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KINETIC ROUGHENING IN MODELS OF MOLECULAR–BEAM EPITAXY GROWTH[1]

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A brief survey of recent progress in understanding the kinetic roughening in growth models with surface diffusion, which are relevant for growth by molecular–beam epitaxy, is given. The main emphasis is on results of computer simulations. Properties of several different models are described and compared. In particular, results for two models, the Wolf–Villain model (and its modifications) and the full–diffusion model, in 1+1, 2+1 and also in higher dimensions are presented. The asymptotic behavior of the Wolf–Villain model is of an Edwards–Wilkinson type. Both models show an unusual scaling behaviour of the height-height correlation function.

1. Introduction

In recent years, the kinetic roughening of surfaces under the action of a driving force has become a field of increasing interest [1]. Kinetic roughening is a nonequilibrium process in which surface fluctuations exhibit an universal behavior leading to the scaling in both time and space with two characteristic scaling exponents ζ and z. This universal behavior has been observed in a wide variety of growth models and there has been considerable effort in finding different possible universality classes. However, most of the growth models studied so far (for example ballistic aggregation, Eden model, restricted solid–on–solid (SOS) model and so on) belong to the Kardar–Parisi–Zhang (KPZ) universality class [2].

Recently, properties of models with surface diffusion have been intensively investigated. Physical motivation, besides the pure theoretical interest, is understanding

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growth processes in molecular beam epitaxy (MBE). Different variants of simplified (toy) discrete models with relaxation after deposition as well as more realistic full diffusion (FD) models have been formulated [3-10]. Here we briefly describe recent results of numerical simulations and mention some experimental results as well. Since this field is a subject of intensive research (both theoretical and experimental) our review is necessary incomplete.

2. Kinetic roughening

One of the quantities which characterize the state of the surface is its roughness. Quantitatively it can be described by the surface width w. Let us consider a surface in d-dimensional space given, in the case of a strip geometry, by a single valued function $h(x,t)$ of the d'-dimensional ($d = d' + 1$) substrate coordinate x. Then in the case of a discrete model (the usual situation in numerical simulations) the surface width w is given by $w^2 = \langle \frac{1}{N}\sum_i (h_i - \overline{h})^2 \rangle$, where $\overline{h} = \frac{1}{N}\sum_i h_i$ is the average height, $h_i = h(x_i)$, $N = L^{d'}$, L is a linear size of the system and $\langle ... \rangle$ means a statistical average. Growing surfaces are in general more rough then surfaces in thermal equilibrium. Kinetic roughening describes the way in which the surface becomes rough, in nonequilibrium situation, i.e. how w evolves in time t. A remarkable fact is the existence of self–affine scaling. If the lenght in a direction parallel to the surface is scaled by a factor b and simultaneously the lenght in the perpendicular direction and the time by factors b^ζ and b^z, respectively, then the surface profile and its properties are statistically invariant. It has been found [11] that starting from an initially flat substrate, the surface width w obeys the dynamical scaling law $w(t,L) \propto L^\zeta f(t/L^z)$, where the scaling function $f(x)$ has properties $f(x) = \text{const}$, $x \gg 1$ and $f(x) \propto x^\beta$, $x \ll 1$, $\beta = \zeta/z$. Thus, w grows according to a power law, $w \propto t^\beta$, until a steady state characterized by a constant value of the width is reached after a time t_{sat} proportional to L^z. The value of the saturated width w_{sat} varies with the system size according to $w_{sat} \propto L^\zeta$. The exponents ζ and z (or ζ and β) characterize the scaling behavior of the roughness for a particular model and determine its universality class in analogy with theory of critical phenomena.

Alternatively, one can study the surface roughness using the height–height correlation function $G(\mathbf{r},t) = \langle [h(\mathbf{x}+\mathbf{r},t) - h(\mathbf{x},t)]^2 \rangle$ which obeys the scaling relation [1] $G(r,t) \propto r^{2\zeta} g(r/t^{1/z})$, where the scaling function $g(x)$ is constant for $x \ll 1$ and $g(x) \propto x^{-2\zeta}$ for $x \gg 1$. Equivalently, the structure factor S can be used. In many growth models the exponents obtained using the two different methods are equal [1]. However, in case of models with surface diffusion anomalous behavior has been found [12], attributed to the power–law increase of the average step height.

There are two main theoretical approaches to kinetic roughening: i) the study of macroscopic stochastic continuum equations of motion which are formulated using phenomenological and/or symmetry arguments, and ii) numerical simulations of discrete models defined by a set of local rules corresponding to physical processes during growth (deposition, desorption, diffusion) on atomic level.

3. Continuum approach

Supposing coarsed-grained picture one can write stochastic differential equations for evolution of the surface in the form

$$\frac{\partial h(\mathbf{r},t)}{\partial t} = \mathcal{F}\{h(\mathbf{r},t)\} + \eta(\mathbf{r},t) \quad , \tag{1}$$

where $\mathcal{F}\{h(\mathbf{r},t)\}$ is functional of the derivatives of h and $\eta(\mathbf{r},t)$ is a zero mean, random noise term in the incoming flux. The scaling exponents are usually calculated using renormalization group (RG) calculations within the one–loop approximation (see e.g. [6, 13]) or a Flory–type approximation [3]. The choice $\mathcal{F}\{h(\mathbf{r},t)\} = \nu\nabla^2 h + \lambda(\nabla h)^2$ gives the KPZ equation [2] which describes growth in the case when lateral interactions are important. The scaling exponents for the KPZ class are known only in 1+1 D, $\zeta = 0.5$ and $\beta = 1/3$ ($z = 3/2$). In higher dimensions, only results of numerical simulations are available ($\zeta \approx 0.39$ and $\beta \approx 0.24$) [1].

In a number of recent theoretical studies [3, 4, 6, 8, 13], models in which surface diffusion is the dominant physical mechanism of the surface smoothing were studied. These models are conserving (evaporation is not taken into account since it is negligible in a typical experimental situation for MBE growth) with the functional $\mathcal{F}\{h(\mathbf{r},t)\}$ in the form $\mathcal{F}\{h(\mathbf{r},t)\} = -\nabla \cdot \mathbf{j}(\mathbf{r},t)$. This corresponds to MBE growth at sufficiently high temperatures when no voids or overhangs are formed. The scaling relation $2\zeta = z - d'$ holds for these models [4]. The current $\mathbf{j}(\mathbf{r},t)$ is a function of the derivatives of $h(\mathbf{r},t)$. The most often studied cases were $\mathbf{j} \propto -\nabla h$ (Edwards–Wilkinson (EW) model [14]), $\mathbf{j} \propto \nabla\nabla^2 h$ (the *linear* diffusion model [3, 15]), and $\mathbf{j} \propto \nabla(\nabla h)^2$ and $\mathbf{j} \propto (\nabla h)^3$ (the *nonlinear* diffusion models [4, 6, 12] which we will denote I and II, respectively). The predicted values of exponents are [3, 6, 13] $\beta^{EW} = (3-d)/4$, $\zeta^{EW} = (3-d)/2$, $\beta^{lin} = (5-d)/8$, $\zeta^{lin} = (5-d)/2$, $\beta^{nonlin-I} = (5-d)/(7+d)$, $\zeta^{nonlin-I} = (5-d)/3$, and $\beta^{nonlin-II} = (5-d)/2(3+d)$, $\zeta^{nonlin-II} = (5-d)/4$. In general, one can expect also different higher order terms. However, asymptotically only some of them are relevant and lead to the values of exponents given above. Crossover to these true exponents which is caused by asymptotically irrelevant terms may be, however, rather slow.

4. Results of numerical simulations

Alternative approach to the study of kinetic roughening is to employ a powerful computer and study discrete models with microscopic rules reflecting physically important surface processes. There are two main categories of discrete models considered in connection with the study of kinetic roughening during MBE growth. In both cases SOS approximation is used. Simple (toy) relaxation models form the first group. Variety of these models has been formulated in literature [4-8]. Here we focus our attention on the model proposed by Wolf and Villain [4] (WV). The microscopic rules of the basic model are as follows. In every time step, a particle is added at a randomly chosen lattice site and then relaxes toward a nearest–neighbor site which offers the highest *coordination* (the number of nearest neighbors) where it sticks for the rest of the simulation. If the number of nearest neighbors cannot be increased, particularly in the case of *tie* (one

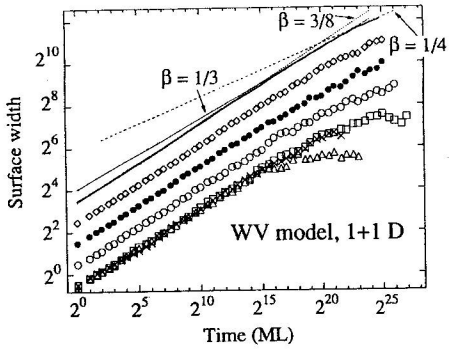

Fig. 1 - Surface width *vs.* time for the WV model in 1+1 D ($L = 150(\triangle), 300(\times), 600$ (□), $800(\circ), 1000(\bullet), 2000(\diamond)$, and 40 000 (▬)). Notice that data for larger lattice sizes ($L \geq 800$) were offset to avoid overlapping of data points.

or more neighboring sites have the same coordination as the original site) the particle stays at the initial position.

Original simulations of the model in 1+1 D [4] yielded exponents $\beta_{\rm eff} = 0.365 \pm 0.015$ and $\zeta_{\rm eff} = 1.4 \pm 0.1$ (thus $z_{\rm eff} = 3.8 \pm 0.5$) in agreement with the theoretical prediction of the *linear* model. However, a subsequent numerical work [7] has shown that in 2+1 D the values of the exponents are $\beta_{\rm eff} = 0.206 \pm 0.02$ and $\zeta_{\rm eff} = 0.66 \pm 0.03$ (thus $z_{\rm eff} = 3.2 \pm 0.5$) which correspond to the prediction of the *nonlinear* model I. The puzzling difference between the behavior of the model in 1+1 and 2+1 D has been confirmed in another numerical study [8] and stimulated further work. In recent large–scale simulations of the WV model in 1+1 and 2+1 D [16] we have found that the WV model shows complicated crossover behavior. In the case of exponents obtained from the surface width there are two crossovers in 1+1 D (Fig. 1): (i) a crossover from $\beta_{\rm eff} \approx 0.37$ (β^{lin}) to $\beta_{\rm eff} \approx 0.33$ ($\beta^{nonlin-I}$) and (ii) a crossover from $\beta_{\rm eff} \approx 0.33$ to $\beta_{\rm eff} \approx 0.25$ (β^{EW}). In 2+1 D we observed one crossover from $\beta_{\rm eff} \approx \beta^{nonlin-I}$ to the scaling behavior of the EW model, i.e. a logarithmic increase of w. Crossover times for the change to the EW behavior, $\approx 2 \times 10^6$ (1+1 D) or $\approx 3 \times 10^4$ (2+1 D) deposited layers, agree quite well with the prediction by Krug *et al.* [17]. This prediction was based on the study of the inclination–dependent diffusion current which is supposed to generate the EW term ($\nabla^2 h$) in continuum differential equations. The EW term is more relevant (in RG sense) than all allowed nonlinear terms [13] and governs the asymptotical behavior of the model. The long time needed to observe the asymptotic regime may be explained as due to a very small coefficient in front of the EW term. Another indication that the WV model belongs to the EW universality class has been very recently obtained by the study of the dependence of the saturated average step height $G_{sat}(1,t)$ for $t \to \infty$ on the system size L [18]. In higher unphysical dimensions (3+1 D and 4+1 D) the WV model shows an instability leading to a mounded surface profile [19].

It has been found recently that the WV model in 1+1 D does not fulfil standard scaling and that different values of the exponents are obtained from behavior of the surface width and the correlation function and/or the structure factor [12]. The exponents obtained in 1+1 D from the behavior of the structure factor were $\zeta^c_{\rm eff} = 0.75 \pm 0.05$ and $z^c_{\rm eff} = 2.4 \pm 0.1$ (thus $\beta^c_{\rm eff} \approx 0.31$). Exponents calculated from the correlation function are very close to the nonlinear model II predictions in 1+1 D ($\zeta^c_{\rm eff} \approx 0.75$) [12, 16], whereas they are close to the nonlinear model I ($\zeta^c_{\rm eff} \approx 0.65$)[16] in 2+1 D.

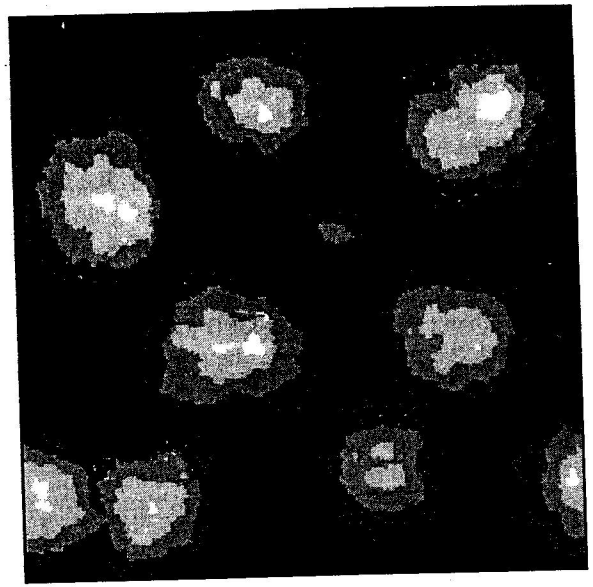

Fig. 2 - Example of the surface morphology for 300 × 300 lattice obtained in the FD model with additional step–edge barrier E_b.

FD models in which particles on the surface can move not only after deposition but during the whole simulation form the second group of discrete models. In these models there are two basic rates, one for deposition of new particles and another for surface diffusion. One can consider different mechanismus of incorporation of arriving particles, the simplest case being random deposition. A prescription for the calculation of the hopping rates can have different forms. Models with Arrhenius dynamics, in which the diffusion rate is given only by the bonding energy at the initial position, have been successfully applied in the study of early stages of MBE growth [20]. This kind of models has been first suggested in connection with kinetic roughening (in 1+1 D) by Das Sarma and Tamborenea [5] who calculated, however, only the exponent $\beta_{\rm eff}$. They found that it changes with the temperature and estimated a value $\beta_{\rm eff} \approx 0.375$ for a situation where the arrival and hopping rates in their model were approximately equal. Later Wilby et al. [9] obtained for a similar model crossover from $\beta_{\rm eff} = 0.375$ to $\beta_{\rm eff} = 0.33$ in 1+1 D , $\beta_{\rm eff} = 0.2$ in 2+1 D and $\beta_{\rm eff} = 0.09$ in 3+1 D. Siegert and Plischke [10] studied a FD model in which the hopping rate for jumps to n.n. sites i and j is given by $w_{i \to j} = k[e^{\beta \Delta E} + 1]^{-1}$. The change of the energy ΔE is calculated using the Hamiltonian $H = J \sum_{<ij>} |h_i - h_j|^n$. Results obtained for $n = 2$ (a special

case without the Schwoebel effect, see below) are $\zeta_{\rm eff} = 1.2\pm0.2$, $\beta_{\rm eff} = 0.35\pm0.01$, and $z_{\rm eff} = 3.6\pm0.4$.

In some experiments (see below) much larger values of exponents (even $\beta_{\rm eff} \approx 1$) than predicted by continuum equations with linear $\nabla^4 h$ or nonlinear $\nabla^2(\nabla h)^2, \nabla(\nabla h)^3$ terms have been obtained. It is believed that this is usually due to an instability of the growing surface caused by supression of interlayer transport due to a step–edge barrier near a step which can prevent hops of atoms off the upper terrace. This effect has been studied already some time ago by Schwoebel [21] and recently by Villain [3]. Very recently, the Schwoebel effect in the context of kinetic roughening has been studied also in numerical simulations. Zhang et al. [22] used a FD model in 1+1 D. They studied the dependence of the exponent $\beta_{\rm eff}$ on the value of the step–edge barrier E_b and obtained an increase of $\beta_{\rm eff}$ up to the value 0.5. Johnson et al. [23] investigated similar model in 2+1 D and found that growth on a singular surface is unstable and that this instability drives formation of mounds (Villain's "sawtooth" profile [3]) on the surface. An example of such surface morphology obtained from a Monte Carlo simulation of a FD model with parameters corresponding to homoepitaxy on GaAs(100) surface [24] after 300 ML have been deposited is shown in Fig. 2.

Šmilauer and Kotrla [25] found that the FD model used in [9] behaves in 1+1 D very similarly to the WV model, in particular it exhibits the same anomalous scaling due to the power–law increase of the average step height. However, in 2+1 D (and in the higher dimensions as well) both models behave very differently, the FD model providing smoother surfaces (a lower value of the exponent $\zeta_{\rm eff}^{(c)}$).

It has been also found that the linear increase of the roughness with time observed in some experiments (see the following section) can be obtained in 2+1 D in a simple modification of the WV model in which a particle in case of tie sticks at the highest position. Surface morphology obtained in this modification of the WV model containes deep grooves. The same rapid increase of the roughness in the FD model can be achieved by forbidding hops of adatoms down step edges, while allowing for hops up.

5. Experimental results

So far, experimental studies of kinetic roughening during MBE growth are not numerous and no clear identification of the experimentally determined exponents with available theories seems to be established. Chevrier et al. [26] studied epitaxial growth of Fe on Si(111) surface using reflection high–energy electron diffraction and reported the exponent β beetwen 0.22 and 0.3. You et al. [27] carried out combined X–ray and scanning tunneling microscopy (STM) measurements for Au sputter–deposited onto Si and found $\zeta^{(c)} = 0.42\pm0.03, \beta = 0.40\pm0.02 - 0.42\pm0.02$ decreasing with the increasing temperature. He et al. [28] obtained $\zeta^{(c)} = 0.79\pm0.05$ using high–resolution low–energy electron diffraction. Krim et al.[29] performed in situ STM measurements of Fe(100) bombarded by Ar^+ ions with the result $\zeta^{(c)} = 0.53\pm0.02$. Ernst et al. [30] investigated the growth of a Cu(100) crystal with He atom scattering in real time. They found that a growing singular surface is unstable and the resulting state is characterized by pyramidlike structures with the slope decreasing with the increasing temperature

(cf. [3, 23]). They obtained exponents $\beta = 0.26$ for $T = 160$ K and $\beta = 0.58$ for $T = 200$ K, i.e. the roughness increasing with the temperature, and the roughness exponent $\zeta^{(c)} = 1$ at both temperatures. Experiments with growth of Si on Si(001) [31] lead to a surprising result that the roughness linearly increases with time, i.e. $\beta_{\rm eff} \approx 1$. Very recently, high values of exponents (e.g., $\zeta^{(c)}$ between $0.6 - 1.2$) have been reported by several experimental groups [32] for metal–on–metal growth. It is our belief that in most cases the high values of the exponents are due to instability caused by the Schwoebel effect.

6. Conclusion

Results of extensive simulations of simplified relaxation models of the WV model type in 1+1 D and 2+1 D (without step–edge barriers) show that their asymptotic behavior is of the EW type. The crossover to the asymptotic region is, however, very slow and the effective exponents are much larger at intermediate times and length scales. FD models offer more realistic description of the growth process and additional possibilities, in particular for the description of the Schwoebel effect by introducing barriers to hopping at step edges. Models of this type belong either to the Edwards–Wilkinson universality class or show an instability with large exponents. A special intermediate case is the FD model with diffusion rates given only by bonding at the initial site (Arrhenius dynamics). It seems that it belongs to a new universality class [25]. Both relaxation and FD models show breakdown of conventional scaling at intermediate times, caused by an increase of the average step height. Description of this effect in a continuum picture remains to be done. In reality one expects that the Schwoebel effect will be present in many cases and that two types of generic behavior are possible for MBE growth at sufficiently high temperatures and realistic deposition rates: 1) an EW type of growth with stable and only logarithmically rough surfaces (possibly amplified by a negative Schwoebel effect, i.e. an increased probability of hopping down the step), and 2) unstable growth, caused by the positive Schwoebel effect. The surface profile in the latter case will be either grooved or composed of pyramidlike objects with the characteristic parameters (a distance between grooves, a size and an angle of inclination of the pyramids) controlled by both the model and the external parameters. In case of low temperatures or fast growth other scenarios are possible as well. If voids and overhangs are formed, growth may be described by the KPZ equation. To explain experimental results, more realistic binding energies and lattice structures should be taken into account.

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