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Modelling of epitaxial film growth with an Ehrlich–Schwoebel barrier dependent on the step height

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Abstract
The formation of mounded surfaces in epitaxial growth is attributed to the presence of barriers against interlayer diffusion in the terrace edges, known as Ehrlich–Schwoebel (ES) barriers. We investigate a model for epitaxial growth using an ES barrier explicitly dependent on the step height. Our model has an intrinsic topological step barrier even in the absence of an explicit ES barrier. We show that mounded morphologies can be obtained even for a small barrier while a self-affine growth, consistent with the Villain–Lai–Das Sarma equation, is observed in the absence of an explicit step barrier. The mounded surfaces are described by a super-roughness dynamical scaling characterized by locally smooth (faceted) surfaces and a global roughness exponent $\alpha > 1$. The thin film limit is featured by surfaces with self-assembled three-dimensional structures having an aspect ratio (height/width) that may increase or decrease with temperature depending on the strength of the step barrier.

Some supplementary data available from stacks.iop.org/JPhysCM/23/292201/mmedia

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Most experiments on film production are performed in conditions far from equilibrium, in which kinetic phenomena usually overcome any thermodynamic effects in the film properties. Morphology, one of the most important features in film production, is ruled by distinct diffusion mechanisms [1, 2], the deposition rate [3] and temperature [4, 5] among other factors. In particular, the emergence of self-assembled and/or three-dimensional structures in the form of mounds has been observed during the growth of a wide diversity of films, ranging from metals [6–8] to inorganic [5, 9] and organic semiconductor materials [10, 11]. This mound instability may be an undesirable feature in the production of atomically flat interfaces but becomes a valuable tool to produce structured surfaces, both systems with important technological applications.

The three-dimensional growth is attributed to kinetic barriers that promote the imbalance between up- and downhill currents in stepped surfaces. A number of different mechanisms have been proposed to explain these barriers, but there are only a few cases in which the observation of an unstable growth can clearly be associated to a particular barrier [5]. In two seminal papers, Ehrlich \textit{et al} [12] and Schwoebel \textit{et al} [13] independently reported that adatoms lying in the edge of terraces diffuse backward in the same layer more frequently than they move to a lower step.
Organized the paper as follows. In section 2, we develop the model and present the simulation procedures. In section 3, the simulation results are presented and discussed. Finally, our concluding remarks are made in section 4.

Figure 1. The barrier model in multilayer steps. (a) A particle in an ascending step detaches from its initial position (empty square) and starts an unbiased random walk along the normal direction (red). (b) The particle can be reflected in the top due to the presence of an ES barrier. (c) A particle in a descending step (empty square) overcomes the ES barrier with probability \( p = \exp(-E_b/k_B T) \) and then diffuses exactly as in (b). (d) Random walk problem including the absorbing/reflecting boundaries indicated by X.

These experiments led to a widely accepted hypothesis of an additional activation energy to downhill diffusion known as the Ehrlich–Schwoebel (ES) barrier. Besides the ES barrier, alternative mechanisms have been proposed to explain mound instability. Examples include the short-range attraction of adatoms towards ascending steps [14] and fast edge diffusion [15–17].

Kinetic Monte Carlo (KMC) is a standard simulation method used to investigate the morphology and dynamics during epitaxial film growth [2]. In a KMC simulation, the events (deposition, diffusion, evaporation, etc.) are implemented, one at a time, with specific rates. In particular, diffusion is a thermally activated process occurring at a rate given by an Arrhenius law \( D = v_0 \exp(-E_D/k_B T) \), where \( v_0 \) is the attempt frequency, \( T \) is the growth temperature and \( E_D \) is the diffusion activation energy [18]. An ES barrier is usually included in KMC simulations by raising \( E_D \) when the particles are lying in descending steps. This additional energy effectively inhibits downhill diffusion and, consequently, increases the chance of nucleation of new layers on top of the terraces. The result is an instability responsible for the emergence of three-dimensional structures [19].

Molecular dynamic simulations show that an efficient upward diffusion can also be present in epitaxial growth [20]. Additionally, molecular dynamic simulations also show that the ES barrier can change substantially from monolayer to multilayer steps [21]. With these motivations, we investigate a model with a step height dependent ES barrier using KMC simulations. We are interested in the effect of these barriers in a model with a step height dependent ES barrier using KMC simulations [21]. With these motivations, we investigate the ES barrier can change substantially from monolayer to multilayer steps and the KMC simulations were performed as follows. Particles are deposited on a substrate kept at a constant temperature \( T \). The substrate, represented by a triangular lattice with periodic boundary conditions, is initially flat. Deposition occurs normally to the substrate at a constant rate \( F \) and obeys the solid-on-solid restriction to prevent overhangs. The height profile \( h_j \) is therefore the number of particles adsorbed in the site \( j \). The deposition involves two steps: firstly a site is picked up at random and secondly a new particle is deposited in the most energetically favourable (largest bond number) site among the chosen one and its nearest neighbours. If there are multiple options, one is randomly chosen. This deposition rule is equivalent to the classical Wolf–Villain model [22].

Intralayer diffusion from a site \( j \) to \( j' \), \( \Delta h(j, j') = h_{J'} + 1 - h_j = 0 \), has an activation energy \( E_D(j, j') = E_0 + n_j E_N \), where \( E_0 \) represents the interaction with the substrate and \( E_N \) the contribution of each one of the \( n_j \) lateral bonds. The diffusion rate is therefore given by the Arrhenius law:

\[
D(j, j') = v_0 \exp\left(\frac{-E_D(j, j')}{k_B T}\right).
\] (1)

Apart from the step barrier, this diffusion rule is similar to that used in the model proposed by Šmilauer and Vvedenski [18]. Interlayer diffusion is allowed for particles lying in both ascending or descending steps. For monosteps, \( \Delta h(j, j') = \pm 1 \), the barrier is implemented as usual: an additional barrier \( E_b \) is present in a descending but not in an ascending step. For steps higher than a monolayer, the interlayer diffusion is implemented in two parts. If the diffusing particle lies in an ascending step, it first detaches from its initial layer at a rate given by equation (1) and then executes a non-directed one-dimensional random walk, perpendicularly to the substrate, as schematically illustrated in figure 1(a). A step barrier is also present in the top of the step (figure 1(b)). The rule for a particle in a descending step is very similar except that it has to overcome the ES barrier before attaching to the lateral (figure 1(c)). Notice that the barrier in the top makes the model symmetric in relation to down- and upward diffusion in multilayer steps, an improvement of the usual rule without height restrictions.

The determination of the probability of the particle moving to a neighbour site or being reflected to its initial position involves the solution of a one-dimensional random walk with absorbing/reflecting boundaries. When the particle visits the boundaries, it becomes trapped with a given probability or is reflected with complementary probability. This problem is schematically illustrated in figure 1(d) for a step of height \( \ell = 4 \). In our model, each boundary represents either the initial or neighbour sites. The reflection is due to the ES barrier and, therefore, we set adsorption probabilities 1 and \( p = \exp(-E_b/k_B T) \) for the bottom (\( i = 0 \)) and the top
Figure 2. Surface morphologies for growth times varying from 5 to 400 ML. The growth temperature is \( T = 573 \) K and a mild step barrier \( E_b = 0.02 \) eV is used.

\[
q(\ell + 1|i) = \frac{i}{\ell + 1/p}.
\]

For an ascending step (figure 1(a)), it is easy to see that the probability of moving to the other layer is \( q(\ell + 1|1) \), while for a descending step this probability becomes \( q(\ell + 1|\ell)p \). In the latter case, the factor \( p \) takes into account the step barrier when the particle crosses the kink (figure 1(c)). The resulting probability of diffusion through the step, for both cases, reads as

\[
P_{\text{cross}} = \frac{p}{1 + p(|\Delta h| - 1)}, \quad |\Delta h| \geq 2,
\]

where \( \Delta h = h_j + 1 - h_j \) is the height of the step to be crossed.

3. Results

The KMC simulations were performed with fixed parameters \( E_0 = 1 \) eV, \( E_N = 0.11 \) eV, \( F = 1 \) monolayers (ML) s\(^{-1}\), \( \nu_0 = 10^{13} \) s\(^{-1}\). Temperatures \( T = 473, 523 \) and \( 573 \) K were simulated for deposition times of \( 10^4 \) ML. Except when explicitly mentioned, we report results for \( 256 \times 256 \) sites. The initial surface evolution for a growth temperature of \( T = 573 \) K and a weak ES barrier of \( E_b = 0.02 \) eV can be followed in figure 2. One can see the surface coarsening and the emergence of three-dimensional structures with a characteristic size in the form of mounds. For very long times, however, the instability is highly enhanced, turning the surfaces into deeply grooved morphologies with large terraces.

A rich variety of spatio-temporal properties is observed in the thin film phase. The present model has an intrinsic step barrier independent of the parameter \( E_b \) when \( |\Delta h| \geq 2 \). If we set \( E_b = 0 \), the system has a transition from a rough (fractal) surface at low temperatures to an almost layer-by-layer growth at high temperatures (inset of figure 5(b)). The layer-by-layer regime is transient and becomes self-affine for asymptotic times, as we will see later. However, a weak barrier of \( E_b = 0.02 \) eV is sufficient to induce a temperature-driven instability and to form mounds. In the top row of figure 3(a), we can see the temperature effect for this weak barrier. The mound aspect ratio (height/width) decreases with temperature, a behaviour also observed in homoepitaxy of
Figure 4. Characteristic length against time for (a) $T = 473$ K and (b) $T = 523$ K using distinct step barriers. The inset shows the correlation functions for different deposition times and $E_b = 0.07$ eV. The curves correspond to averages over 20 independent samples. Error bars smaller than the symbols were omitted. The dashed lines are power laws with the indicated exponents as guides to the eyes.

Figure 5. Interface rms roughness evolution for (a) $T = 473$ K and (b) $T = 523$ K. The inset shows the initial layer-by-layer growth for $T = 523$ K. The dashed lines are scaling laws obtained from data regressions.

some semiconductor materials [24]. If a step barrier $E_b = 0.07$ eV is used instead, the surface morphology changes considerably. The uphill diffusion becomes strong at higher temperatures, generating self-assembled three-dimensional structures, as shown at the bottom of figure 3(a). Details are illustrated in figure 3(b), where isolated grains for temperatures $T = 523$ and 573 K are shown. Higher temperatures make the grains higher, resulting in a large increase of the aspect ratio.

The thick film limit also exhibits very interesting dynamics. A basic quantity to characterize the surface morphology and dynamics in this regime is the height–height correlation function defined as [15]

$$\Gamma(r) = \langle h(x)h(x+r) \rangle_x, \quad (4)$$

where $h(x)$ is the surface height in the mean height reference and $\langle \cdot \cdot \cdot \rangle_x$ represents the average over the surface. The first zero of $\Gamma(r)$, denoted by $\xi$, is a characteristic length of the surface. Typical correlation functions for distinct growth times are shown in the inset to figure 4(a). The oscillating behaviour observed in the correlation functions is the hallmark of mounded surfaces [1, 15]. The length $\xi$ is expected to increase as a power law of time, $\xi \sim t^\zeta$, where $\zeta$ is the so-called coarsening exponent [15]. Another basic quantity to characterize the surface dynamics is the interface rms roughness given by $w = \sqrt{\langle h^2 \rangle}$. For a kinetic roughening, this quantity grows as a power law of time, $w \sim t^\beta$, where $\beta$ is the growth exponent. In dynamical scaling theory [1], the coarsening and growth exponents are related with the roughness exponent by the scaling relation $\alpha = \beta / \zeta$. These exponents are very useful because they may determine the universality class of the system and, from the framework of the generic dynamical scaling theory, they may connect scaling and morphological properties of the surface [25].

For all investigated temperatures the scaling exponents change depending on whether an explicit step barrier is present or not. For the lower temperature of $T = 473$ K and in the absence of an explicit step barrier ($E_b = 0$), we found a power law regime with coarsening and growth exponents $\zeta = 0.32$ and $\beta = 0.19$, respectively, as shown in figures 4(a) and 5(a). The surface evolution for these parameters is shown in movie 1 of the supplementary material (available at stacks.iop.org/JPhysCM/23/292201/mmedia). These exponent values are in good agreement with the exponents $\zeta = 3/10$ and $\beta = 1/5$ obtained for the classical Villain–Lai–Das Sarma (VLDS) growth equation [19, 26]

$$\frac{\partial h}{\partial t} = -\nu \nabla^4 h + \lambda \nabla (\nabla h)^2 + \eta, \quad (5)$$
where $\eta$ is a Gaussian noise. Similar exponents are obtained for higher temperatures and $E_b = 0$. Interestingly, in the limit of zero temperature, our model corresponds to the Wolf–Villain model [19] that has a crossover [27, 28] to the universality class of the Edwards–Wilkinson (EW) equation \( \partial_t h = \nabla^2 h + \eta \) with exponents $\beta = 0$ (logarithmic growth) and $\xi = 0.5$ in $d = 2 + 1$ [29]. In contrast, our simulations at higher temperatures point out the opposite behaviour: a small growth exponent (consistent with a logarithmic growth) and a coarsening exponent close to $1/2$ are observed for early growth times whereas $\beta \approx 1/5$ and $\xi \approx 3/10$ are seen only at long times. The results suggest that the diffusion, not the deposition rules, is governing the dynamics.

In the presence of an explicit ES barrier, the scaling exponents for $T = 473$ and 523 K converge (more slowly for $E_b = 0.02$ eV than for 0.07 eV) to $\beta = 0.33$ and $\xi = 0.22$, as shown in figures 4 and 5. A growth exponent $\beta = 0.33$ were also obtained for the higher investigated temperature of $T = 573$ K, but the measured coarsening exponent, $\xi \approx 0.18$, seems not be the asymptotic value. Anyway, the corresponding roughness exponents, given by $\alpha = \beta/\xi$, are larger than 1, implying that the explicit barrier causes the so-called super-roughness dynamical regime [25]. Super-roughness is featured by a locally smooth surface with rms roughness in windows of size $\ell$ scaling as $w(\ell) \sim \ell$ for $\ell \ll \xi$. The left panel of figure 6 shows surfaces for a long deposition time obtained for $E_b = 0.07$ eV. For $T = 473$ K, the system evolves from an initially rough (figure 3(a)) to a faceted morphology exhibiting pyramidal mounds, as can be seen in movie 2 of the supplementary material (available at stacks.iop.org/JPhysCM/23/292201/mmedia). Similar structures are obtained for a mild barrier $E_b \approx 0.02$ eV, but the faceted morphology is less evident. The surface dynamics becomes richer for higher temperatures. Initially, self-assembled and fast growing structures are formed due to the instability caused by the step barrier. Therefore, these structures start to coalesce and to form large scale mounds also exhibiting pyramidal morphology. The surface evolution can be followed in movie 3 of the supplementary material (available at stacks.iop.org/JPhysCM/23/292201/mmedia).

4. Concluding remarks

The pattern formation in semiconductor films produced with epitaxial techniques is a subject of basic and technological interest. The formation of three-dimensional structures is related with the so-called Ehrlich–Schwoebel (ES) barrier. This barrier is usually implemented in kinetic Monte Carlo (KMC) simulations by reducing the diffusion rate of particles in descending steps. In the present work, we investigate the homoepitaxial growth of films by performing KMC simulations where the ES barrier depends explicitly on the step height. In addition, our model includes a barrier to interlayer diffusion in ascending steps when their heights are larger than a monolayer.

The model yields surfaces with mounded morphology even for a mild step barrier of $E_b = 0.02$ eV, a value much smaller than the typical nearest neighbour interactions of $E_N = 0.11$ eV used in the present KMC simulations. However, a layer-by-layer growth, lasting during the deposition of several layers, is obtained if the explicit ES barrier is null. For an intermediate barrier, $E_b = 0.07$ eV, self-assembled three-dimensional structures are obtained. The aspect ratio (height/width) of these structures decreases with temperature for a mild barrier but increases for the intermediary one.

We have also investigated the thick film limit using the growth and coarsening exponents, defined as $w \sim t^\beta$ and $\xi \sim t^\xi$, respectively. In the absence of an explicit barrier, the interface undergoes kinetic roughening with exponents $\beta \approx 0.19$ and $\xi \approx 0.30$, which is consistent with the non-linear Villain–Lai–Das Sarma (VLDS) equation [19, 26] for molecular beam epitaxy (MBE). In the presence of a step barrier, the exponents change to $\beta \approx 0.33$ and $\xi \approx 0.22$, resulting in a roughness exponent of $\alpha = \beta/\xi > 1$ which represents the so-called super-roughness scaling regime [25] for the surface evolution. This regime features large pyramidal structures that are observed in MBE systems like Ge(100) [24] and Cu(100) [30]. Even though a large number of models have been proposed to reproduce specific systems with pyramidal morphology [2], our model is quite simple, robust and does not require any kind of parameter tuning.

The present model addresses two features that, to our knowledge, were neglected in the broad literature of KMC modelling of epitaxial growth: the influence of the step height in the ES barrier and the explicit barrier when the particle is executing an uphill diffusion. This last one is particularly interesting since the rules for the movement through a kink become symmetric in relation to up- or downhill movements (figure 1(b)). Finally, the specific form of the interlayer
diffusion probability (equation (3)) is not essential to the surface dynamics. We have also simulated, but not shown in this paper, a model where equation (3) is replaced by $P_{\text{cross}} = p$ for $|\Delta h| \geq 2$ and observed that all the morphological properties and scaling exponents are preserved.

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