A New Universality Class for Kinetic Growth: One-Dimensional Molecular-Beam Epitaxy

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We study a new model for kinetic growth motivated by the physics of molecular-beam epitaxy where the deposited atoms can relax to kink sites maximizing the number of saturated bonds. The model is thus intermediate between the well-known random-deposition model with no relaxation and the random-deposition model with perfect relaxation, producing growth exponents which are in between these two extremes. In particular, the growth exponent β, defining the interface width $W \sim t^{\beta}$ at intermediate times, is found to be $\beta \approx 0.375 \pm 0.005$ in $d=1+1$ dimensions. Our estimated α for this model is around 1.5 for $1+1$ dimensions.

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Kinetic growth phenomena have been a very active research area[1-5] in the last five years. There are, in our opinion, three central issues in the theoretical study of kinetic growth: (1) What are the universality classes[1,2] for various kinetic growth models? (2) What are the critical dimensionalities[4] for the various models? (In particular, are there kinetic phase transitions in physically realizable dimensions?) (3) What, if any, is the relationship between various kinetic growth models and actual vapor-deposition growth processes?

In this Letter we present results for a new kinetic growth model which touches upon the first and the third of these three questions. In fact, we believe that our model represents a new universality class which has not been, at least to the best of our knowledge, discussed before in the literature. In addition, we feel that our growth model is close in spirit to the technologically important molecular-beam-epitaxy (MBE) technique of crystal growth. In that sense, this paper represents an effort towards bridging the gap that currently exists in the literature between the MBE growth simulation studies[6] and kinetic growth studies[5]—it is quite surprising that these two subjects have evolved in the literature *totally* independent of each other even though there is obviously a close connection between the two.

Kinetic growth has been studied[7] via two different deposition models, namely, the random-deposition (RD) and the ballistic-deposition (BD) model, each of which can incorporate relaxation (RDR and BDR models, respectively). It is believed that all these models exhibit dynamic scaling properties given by

$$W \sim t^{\beta} \text{ for } t_0 \ll t \ll t_s \,, \qquad W \sim L^{\alpha} \text{ for } t \gg t_s \,, \tag{1}$$

where α and β are the kinetic growth exponents; W is the interface width (or roughness) defined as $W^2 = \sum_i N^{-1}(h_i - \bar{h})^2$, with $\bar{h}$ the average height of the interface, h_i the height of ith site (in a lattice model), and N the total number of surface sites; L is the (finite) size of the growing surface in $d-1$ dimensions; and t_0 and t_s are model-dependent characteristic time scales which define the initial transient and the saturation time, respectively. The general dynamic scaling law for kinetic growth in a d-dimensional system (the growing surface has $d-1$ dimensions) is $W(L,t) \sim L^{\alpha} f(t/L^{z})$, where f is the scaling function and the exponent $z = \alpha/\beta$. It is clear that $t_s \sim L^{z}$, and $f(x) \sim 1$ for $x \gg 1$ and $f(x) \sim x^{\beta}$ for $x \ll 1$. It is known[7] that in $d=1+1$ dimensions, $\beta = \frac{1}{2}$ (RD), $\frac{1}{4}$ (RDR), $\frac{1}{3}$ (BD and BDR).

An important point to note is that the dynamic scaling law for kinetic growth depends crucially on there being no characteristic time scales in the RD (and RDR) and BD (and BDR) models. Any relaxation or diffusion of individual atoms is instantaneous in these models and, therefore, atoms are allowed to relax only *once*, after which they are fully incorporated in the system and do not participate in the growth dynamics at all. In MBE growth, on the other hand, atomic diffusion follows an Arrhenius activation behavior where each atom hops continuously (i.e., at all times) according to a hopping rate $R = R_0 e^{-E_A/k_B T}$, with E_A the site-dependent activation energy usually taken to be $E_A = E_0 + nE_B$, where E_0 is the activation energy of a free atom with no bonds, E_B is the binding energy per bond, and n is the number of nearest-neighbor bonds that the hopping atom has in its initial site. (Usually, R_0 has a weak temperature dependence given by $R_0 = 2k_B T/h$.) Thus, MBE growth differs in an important respect from the kinetic growth models in the sense that in MBE one has two different and competing kinetic rates (or, equivalently, two competing time scales), namely, the atomic deposition rate R_D (defined as the number of atoms deposited per site per unit time) and the hopping rate R, whereas in kinetic growth models one only has a deposition rate. Clearly, for all nonzero values of R/R_D, MBE growth is in a crossover regime where random deposition and continuous hopping compete with each other and produce an effective growth exponent. As we argue below there is a fundamental difference between MBE growth and the Edwards-Wilkinson (or Kardar-Parisi-Zhang) model in that MBE belongs to a different universality class. In

MBE, the atoms chemisorb to saturate bonds and, therefore, the atoms relax to the nearest local energy minima (which is not necessarily the local height minima as in these other models). Thus, the crossover in MBE growth discussed above is to a new universality class to be discussed below.

As a starting point for more complete studies, the simulation results presented in this paper are for two-dimensional growth whereas real MBE obviously deals with the growth of three-dimensional crystals. This simplification allows comparison with definitive kinetic growth results described above and serves the purpose of establishing a new universality class associated with MBE. To avoid any confusion we refer to our growth process as one-dimensional MBE (1D MBE), emphasizing the fact that the growing interface in our simulation is one dimensional. The crossover phenomena discussed above can be seen in Fig. 1 where our calculated β for the 1D MBE growth[6] of a square lattice is plotted as a function of R/R_D for finite systems varying in size between $L=64$ and 32768. Note that for two-dimensional square-lattice simulation the number of nearest-neighbor bonds varies between 1 and 3, and therefore, the hopping rate R can take three different values depending on the number of bonds to be cut to enable the atom to hop. For the 1D MBE growth simulation results shown in this paper we choose $E_0=0.3$ eV and $E_B=1$ eV which are semiquantitatively consistent with Si and GaAs. All the results shown in this paper correspond to $R_D=1$, i.e., we grow one atomic monolayer per second. We are also using a nearest-neighbor hopping model and the solid-on-solid (SOS) approximation which implies that the only possible n entering the definition of the hopping rate R are 1 and 2, producing exponentially different hopping rates. In turns out that our numerical results change little if we set $R=0$ for $n=2$ and the value of R used to define the abscissa R/R_D in Fig. 1 is the fastest diffusion rate in the problem corresponding to $n=1$. As an inset of Fig. 1 we also show the β as a function of growth temperature.

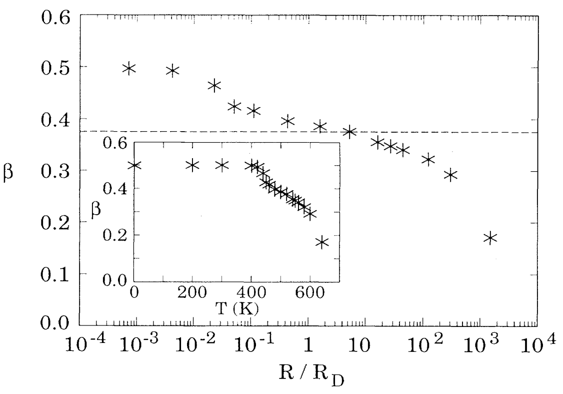

FIG. 1. The calculated *effective* exponent β as a function of the ratio R/R_D of the deposition rate to the hopping rate in the 1D MBE growth model. Inset: The calculated β as a function of temperature T which defines the hopping rate via an Arrhenius rule (one has $R_D \approx R$ for $T \approx 500$ K for the parameters used). Dashed line: $\beta=0.375$. (The system size is between 64 and 16384 depending on T.)

For $R/R_D=0$, clearly, the growth is via random deposition with no diffusion with a growth exponent $\beta=0.5$ corresponding to the Poisson distribution of the deposited columns. For high temperatures, or equivalently $R/R_D \gg 1$, each atom hops many times before the next atom is deposited and the effective exponent in a finite-size calculation approaches zero, indicating a smooth surface arising from saturation. The actual dependence of the effective exponent β on R/R_D in Fig. 1 is controlled by finite-size effects which are unavoidably present in this crossover regime of 1D MBE growth. In particular, for $R/R_D \ll 1$, we are limited by finite-time effects in our simulation with the effective β close to 0.5, whereas for $R/R_D \gg 1$, we are limited by finite-size effects and the effective exponent tends towards zero. We argue later that the actual 1D MBE growth exponent is $\beta \approx 0.375$, and it is interesting to note that around $R/R_D \sim 1$ (where the finite-size effects are minimal), β is indeed close to that value.

To answer the question of which universality class the 1D MBE growth process really belongs to, we now introduce a new growth model incorporating the atomistic hopping of 1D MBE growth without, however, the problems of having two competing kinetic rates and the continuous hopping of all the atoms. In our new model we preserve the random deposition and the solid-on-solid aspects of the other model, but incorporate diffusion by letting each atom hop with a probability p to the *nearest kink* site within a finite distance l of its initial deposition. We avoid any equilibration crossover problem of the 1D MBE growth process (cf. Fig. 1) by letting each atom complete its diffusion instantaneously (i.e., before the next atom is deposited). Thus, the new model has two crucial differences from the 1D MBE growth model: (1) We have only one rate in the problem, namely, the deposition rate (and *no* diffusion or hopping rate), and (2) each atom is allowed to relax only once and *not* continuously. As mentioned above our model is also fundamentally different from the RDR (and the BDR) model because the *atom relaxes only to the nearest kink site* and *not* to the nearest local height minima. The probability p (with $0<p\le 1$) and the diffusion length l (with l an integer satisfying $0<l<L$, where L is the lateral system size) allow us to study the details of the model and the nature of the universality class.

It is clear that our new model is in between the RD and RDR models in its surface morphology (i.e., W in the new model should be smoother than the RD model but rougher than the RDR model). Thus, β in the new model should satisfy the inequality

$$\tfrac{1}{4} < \beta < \tfrac{1}{2} .$$

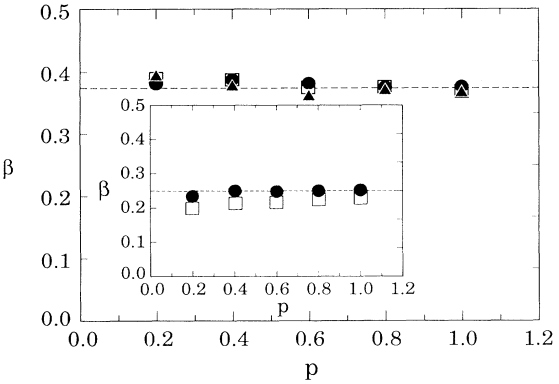

FIG. 2. The calculated β as a function of the diffusion probability p for a number of different diffusion lengths l in the new growth model. Inset: The same for the well-known model of random deposition with instant and complete relaxation (Refs. 3 and 7). The different symbols indicate different values of l [$=1$ (circle), 2 (square), 3 (triangle)]. (The system size $L=1024l$ and larger.)

Our simulation results indicate that β in our new growth model[8] is given by

$$\beta \approx 0.375 \pm 0.005\,, \tag{2}$$

independent[9] *of the values of l and p.* We, therefore, propose that 1D MBE growth (for $R/R_D \neq 0$) belongs to a new universality class which is described neither by the Edwards-Wilkinson[1] nor by the Kardar-Parisi-Zhang[2] model, but by a new universality class lying in between the RD and the RDR models and defined by the exponent of Eq. (2).

In Fig. 2 we show our calculated values of β for the new model as a function of p for different values of the diffusion length l. We have carried out a large number of simulations on system sizes varying between $L=32$ and 32768, and have typically averaged over 32 to 1024 runs to obtain our β from a linear regression analysis of the $\log W$ vs $\log t$ plots. As an inset we also show our results for β in the RDR model where we introduce l and p exactly in the same fashion as discussed above except that the atoms now diffuse to the *lowest* local kink rather than the nearest kink (within a distance l of the original deposition and with a probability of diffusion p). We have explicitly verified that for either l or $p=0$ we recover the expected RD result of $\beta=0.5$. One can see from our result in the inset of Fig. 2 that $\beta \approx 0.25$, consistent with the Edwards-Wilkinson result.

In Figs. 3(a) and 3(b) we show some actual plots (averaged over 32 to 1024 runs) of W as a function of time in our new model for various values of p and l. Consistent with the scaling laws of Eq. (1) one has three different regions in the W-t plot—for short t ($\ll t_0 \sim 1$, we use 1 layer/sec deposition rate), one always has $\beta \approx 0.5$ due to the random-deposition process; for intermediate t, we have $\beta \approx 0.375$ defining the new universality class; and then for large t ($\gg t_s$), one has saturation so that $\beta \approx 0$ [the saturation regime is not shown in Fig. 3(b)]. In Fig. 3(c) we plot W as a function of t for the 1D MBE growth model at $T=500$ K for various system sizes.

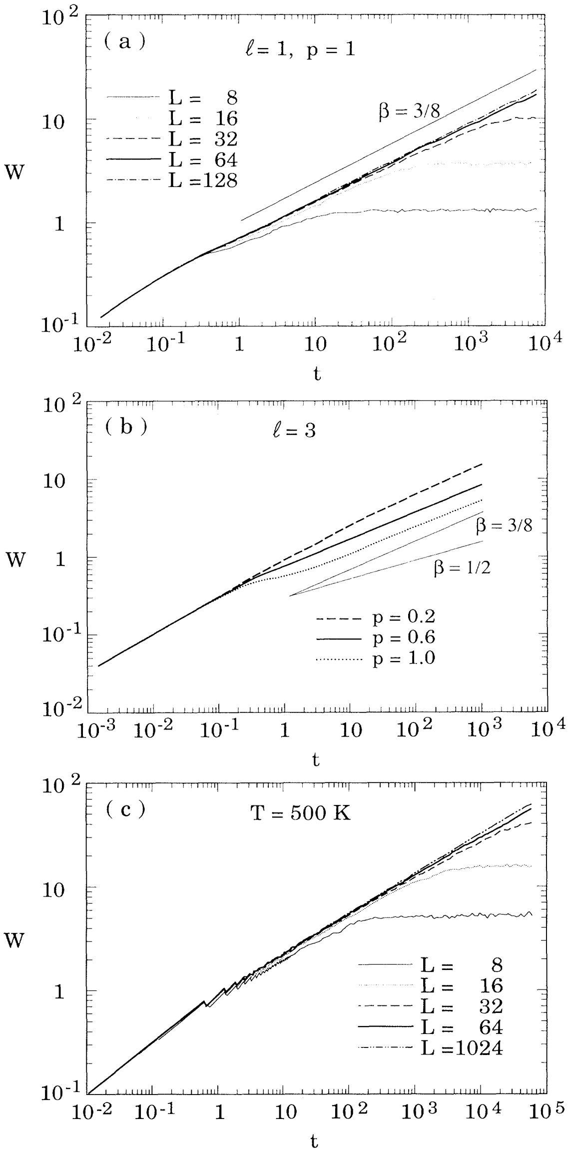

FIG. 3. Log-log plot of the calculated interface width (W) against the growth time (t): (a) in the new model for $l=1, p=1$ and for different system sizes L; (b) in the new model for $l=3$, $L=1024$ and three values of p; (c) in the 1D MBE model at $T=500$ K for various values of L.

It is interesting to speculate on the nature of the continuum model which corresponds to this new universality class (in the sense that the Edwards-Wilkinson[1] or the Kardar-Parisi-Zhang[2] equation corresponds, respectively, to RDR or BD growth). It turns out that the macroscopic linear Langevin equation, describing[10] mass diffusion under a chemical potential gradient, which is given by $\partial h/\partial t = \nu\nabla^4 h + \eta$ (where η is the noise and ν is related to surface diffusion), has the dynamical exponents $\beta = (5-d)/8$, $\alpha = (5-d)/2$ with the scaling law $\alpha/\beta = 4$. (This result is obtained by a trivial Fourier transformation of the linear equation.) In $d = 1+1$, these exponents agree very well with our simulated results. Thus, the exponents of 1D MBE growth are *apparently* well described by this simple diffusion equation which has been known for a long time. It is not clear why our growth model should be *exactly* described by this continuum equation containing *only* the fourth-order gradient term of chemical-potential-gradient-driven diffusion. One would have thought, particularly for $l > 1$, that the continuum equation for 1D MBE growth should be substantially more complicated. We consider the issue of the connection between our atomistic growth model and continuum growth equations to be open and we are currently exploring this connection.

In summary, we propose a new growth model (where the atoms relax to the nearest kink sites) which we believe represents a new universality class closer in spirit to the 1D MBE growth. The growth exponent β for the interface roughness in this model has a value of 0.375 ± 0.005 in $d = 1+1$ dimensions, which lies intermediate between the RD ($\beta = 0.5$) and the RDR ($\beta = 0.25$) models. We emphasize that the higher value of β implies a much rougher surface growth profile in this model than in either the BD ($\beta = 0.33$) or the RDR ($\beta = 0.25$) growth model. Since 1D MBE itself is not a realistic growth process, we propose experiments on vicinal semiconductor surfaces where atoms will chemisorb at kink sites and the step propagation should follow the proposed new universality. Many more results with accurate values for α ($\approx 1.47 \pm 0.1$, according to our preliminary estimate) and with the details of the surface morphology will be published in a longer paper.

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Note added.—After our work was submitted for publication we received a preprint from Wolf and Villain[11] which independently obtains some of the results contained in this paper. Another recent paper (Golubović and Bruinsma[12]) discusses the continuum growth equation[10] without any numerical results.

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[8] We have calculated β for various values of L ($32 < L < 32768$) for various l and p. Our $\log W$-$\log t$ plots are linear over 2 to 6 decades of time. The error estimate is based on variation in β extracted from various L values. Details will be given in a longer publication.

[9] Note that for any finite-size system (i.e., $l/L \neq 0$), large values of l will tend to cause saturation of the interface widths at shorter times, making it more difficult to extract an unambiguous β for larger l values.

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