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Surface roughness in thin-film growth: The effect of mass transport between layers

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We present one-dimensional simulations using kinetic parameters and deposition conditions typical to epitaxy. We study how various microscopic kinetic parameters influence surface morphology and test scaling predictions made by continuum theories.

Several continuum equations, meant to describe how the shape of a crystal surface changes when atoms are deposited on it, have been proposed and studied recently.[1–4] For one-dimensional systems these equations predict that the surface width ξ satisfies[4] the scaling relation $\xi \sim t^{\beta}$ for $t_0 \ll t \ll t_s$, where t_0 and t_s are model-dependent time scales defining the initial transient and the final saturation time.[4] The surface thickness is defined by $\xi(L,t)=[\langle h^2(x,t)\rangle - \langle h(t)\rangle^2]^{1/2}$, where $h(x,t)$ is the surface height at the point x, L is the linear dimension of the substrate.

In this paper we use one-dimensional computer simulations to investigate the kinetic processes that control the roughness of a growing surface, and to test whether the scaling relations have the form predicted by the continuum theories. We perform these simulations under conditions relevant to molecular-beam epitaxy (MBE). Some of the more interesting applications of MBE are to the growth of very small structures, such as quantum wells, wires, or dots. In these structures the distances between interfaces are very small, and the optical and electronic properties of the system are degraded by surface roughness. Because of this it is important to understand how one can grow smooth surfaces.

The desire of working under conditions relevant to epitaxy places restrictions on the kinetic model and the parameter values used in it. Because MBE is often used to grow thin films with small area, we are not interested here in scaling relations valid for infinitely thick samples, having an infinitely large surface. Furthermore, since smooth films with few defects are desired, epitaxy is performed at high surface temperatures, to ensure that an atom landing on the surface has ample time to sample a variety of surface sites and pick the one leading to a periodic crystal structure.

It is not difficult to decide what atomic properties determine whether the surface will grow smooth or rough. When the deposition starts, on a flat substrate, the atoms run along the surface, meet each other, and form small clusters. The atoms deposited next will stick to these clusters to form islands. As the islands grow larger it is more and more probable that arriving atoms land on top of them. The fate of these atoms determines whether the growth is vertical or lateral. If the atoms deposited on top of an island leave it very rapidly, by descending onto the surface on which the island is located, they will not have a chance to meet and form a new island on top of the one on which they landed. These growth conditions generate smooth surfaces and the growth is said to be "two dimensional," or "horizontal," or "layer by layer," and the deposited atoms "wet" the surface. The reflection high-energy electron diffraction (RHEED) or He scattering signal will oscillate[5] as the growth of one layer after another is completed.

In the opposite limit, the atoms landing on top of an island have difficulties leaving it. This increases the probability that the atoms located on the same island meet to form dimers, trimers, etc., which evolve into new islands. The growth is "vertical," the surface is rough, the newly deposited atoms do not wet the surface. The RHEED and He scattering signal has few or no oscillations.[5]

The parameters controlling these processes are the barrier to diffusion of single atoms on top of the island, the deposition rate, and the barrier encountered by the atoms leaving the island. With few exceptions,[6,7] previous work[8–13] have set the latter barrier to either zero or equal to the first barrier. Calculations[14,15] based on semiempirical[14] potentials or effective medium theory[15] show that, for either metals or semiconductors, this barrier is finite and in fact is always higher than the diffusion barrier of an isolated adatom on the flat part of the surface. This conclusion is backed up by experimental evidence.[16–18] Furthermore, if this barrier was zero for many materials, RHEED or He scattering oscillations, whose amplitude is recovered almost completely each period and which persist indefinitely, would be observed frequently. This is not the case. We believe that the barrier preventing an atom from descending from an island onto the surface on which the island is located is an important parameter, and we study here in detail how sur-

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face roughness depends on it using microscopic model simulations. The effect of surface morphology and roughness on step-edge-induced diffusion bias between layers has also been discussed by Villain using continuum models.[7]

We use a kinetic simulation in which the atoms move on a one-dimensional lattice with 100 sites. Tests with lattices having twice as many sites lead to the same results. At the temperatures studied here, an atom newly deposited to the cliff area of a step has sufficient mobility to quickly reach the kink site located at the lower corner of the cliff.[19] Therefore, we use a solid-on-solid model for initial deposition, such that an atom landing at the site x_i always increases the column height $h(x_i,t)$ by one unit. Overhang structures can be formed only when an atom crossing a step edge and a newly deposited atom reach the same cliff area at the same time; such events are infrequent. An atomic move i is performed with the probability $P_i = \exp[-(V_i - V_0)/kT]$. Here V_i is the energy barrier opposing the move i and V_0 is the barrier opposing a reference move. In the present simulations V_0 is the barrier encountered by a single atom moving along the surface. The potential energy barrier opposing the site-to-site movement of an atom that has n nearest neighbors is $V_i = V_0 + (n-1)V_b$, where V_b is the energy needed to separate an adatom pair. The choice made here is to speed up the simulations: an isolated atom on a flat surface makes a hop for every attempt. The barrier opposing the descent from an island to the island below is $V_i = V_0 + (n-1)V_b + V_e$. Since V_e is an essential parameter we vary it between 0 and 0.5 eV. The latter value is what one might expect on a semiconductor surface.[14] The rate to climb up a step and the rate for a border atom to leave the island are both small,[14,15] and unless the temperature is very high they do not play an essential role in kinetics. Nevertheless, they are included in the simulation since their presence does not cause too much hardship. We use in all simulations $V_0 = 1$ eV, $V_b = 0.5$ eV, temperatures between 500 and 700 K, and a deposition rate of a monolayer per second. These are reasonable values for epitaxy on semiconductor surfaces.[14,20,21]

The evolution of $\log_{10}(\xi)$ with the logarithm of the mean height h of the film (which is proportional to the deposition time), at 600 K is shown in Fig. 1, for several values of V_e. The plots are straight lines, as predicted by the scaling formula $\xi \sim t^{\beta}$, only if $V_e > 0.2$ eV. For $V_e = 0.15$ and 0.1 eV, a straight line is obtained only for higher values of h. If V_e is small, the plots oscillate, in disagreement with the scaling relation. However, if we smooth out the oscillations, the log-log plot of the smooth curve is of the form t^{β}.

These results indicate that when V_e is small the growth is practically layer by layer.[22] An ideal layer-by-layer growth—in which a whole surface layer is completed before the growth of a new one is started—cannot be achieved in practice. As a layer nears completion the number of vacant sites between islands becomes smaller. An atom landing on top of an island will have difficulties finding those few vacancies; it is more likely that it will meet another atom and start a new island. This happens even when $V_e = 0$. A typical surface morphology for this case is shown in the upper panel of Fig. 2.

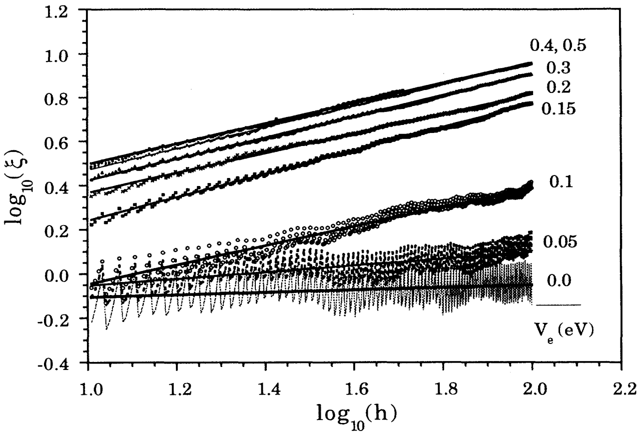

FIG. 1. The evolution of the interface width ξ as a function of the mean number of layers h deposited on the surface, at $T = 600$ K, for different values of V_e. Note the log-log scale.

The oscillations indicating the presence of layer-by-layer growth have been documented experimentally for many systems.[5] In all the experiments the amplitude of these oscillations decays in time, as the smoothness of the surface deteriorates. This is contrary to the results of the simulations for $V_e = 0$ (Fig. 1) where the oscillations are not as regular as one would see for perfect layer-by-layer growth, but are remarkably persistent in time; their amplitude fluctuates but it does not decay. We take this to mean that the existence of a system in which $V_e = 0$ has not yet been experimentally documented. Oscillations resembling more closely those observed experimentally are obtained for $V_e = 0.1$ and 0.15 eV. This does not mean that V_e has these values in the experimental system: the shape of these curves also depends on the hopping rate of a single atom along the surface, on temperature, and on the deposition rate.

The crossover from an oscillatory dependence of ξ to the form $\xi \sim t^{\beta}$ takes place in a narrow range of values of V_e.

The dependence of β on V_e, obtained from the log-log plots described above, is shown in Fig. 3, for two temperatures. If T is 600 K, β first increases rapidly over a very narrow range of V_e and then it levels off to a saturation value $\beta_s \approx 0.5$.

The dependence of β on V_e at 500 K is qualitatively similar to that observed at 600 K. There are, however, quantitative difference when V_e is small: at 500 K the thickness of the interface grows more rapidly with the deposition time. This happens because at the lower temperature the surface mobility is lower. As a consequence, even though no barrier prevents the atoms from leaving the island (i.e., $V_e = 0$) most particles cannot reach the border fast enough to avoid aggregation with the other atoms on the island; the surface is rougher than at 600 K and the simulations show that four or more layers are be-

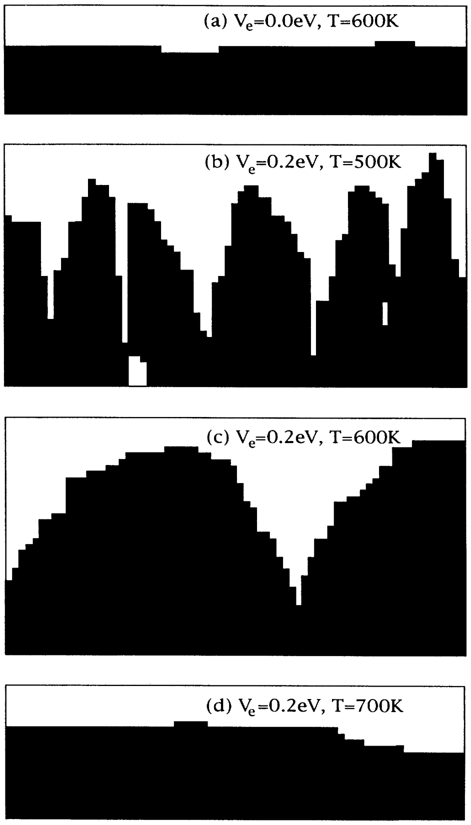

FIG. 2. Typical interface morphology for $V_e=0$ and $T=600$ K and for $V_e=0.2$ eV and $T=500$, 600, and 700 K. V_e is the excess energy barrier encountered by an atom when it descends from an island onto the layer below.

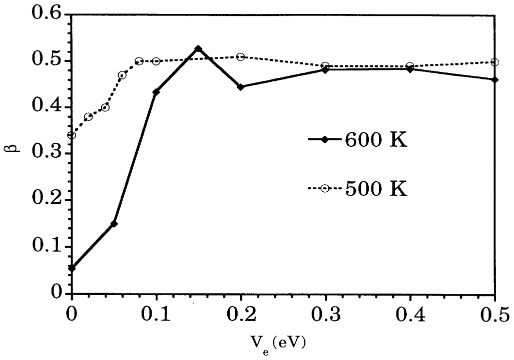

FIG. 3. The dependence of the exponent β on the edge energy V_e for $T=600$ and 500 K.

ing completed simultaneously. The value $\beta \approx 0.34$ (for $V_e=0.0$ eV) is close to that obtained by several previous simulations,[4,8–11] which either neglected surface mobility or treated it unsatisfactorily, and is also close to the prediction made by the continuum models. We regard this agreement as accidental: at 600 K we obtain $\beta=0.05$, which is very different from the prediction of the continuum equations in long-time growth limit.

We recall that in the simplest case of random deposition without surface diffusion,[23] the interface width grows as $\xi \sim t^{(1/2)}$. This case certainly does not apply to the present situation, as the adatoms always have high mobility on the flat part of the surface. It is the step-edge-induced diffusion barrier increase that drives the growth mode from being smooth to rough. Qualitatively, the value $\beta=0.5$ for high edge energies could be attributed to the lack of interactions or communication (by exchanging adatoms) between the growing islands. Physically, V_e originates from the excess surface stress at the step.[14–18] The rough, $\beta=0.5$ regime obtained here might correspond to the spinodal decomposition of the interface in the nonlinear Edwards-Wilkinson model, which includes full surface diffusion and surface stress, and predicts the same scaling exponent in some limits.[24]

The value of V_e depends on the particular system. It is smaller for metal-on-metal growth[17,18] and larger for semiconductor-on-semiconductor growth.[14,16] For most applications, one is interested in growing thin, smooth films. Once a material is chosen, one cannot change V_e except by changing the crystal face on which the growth takes place or by chemical modification. Since high deposition rates are normally desired, one is practically left, for a given system, with one variable parameter, the temperature. As illustrated by Fig. 2 the surface is smoother if the temperature is higher.

To summarize, we have investigated systematically the dependence of surface morphology and thickness of the energy barrier V_e which blocks the migration of an atom down a step from the top of an island to the layer on which the island stands. The parameters of the model were chosen to mimic the conditions used in epitaxy for material of practical interest. We found several interesting phenomena. There is rapid crossover in the dynamical behavior of the interface width, from persistent oscillations corresponding to smooth layer-by-layer growth, to a regime where the interface width increases as $\xi \sim t^{\beta}$ with $\beta=0.5$. This value is larger than that predicted by several continuum theories,[1–4] but agrees with a prediction in the nonlinear Edwards-Wilkinson model.[24] It is possible that the scaling relations derived from the Kardar-Parisi-Zhang[1] theory are valid on time and space scales too large to be relevant to the conditions (i.e., thin layers with small surfaces) used in practical epitaxy. In the layer-by-layer growth regime the evolution of the surface thickness in time consists of an exponential term t^{β} and an oscillating part which can be removed by smoothing out the "data." The values of β for these smoothed out curves are smaller than 0.5 and depend on both V_e and the surface temperature.

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