the density ρ. This yields the relation

$$v(u) = J/\rho(u). \tag{6}$$

For deposition processes, a non-linear variation of the growth velocity with surface inclination is therefore solely due to a corresponding variation of the deposit density. Two classes of deposition processes may be distinguished, depending on whether the deposit has a regular (crystalline) or an irregular (amorphous) structure. In the first case, to be referred to as class I in the following, the deposit density is clearly insensitive to changes in the surface inclination. Class I processes are therefore described by the linear theory [7]. An example of such a process is the model introduced by Family [8] and extended to $d = 2$ by Liu and Plischke [17]. For this model the predictions (2) were verified both in $d = 1$ and 2. On the other hand, if the deposit is amorphous (class II processes), its internal structure and its density will respond even to slight

variations of the surface inclination. The function $\rho(u)$ has been determined for various deposition models [15, 18, 19] and also in vapour deposition experiments [20, 21]. It has a maximum at $u = 0$ which gives rise to a non-zero λ for normal incidence deposition. Typical class II deposition models are the lattice [5, 22] and off-lattice [18, 19] versions of Vold's ballistic deposition model [1], and variations of it which include partial restructuring [18, 19], a finite concentration of incoming particles [23] or spatial correlations in the particle flux [24]. In all these cases the measured scaling exponents are consistent with the predictions [9-12] of the non-linear theory.

A different scaling behaviour is observed [15, 18, 25] in the limit of grazing particle incidence ($u \to \infty$), where the deposit density $\rho(u)$ vanishes. In this regime the surface fluctuations are dominated by the discontinuous ('columnar') deposit structure. This changes the roughness exponent to $\zeta = 1$ and implies the breakdown of the hydrodynamic description [15].

The multiple restructuring model introduced by Visscher and Bolsterli [26] is somewhat intermediate between class I and class II processes. While an exact implementation of the deposition rules leads to a close-packed crystalline deposit structure, any amount of fluctuations in the sizes or positions of the particles destabilises the regular packing and generates a random packing of lower density [18, 19, 26]. Since such fluctuations are inevitable both in off-lattice computer simulations and in real experiments, the class I character of the process prevails only if the lattice structure is imposed from the outset. This explains the surprising difference found in [18, 19] between lattice and off-lattice simulations of the same model. The lattice simulations agree with the predictions [7, 8, 11] of the linear theory both in one [18] and two [19] dimensions, and also for $d = 1$ with spatial correlations in the particle flux [24]. The off-lattice simulations in $d = 1$ show a crossover from the exponents (2) at early times to those of the non-linear theory at late times [18], which is indicative of a small but non-zero value of λ [16]. In $d = 2$ the off-lattice results are inconclusive [19]. In view of the amorphous deposit structure the asymptotic behaviour is predicted to be governed by the non-linear theory. The same argument applies to possible experimental realisations of this model, such as the sedimentation of macroscopic particles from a viscous fluid.

Since the rate of growth at a point of the surface is determined by the incident particle flux, deposition processes may be termed *flux-limited.* In contrast, *growth processes* such as the Eden model [2, 27-31] are *reaction-limited* in the sense that the rate of growth depends on the number of available growth sites rather than on the supply of new material. In these cases the growth velocity is given by [27]

$$v(u) \simeq \sqrt{1+u^2}\, n(u) \tag{7}$$

where $n(u)$ is the number of growth sites per unit area of the inclined surface. For the Eden model $n(u)$ is almost independent of u [28]. Other examples of growth processes are the single-step model [17, 22, 32], the PNG model of crystal growth [13, 33, 34] and the restricted SOS model [35]. For the former two models the function $v(u)$ is known exactly in $d = 1$ [13]. In general, there is no reason to expect that $n(u)$ should compensate the non-linear prefactor in (7). Thus generic growth processes are governed by the non-linear theory. This is in accord with the simulations [6, 13, 17, 22, 29-32, 35]. Van Saarloos and Gilmer [34] interpreted their numerical results on the two-dimensional PNG model in terms of the linear theory, extracting the dynamic exponent $z = 2$. However a re-analysis of the data shows that a better fit is obtained if $z < 2$, in agreement with preliminary large-scale simulations of the model

[36]. Note that in contrast to the deposition processes discussed above, the underlying lattice is irrelevant for growth processes.

A transition from a class I deposition process to a reaction-limited growth process was recently observed in a lattice model of deposition with a finite concentration of falling particles [37]. In the limit of low concentration the model is equivalent to the lattice version of the multiple restructuring model [18] and belongs to class I. For high concentrations the surface is covered by a layer of close packed, but mobile 'mud' particles. This layer forms a reservoir from which particles are incorporated into the deposit at the available growth sites (local minima of the surface), independent of the local particle flux. The expected change in the scaling exponents as a function of concentration was clearly observed in the simulations [37].

In conclusion, a classification of deposition and growth processes has been proposed which covers all models investigated so far, and which explains why some of these models are governed by the linear theory [7]. The classification is based on the inclination-dependent growth velocity $v(u)$. For deposition processes, $v(u)$ is determined by the bulk structure of the deposit. This lends further support to the general expectation [4] that the internal structure and the surface properties of a random aggregate are intimately related.

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