

THE DYNAMICS OF THIN FILM GROWTH: A MODELING STUDY

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For closed-loop control of thin film deposition, one would like to directly control film properties such as roughness, stress, or composition, rather than process parameters like temperatures and flow rates. This requires a model of the dynamic response of film properties to changes in process conditions. Direct atomistic simulation is far too slow to be used in this capacity, but a promising approach we explore here is to derive reduced-order dynamic models from atomistic simulations.

In this paper, we consider film growth on a vicinal surface using a kinetic Monte Carlo model. The temperature range spans the transition from smooth step flow to rough island growth. Proper Orthogonal Decomposition is used to extract the dominant spatial modes from the KMC simulations. Only five spatial modes adequately represent the roughness dynamics for all simulated times and temperatures, indicating that a 5-state model may be sufficient for real-time roughness control.

INTRODUCTION

Control of thin film deposition is becoming increasingly important as the need to deposit thin films with a very precise thickness, composition, and microstructure grows. While current practice is to regulate indirect process parameters that are easily monitored, in many cases it would be preferable to directly sense and control film properties during deposition.

In addition to developing sensors to sense in real time the state of the film, it is also necessary to know how the film state will respond dynamically to changes in controllable process parameters. This requires a model that can compute the dynamic evolution of the film in real time.

Atomistic models are orders of magnitude too computationally expensive for this application, but simpler PDE or rate-equation models are usually derived by making *ad hoc* simplifying assumptions that may not correctly capture the dynamics of this complex, nonlinear process. For example, island nucleation is a strong nonlinear function of adatom concentration; models that assume adatoms are distributed uniformly on the surface may make substantial errors in computing island nucleation rates.

A promising alternative approach is to develop reduced-order dynamical models from atomistic simulation data, such that the dynamic response of, for example, the roughness is captured in the reduced-order model without requiring *ad hoc* simplifying assumptions.

In this paper, we explore the dynamics of film growth on a vicinal surface using a kinetic Monte Carlo (KMC) model, over a temperature range spanning the transition from step flow growth at high temperature to island growth and coalescence on

terraces between rough steps at low temperature. We use the technique of Proper Orthogonal Decomposition to identify spatial modes that represent the major features of the surface profiles.

KINETIC MONTE CARLO SIMULATIONS

We studied the surface morphology of a growing crystal with a kinetic Monte Carlo model. We consider a single-species material on a cubic lattice, and we increment time as in Fichtorn and Weinberg [1] to achieve a physically-based time. Vacancies in the crystal are prohibited. We define reaction rates for adsorption, desorption, and surface diffusion based on a nearest-neighbor bond-counting model:

$$k_{\text{ads}} = \gamma P_j \sqrt{\frac{1}{2\pi m k_b T}} \quad (1)$$

$$k_{\text{des},i} = \frac{k_b T}{h} \exp\left(-\frac{E_{\text{des},0} + i\Delta E}{k_b T}\right) \quad (2)$$

$$k_{\text{dif},i} = \frac{k_b T}{h} \exp\left(-\frac{E_{\text{dif},0} + i\Delta E}{k_b T}\right), \quad (3)$$

where i , ranging from 0 to 4, is the number of adjacent side neighbors, k_{ads} is the adsorption rate, $k_{\text{des},i}$ is the desorption rate for a surface site with i nearest neighbors, and $k_{\text{dif},i}$ is the diffusion rate for a surface site with i nearest neighbors. The Boltzmann constant is denoted with k_b , Planck's constant is h , the sticking coefficient is γ , the mass of the particle is m , temperature is T , and the precursor partial pressure is P_j . The chemistry model has four free parameters: three activation energies and a constant in the adsorption rate. The activation energies $E_{\text{des},0}$, $E_{\text{dif},0}$, and ΔE are the depths of the potential energy wells associated with the occurrence of a surface event. Specifically, $E_{\text{des},0}$ is the energy for the desorption of an atom with no side neighbors, $E_{\text{dif},0}$ is the energy for the diffusion of an atom with no side neighbors, and ΔE is the additional energy barrier associated with a single side neighbor.

We performed simulations on a 256×256 domain and deposited 4 layers of atoms. Eight steps were inserted into the initialized lattice, and periodic boundary conditions were used to simulate an infinite train of steps. The reaction rate parameters were $\gamma(2\pi m k_b)^{-0.5} = 5\sqrt{\text{KPa}}\text{s}^{-1}$, $E_{\text{des},0} = 2.64 \times 10^{-18} \text{J}$, $E_{\text{dif},0} = 3.02 \times 10^{-19} \text{J}$, and $\Delta E = 7.59 \times 10^{-20} \text{J}$. The activation energy for desorption is sufficiently high such that desorption is negligible in the simulations. We considered a nominal partial pressure $P_{j,o} = 1 \text{ Pa}$ and nominal temperatures T_o of 950 K, 1050 K, and 1150 K.

The temperature range was selected to span the transition from step flow growth at high temperature to growth primarily by island nucleation at low temperature. At intermediate temperatures, both processes contribute, as shown in Figure 1. This figure shows that the islands are not distributed randomly on the terrace. Since the steps are sinks for adatoms, the adatom density is highest away from the steps, and it is here that islands preferentially nucleate.

Layer-by-layer growth via island nucleation and step growth results in a periodicity at the monolayer growth frequency that is usually detected as oscillations in the measured RHEED signal. This behavior is seen in plots of the step edge density,

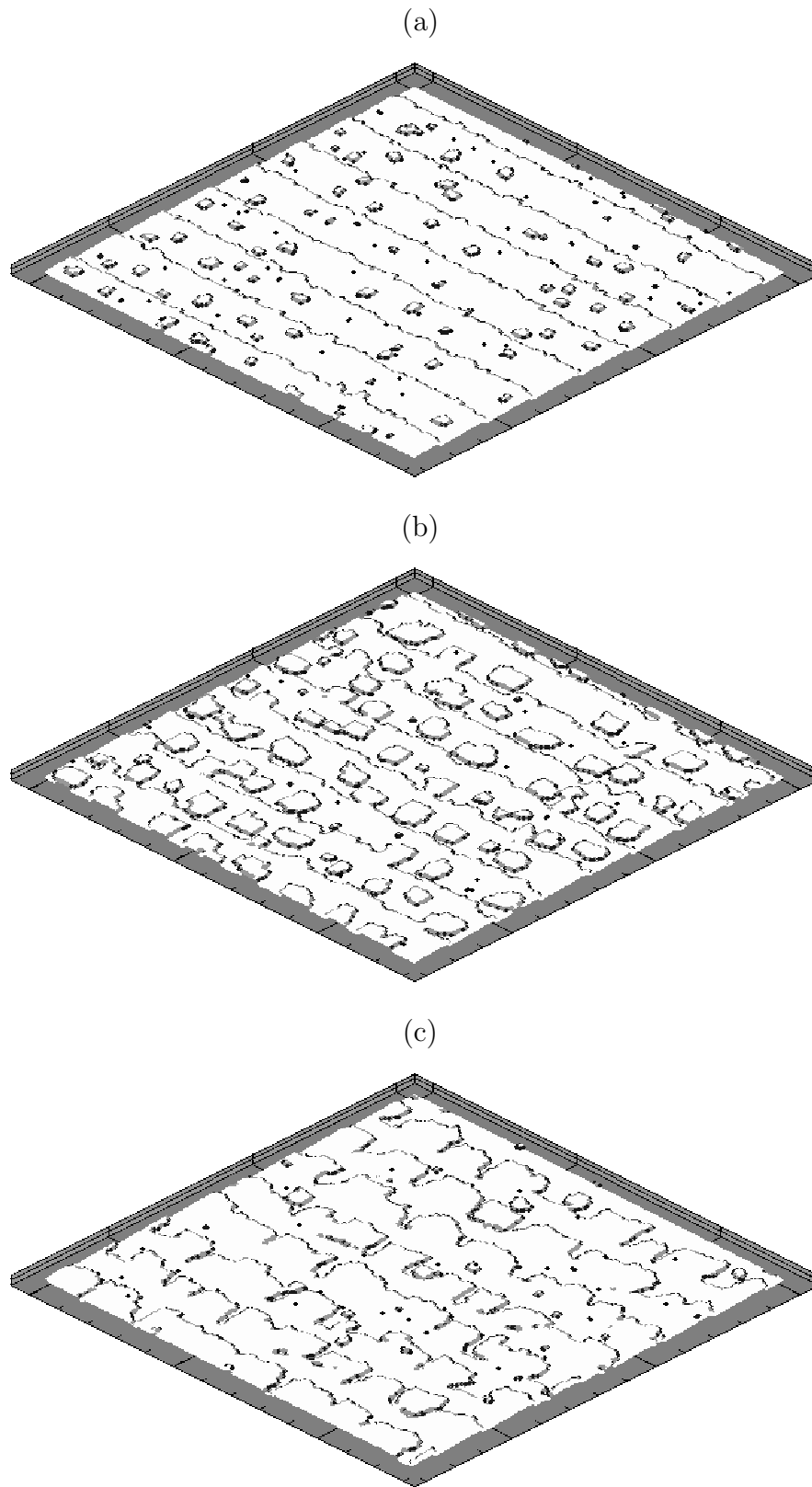


Figure 1: Monte Carlo simulation at 1050 K for various stages of growth: (a) 0.16 layers, (b) 0.54 layers, (c) 1.0 layers.

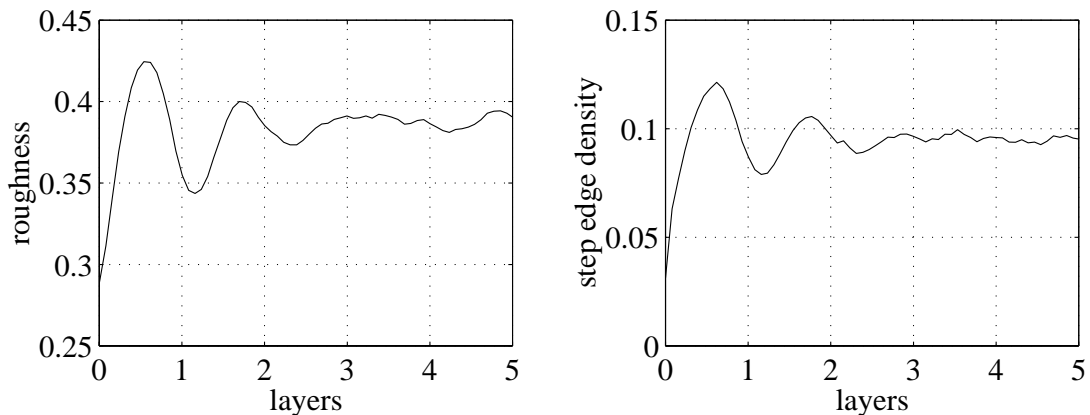


Figure 2: Roughness and step edge density from a KMC simulation at $T = 1050$ K.

which serves as a model for the RHEED signal, and of the roughness

$$W = \left[\frac{1}{A} \int_A (h(x, y) - \bar{h})^2 \right]^{\frac{1}{2}}, \quad (4)$$

where A is the domain area, h is the surface height, and \bar{h} is the spatially-averaged height. These measures are plotted in Figure 2 for the simulation pictured in Figure 1. The oscillations decay due to roughening of the step edges. Note in Figure 1 that the step edge is not straight at the completion of one layer of growth.

The oscillations in surface properties are influenced by the spatial non-uniformities imposed by the steps. Because the islands are concentrated near the original center of the terrace, the collision of steps with islands happens suddenly. Once the steps have collided with the islands, the surface becomes smoother. Notice that in Figure 1(b) the steps have not yet collided with most of the islands, while in Figure 1(c), the step has merged with the islands, creating a minimum in roughness and step edge density.

To better understand the spatial distribution of islands, we examine the shape of the surface height profile for a range of growth conditions, from step-dominated growth at 1150 K to island-dominated growth at 950 K. We also consider growth under sinusoidally-varying temperature and precursor partial pressure in an attempt to excite different spatial modes. To obtain spatial profiles across the terrace, we average the surface height over the direction parallel to the steps. We also average over the eight steps to yield the surface height profile for a single step as a function of the distance along the terrace. Spatial profiles throughout the growth of four layers are shown in Figure 3. At low temperature, islands grow between the steps and dominate the growth. As time advances, the surface becomes more disordered, which may be seen in the smoothing of the surface profile. At high temperatures, growth occurs as adatoms attach to the step edge. The propagation of the step is the dominant spatial feature in this limit. Notice that the vertical scale is greatly expanded, so that the initial stepped surface appears to be a sawtooth wave.

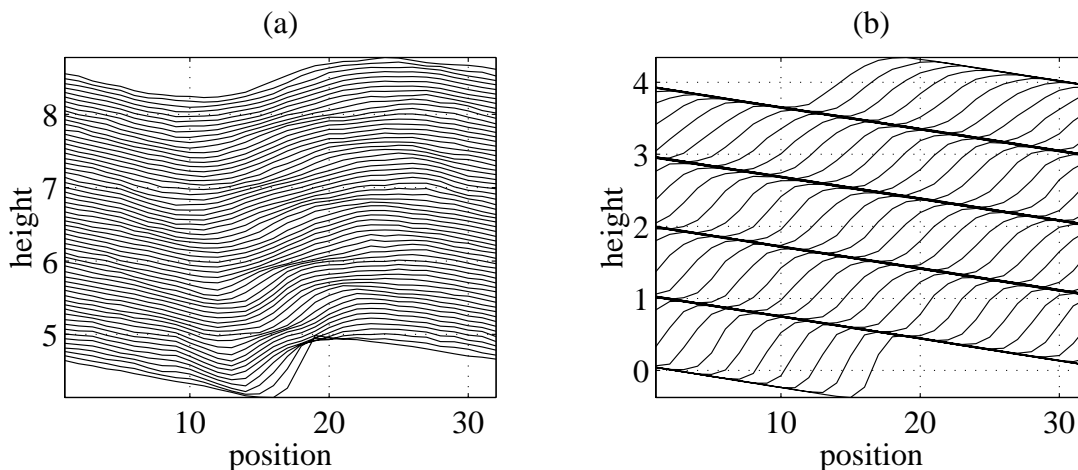


Figure 3: Surface height as a function of position in the direction perpendicular to the step, for KMC simulations at (a) 950 K, (b) 1150 K.

PROPER ORTHOGONAL DECOMPOSITION

We use a method called Proper Orthogonal Decomposition (POD) to extract the typical shape of the surface height profile [2]. The input to POD is a collection of “snapshots” of data. As output, the POD method returns a series of orthogonal spatial modes, which are ordered in decreasing importance. The root-mean-square error between the snapshots and their projections onto the first n modes possesses the minimum root-mean-square error that any n orthogonal modes could (where n is an integer from 1 to the number of snapshots). Thus, the POD modes are considered to be optimal. If low-order behavior exists in the spatial profile, the first few POD modes will capture a large percentage of the total spatial profile.

In this study we take as our snapshots all of the spatial profiles plotted in Figure 3. In Figure 3(b) the surface maintains a typical shape which propagates in time. To capture more energy in fewer POD modes, we preprocess our snapshots with a procedure called template fitting [3], in which the spatial profiles are shifted to line up with each other.

The first five spatial modes determined by POD are plotted in Figure 4. The first mode captures 96.6% of the energy, meaning that the root-mean-square error between the KMC surface profiles and the projections onto the first mode is only 3.4%. The shape of the first mode represents the overall shape of the original step in the KMC simulations. Its corners are rounded, consistent with the smoothing of the profiles seen in Figure 3. The second mode contains 2.7% of the energy, totaling 99.3% for the first two modes combined. This mode has features on its ends which, when added to the first mode, can sharpen or smooth the overall shape. The feature in the center of the second mode represents island growth on the terrace. The third, fourth, and fifth modes are also shown in Figure 4. They contain little energy. The most dominant features of these modes are on the right side. As in the second mode, the features on the right side resemble sinusoids in various phases with each other. These modes can be combined to reconstruct a traveling wave.

The projection size of each snapshot onto each mode is shown in Figure 5. At

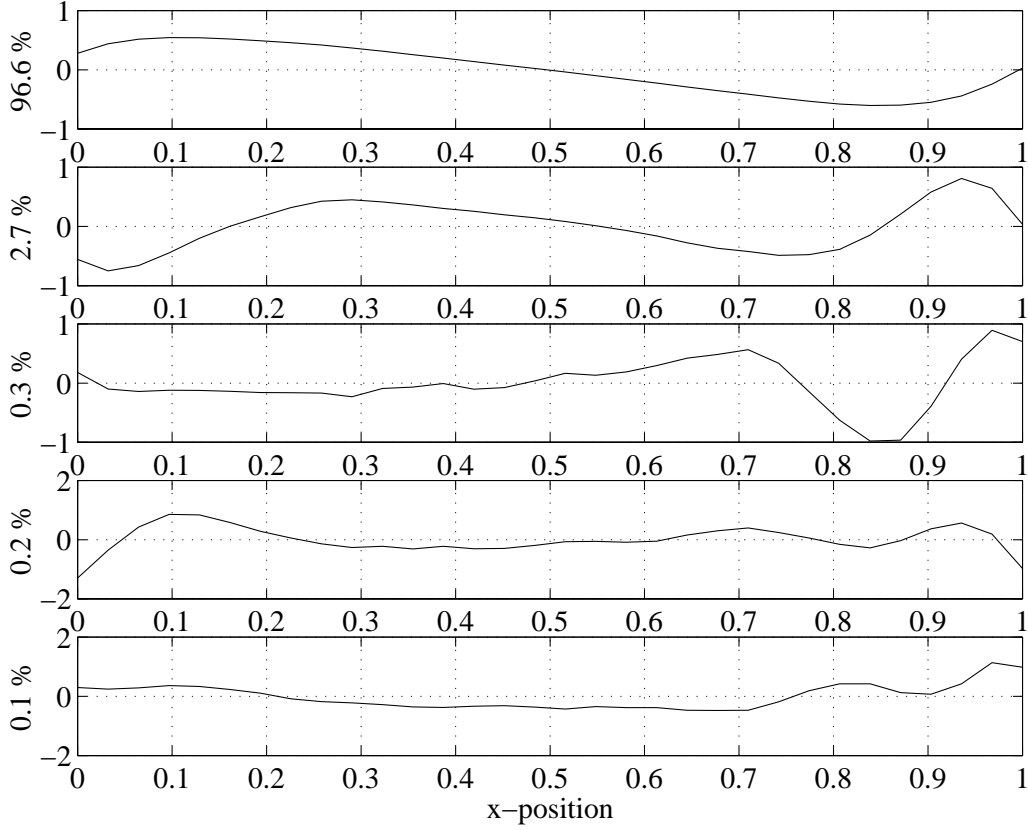


Figure 4: The first five spatial modes of surface height, obtained by Proper Orthogonal Decomposition. The percentages denote the amount of energy captured by each mode.

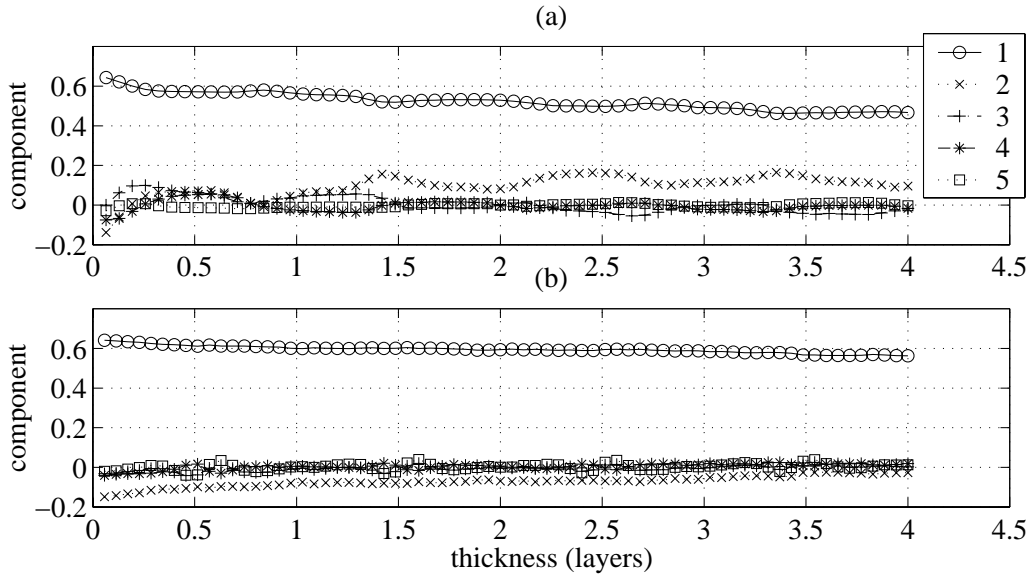


Figure 5: The size of the projection of the first five POD modes onto the snapshots (a) 950 K, (b) 1150 K (shown in Figure 3).

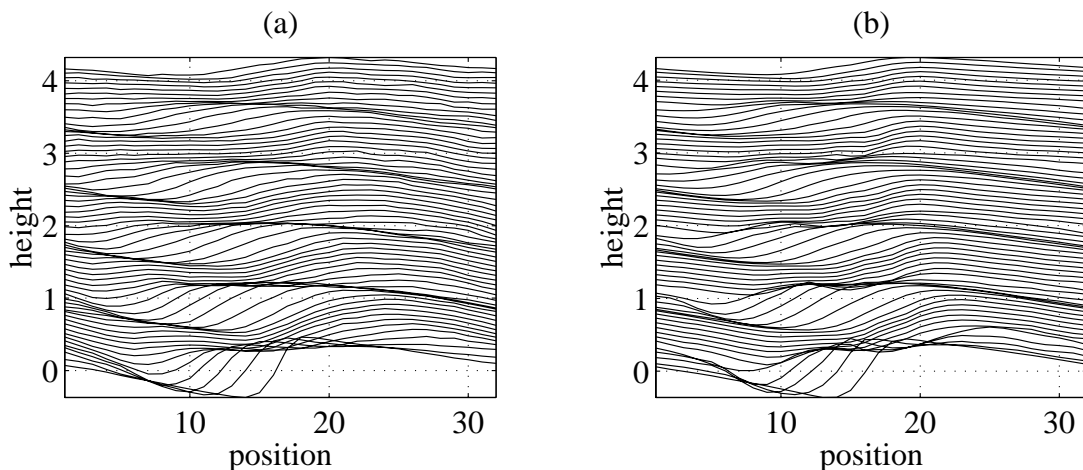


Figure 6: Surface height as a function of position perpendicular to the step, from a KMC simulation at 1050 K with oscillating growth conditions: (a) original snapshots, (b) reconstructed surface height.

the low temperature, the first mode decreases during growth while the second mode increases. This represents an overall roughening of the surface. In addition, oscillations in the second mode occur once per monolayer, signaling that the spatial height profile is oscillating near the monolayer frequency. The higher modes do not appear to contribute significantly to the spatial distribution. At the high temperature, when step flow dominates, oscillations are not evident. The sharp sawtooth-like shape of the step becomes smoother as the step becomes more disordered, as seen in a decrease in the first mode and an increase in the second mode.

COMPARISON TO OTHER SNAPSHOTS

We have demonstrated that the POD method generates modes in which the first few modes capture the majority of the KMC spatial height profile. Next we test these modes on the spatial profiles of a KMC simulation associated with an intermediate temperature, 1050 K, at which both islands and steps grow. The new snapshots are shown in Figure 6(a). In addition, we vary the temperature and partial pressure sinusoidally near the monolayer frequency, with amplitudes of 25 K and 0.95 Pa. We might expect that different spatial modes would be present in such a case. By projecting these profiles onto the previously-computed modes, we test whether or not the previously computed modes represent overall spatial features of film growth on steps.

We project the spatial height profiles of the new simulation onto the previously computed POD modes. The energy captured by the first five modes is 94.7%. While it is not as good as the 99.8% captured for the original data set, most of the energy is captured. This is quite remarkable considering that the spatial profiles contain different features. Because both islands and steps contribute to growth, there is a stationary feature, the islands, and a traveling feature, the steps, in the spatial profile. Template fitting cannot align both features, so a traveling wave is present in the final snapshots. During the early stages of growth, the first, second, third, and fifth modes

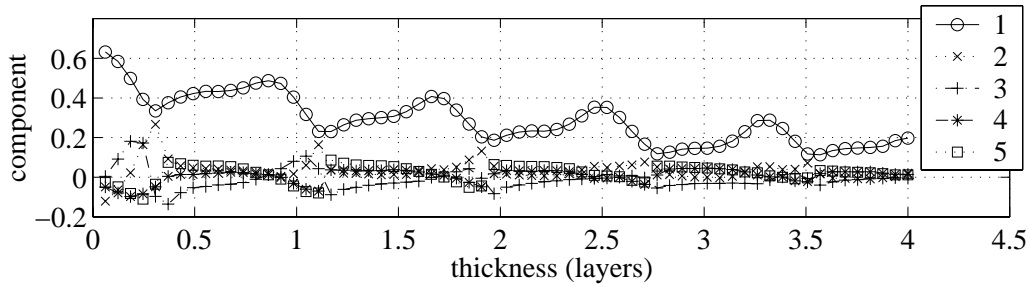


Figure 7: The size of the projection of the first five POD modes onto the snapshots in Figure 6(a).

all appear significantly in the response, as shown in Figure 7. The second, third, and fifth modes contain spatial oscillations near the right side which when combined represent the traveling wave. Even though these modes did not play a large role in the spatial signals from which they were derived, they were captured and play a significant role for other growth conditions. The reconstructed spatial profiles are shown in Figure 6(b). They appear very similar to the original spatial profiles.

CONCLUSIONS

We studied the surface morphology of film growth on a vicinal surface with a kinetic Monte Carlo simulation. To better understand the role of spatial non-uniformities, we searched for characteristic shapes of the surface height. The Proper Orthogonal Decomposition technique produced spatial modes from the kinetic Monte Carlo data. These modes captured 99% of the spatial height profiles for the data used to generate the modes. For simulations performed at another growth condition, the modes captured 95% of the profile.

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