Modeling and Control of Thin Film Morphology Using Unsteady Processing Parameters: Problem Formulation and Initial Results

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Abstract

Thin film deposition is an industrially-important process to which control theory has not historically been applied. The need for control is growing as the size of integrated circuits shrinks, requiring increasingly tighter tolerances in thin film manufacture. In this work we formulate a lattice model of film growth as a control system and take the process parameters as inputs. In the evolution equation, nonlinear functions of the process parameters multiply linear vector fields, yielding a structure similar to a bilinear system.

The process conditions in some deposition methods are inherently unsteady, which produces films with altered morphology. We use the model developed in this study to analyze the effects of fast periodic forcing on thin film evolution. With the method of averaging we develop new effective transition rates which may produce film properties unattainable with constant inputs. These effective rates are the convex hull of the set of rates associated with constant inputs. We present conditions on the convex hull for which the finite-time and infinite-time reachability sets cannot be expanded with fast periodic forcing. An example in which this forcing increases the reachability set and produces more desirable morphology is also presented.

1 Introduction

The deposition of thin layers of material is critical in the manufacture of integrated circuits and other devices. This process, referred to as *thin film deposition*, occurs when precursor material in the gas phase impinges on a solid surface. The precursors react with the surface, resulting in the deposition of a film. The process parameters under which a thin film is deposited must be tightly controlled to prevent defects in the crystal structure, since device performance is strongly dependent on the microstructural properties of the film. Extensive research has been devoted to the control of process variables, such as reactor temperature [8], but not to the film properties which directly determine device performance. The process variables are often controlled about a setpoint, not because it yields the best films, but because the high dimension of film growth models precludes the straightforward application of most established control techniques. Simplified models of growth exist, but usually assume that the process parameters are fixed, neglecting the actuators.

In this study we consider the control of thin film morphology. Morphology refers to the surface height profile, which evolves as the film grows. In the fabrication of a layered structure, a perfectly flat surface is usually optimal. Alternatively, small clusters of atoms on the surface of a film are desired for the manufacture of quantum dots.

Thin film deposition usually occurs under constant process conditions, although some methods are inherently unsteady — one example is pulsed laser deposition (PLD). In PLD, the wafer is placed near a target composed of precursor material. A laser is pointed at the target, heating the target and evaporating the precursor. As the evaporated precursor impinges on the substrate, a film is grown. Because the laser is quickly pulsed, an unsteady flux of precursor material reaches the substrate. It has been observed that the morphology of films grown by PLD differs from the morphology of films grown under steady processing conditions [4, 11]. Recently, many other strategies have been proposed within the physics community to alter film evolution with unsteady flux and temperature [2, 6, 9, 10].

We approach the control of thin film morphology in a different way, from the perspective of control theory. Concepts developed with control theory,

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such as stability, trajectory tracking, and optimal control, could provide more complete answers than ad hoc physical arguments. The primary challenge is to formulate the dynamics of thin film growth in a way to which control theory can be applied. In this study we consider a lattice model of thin film growth. Although the dimension is high, the structure is simple. We exploit this structure to study the response of thin film morphology to fast periodic processing parameters, and show that the reachability set associated with constant inputs may be expanded with unsteady inputs.

2 Lattice model of thin film growth

A thin film may consist of only a few atomic layers, so atomic scale features strongly influence device performance. Thus, our model of thin film growth must contain atomic scale phenomena. Molecular dynamics is one such representation, in which each atom is treated as a point mass connected to neighboring atoms by springs. The time scales associated with atomic motion are of the order of picoseconds, as atoms vibrate within potential energy wells. The molecular dynamics model captures these vibrations, as well as much less frequent events in which an atom overcomes an energy barrier and moves to a new well. It is the latter type of event which contributes to film growth, as atoms move between sites on the crystal lattice.

The time scale of film growth is in minutes or hours, not picoseconds. To capture the dynamics on this time scale, we instead choose a lattice representation of film growth. The motion of atoms between sites is recovered without resolving atomic vibrations. The rates at which the atoms transition between sites are referred to transition rates, and are obtained through molecular dynamics simulations, transition state theory, or experiment. We refer to the transition rates as k_i, where $i = 1 \dots m$, and m is the number of distinct types of transitions.

The key feature of a lattice model is the rigid lattice to which atoms are constrained. This lattice represents the actual crystal structure of the material. At any particular time, a lattice site may or may not contain an atom. As a result, the state of the lattice is completely described by σ, an M-dimensional vector, where M is the number of lattice sites. If the j^{th} lattice site is occupied, then $\sigma_j = 1$, otherwise $\sigma_j = 0$. The number of possible configurations of the system is therefore 2^M. We refer to a configuration as H, with $\{H_l\}$ the set of all configurations. Film growth is an inherently stochastic process, so we express the state of the system not by the current configuration, but by the probability of each of the various configurations.

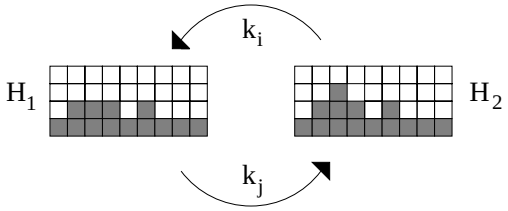

Figure 1: Two configurations in a two-dimensional lattice associated with a one-dimensional surface.

Possible atomic transitions include surface diffusion, in which an atom moves between adjacent lattice sites; adsorption, in which an atom in the surrounding gas takes a site on the lattice; and desorption, in which an atom leaves a lattice site and returns to the gas. As a result of the transitions, the configuration probabilities evolve over time. These transition rates are strongly dependent on the processing conditions, so we take the processing parameters as our actuators.

Consider, for example, the configurations shown in Figure 1. The two configurations differ by a single site, which is not occupied in H_1 and is occupied in H_2. A transition from H_1 to H_2 occurs when an atom is adsorbed from the gas into that site. Similarly, a transition from H_2 to H_1 occurs when the atom desorbs back into the gas. The rates at which these transitions occur are specified by the transitions rates k_i and k_j. Note that two arbitrary configurations will generally not be able to transition directly between each other.

The dynamics associated with the configuration probabilities are described by a "master equation" [5]. The state of this system is the vector of configuration probabilities, which we will refer to as x, and the resulting differential equation is

$$\dot{x} = \sum_{i=1}^{m} k_i(u) N_i x, \tag{1}$$

where $x \in \mathbb{R}^n$, $u \in \mathbb{R}^m$ is the input vector, and $N_i \in \mathbb{R}^{n \times n}$. Note that this system is linear in the state. We take as inputs the processing parameters, which alter the transition rates k_i. The constant matrices N_i contain the information regarding allowable transitions between states. Most elements of N_i are zero, meaning that most configurations cannot transition between each other. Each diagonal element of N_i is negative if there is

any transition out of that configuration; otherwise it is zero. Off-diagonal terms represent allowable transitions into a configuration from another configuration and are never negative. The columns of N_i always sum to 0, which must be the case because the probabilities must always sum to 1. Thus, the system evolves on a hyperplane of dimension $n-1$.

A corresponding linear output equation may be formulated if the outputs are expected film properties. Any property R, such as thickness or roughness, may be computed for each configuration H_l to obtain $R(H_l)$. The expected value is the sum over l of the product of the probability of H_l and $R(H_l)$. Thus, the outputs are linear in the state:

$$y = Cx, \tag{2}$$

where p is the number of outputs and $C \in \mathbb{R}^{p\times n}$.

The system described by (1) and (2) belongs to the class of positive systems [3]. These systems evolve in the positive orthant, which must be the case here since probabilities cannot be negative. Additionally, the inputs u and transition rates $k_i(u)$ are always positive, so that we can only move along the vector fields $N_i x$ in the positive direction. The system (1) and (2) also resembles a strictly bilinear system; if the transition rates $k_i(u)$ can be independently controlled, it is a strictly bilinear system.

To consider a realistically-sized system, we need a lattice containing many sites, for example, a three-dimensional lattice of $M = 100 \times 100 \times 100$ sites. Remembering that the number of configurations is 2^M, and the state dimension is the number of configurations, the dimension of (1) is so large as to make simulation impossible. We may reduce this dimension by making symmetry arguments and physical approximations, but the state dimension remains a prohibitively large number.

Instead of directly simulating (1) and (2), we may perform individual stochastic realizations with the Monte Carlo method, in which a lattice is defined and initialized with atoms at particular lattice sites. This surface evolves by the random execution of transition events, with probabilities proportional to the transition rates. An example of a Monte Carlo simulation is shown in Figure 2. Although Monte Carlo simulations are typically equilibrium calculations, a variant called kinetic Monte Carlo (KMC) captures the correct evolution in time [5]. KMC simulations are tremendously useful in predicting film growth, but they do not have the structure typically required for control analysis.

We have formulated the lattice model of thin film growth in the language of the controls community. To understand how to control this system, we need to determine a number of properties, including (1) the controllability of the system, (2) the reachable set from a particular initial condition, and (3) the existence of a reduced order representation. These results may depend on either the positive or bilinear properties of equations (1) and (2). In this study we present an initial result for system inputs which are periodic with high frequency.

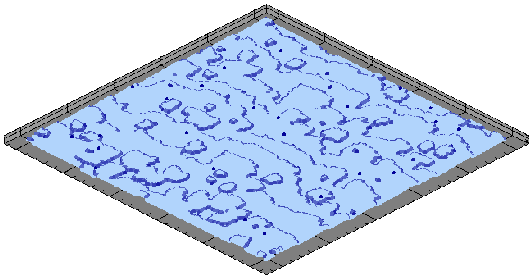

Figure 2: A KMC simulation of thin film growth on a surface with steps. The surface has 256 × 256 sites. The dark points denote atoms with at least one empty side bond.

3 Averaging applied to film growth

In this study we apply the method of averaging with two goals in mind: (1) to explain why fast periodic processing parameters yield altered morphology and (2) to exploit unsteady growth conditions to achieve morphology unattainable with constant growth conditions. The method of averaging may be applied to any system of the form

$$\dot{x} = \epsilon f(x,t), \tag{3}$$

where $x \in \mathbb{R}^n$, $f : U \times \mathbb{R}^1 \mapsto \mathbb{R}^n$, $f \in C^2$, and $U \subset \mathbb{R}^n$. If f is periodic with period τ, we can define its average $\bar{f}$ as

$$\bar{f}(y) = \frac{1}{\tau}\int_0^\tau f(y,t)dt, \tag{4}$$

and the averaged equation as

$$\dot{y} = \epsilon \bar{f}(y), \tag{5}$$

where $y \in \mathbb{R}^n$. The averaging theorem states that (5) approximates (3) when ϵ is small [12]. Specifically,

$$|x(t) - y(t)| = O(\epsilon) \tag{6}$$

on a time scale $O(\epsilon)$ if $|x(t_0) - y(t_0)| < O(\epsilon)$ and $y(t_0) \in U$.

The averaging theorem may be applied to the master equation (1) when the inputs are periodic, i.e. $u = u(\omega t)$ with frequency $\omega = \frac{2\pi}{\tau}$. To obtain an equation of the form (3) we first rescale time by the input frequency ω. Defining a new time $s = \omega t$ and $\epsilon = \frac{1}{\omega}$, the master equation becomes

$$\frac{dx}{ds} = \epsilon \sum_{i=1}^{m} k_i(u(s)) N_i x, \tag{7}$$

which is consistent with (3). When the input frequency is high, ϵ is small and the master equation is approximated by its averaged version,

$$\dot{x} = \sum_{i=1}^{m} \left(\frac{1}{\tau} \int_0^\tau k_i(u(\omega t)) \right) N_i x \tag{8}$$

on a time scale of $O(\frac{1}{\omega})$ in the rescaled time s, and a time scale of $O(1)$ in the physical time t. Similar application of the averaging theorem to mechanical systems may be found in [1].

The form of the original equation (1) is identical to the averaged version (8), except that the functions $k_i(u)$ are replaced by their average values. Physically, this means that the original transition rates are replaced by new "effective" transition rates, $k_{\text{eff},i}$, such that

$$k_{\text{eff},i} = \frac{1}{\tau} \int_0^\tau k_i(u(\omega t)) dt \tag{9}$$

In the remainder of this study we explore the range of effective rates which may be achieved with fast, periodic forcing of the inputs, and compare this set to the set of transition rates associated with constant inputs. Specifically, we consider the set of rates achievable with constant inputs in the range $u_{i,\min} \le u_i \le u_{i,\max}$, and compare this set to the set of all effective rates achievable with periodic inputs such that $u_{i,\min} \le u_i(t) \le u_{i,\max}$. We note that under these constraints, no single effective rate $k_{\text{eff},j}$ will lie outside the set of k_j attainable by constant input, as guaranteed by the mean value theorem. However, the set of rates $\{k_{\text{eff},i}\}$ may be unattainable by constant inputs, particularly when $q < m$, i.e. the number of inputs is less than the number of transition rates.

Because the system is linear in the state, we may view its evolution as the flow along linear combinations of the vector fields $N_i\, x$. When the inputs are constant or fast periodic, these vector fields are multiplied by the constant scalars $k_{\text{eff},i}$, where $k_{\text{eff},i} = k_i$ for the case of constant inputs. We may view this evolution as the evolution of a linear time-invariant autonomous system $\dot{x} = Nx$, where $N = \sum_{i=1}^{q} k_{\text{eff},i} N_i$. The trajectory along which the system evolves is determined only by the relative amounts of each vector field. The speed at which the system travels along the trajectory is determined by the magnitude of the rates. Defining this speed v as $(\sum_{i=1}^{q} k_{\text{eff},i}^2)^{1/2}$, we write N as $N = v \sum_{i=1}^{q} \frac{k_{\text{eff},i}}{v} N_i$. Inputs associated with the same set of coefficients $\{\frac{k_{\text{eff},i}}{v}\}$ produce motion in the same direction in "rate space", the set of $\{k_{\text{eff},i}\}$. Equivalently, they move along the same trajectory, with speed v also determined by $\{k_{\text{eff},i}\}$.

In this study we compare the reachability sets associated with constant inputs to the sets which include fast periodic forcing and show that an unsteady input may expand the reachability set, either for finite or infinite time. If a new direction in rate space is attainable, then the infinite-time reachability set may be expanded. If no new direction is possible, but the speed along an existing direction can be increased, then the finite-time reachability set may be expanded with fast periodic forcing.

4 Piecewise constant inputs

We now consider the case of fast periodic forcing in which the input is a periodic piecewise constant function. Representing the effective rate as a sum instead of an integral, equation (9) becomes

$$k_{\text{eff},i} = \frac{1}{\tau} \sum_{i=1}^{r} k_i(u_r)(\alpha_i \tau) = \sum_{i=1}^{r} \alpha_i k_i(u_r), \tag{10}$$

where α_i is the fraction of period τ spent at input u_i and r is the number of piecewise constant segments. By definition $\sum_{i=1}^{r} \alpha_i = 1$.

We describe the set of effective rates in terms of a convex hull. The convex hull of a set S in $\mathbb{R}^n$ is the smallest convex set containing S [7]. It may equivalently be described as

$$\text{con}\, S = \cup_{1 \le m < \infty} \text{con}_m\, S, \tag{11}$$

where $x \in \text{con}_m\, S \Leftrightarrow x = \sum_{i=1}^{m} \alpha_i s_i$ for some $s_i \in S$ and $\alpha_i \ge 0$ with $\sum_{i=1}^{m} \alpha_i = 1$.

It is clear from equation (10) that the set of all effective rates is the convex hull of the set of all constant rates when r is arbitrarily large. Defining $k = \{k_1, \ldots, k_m\}$ and $k_{\text{eff}} = \{k_{\text{eff},1}, \ldots, k_{\text{eff},q}\}$,

$$\{k_{\text{eff}}\} = \text{con}\,\{k\}. \tag{12}$$

Table 1: Transition rates for Models 1 and 2.

Model 1:	$k_1(u_1,u_2) = u_1$
	$k_2(u_1,u_2) = \nu_2 \exp\left(-\frac{E_2}{k_b u_2}\right)$
	$k_3(u_1,u_2) = \nu_3 \exp\left(-\frac{E_3}{k_b u_2}\right)$
Model 2:	$k_1(u_1,u_2) = u_1$
	$k_2(u_1,u_2) = \nu_2 \exp\left(-\frac{E_2}{k_b u_2}\right)$
	$k_3(u_1,u_2) = u_1\nu_3 \exp\left(-\frac{E_3}{k_b u_2}\right)$

We next quantify $r_{\min}$, the minimum number of piecewise constant inputs u_r required to reach any point in $\operatorname{con}\{k\}$. Carathéodory's theorem provides a well known upper bound on this value [7]. It states that if $X \subset \mathbb{R}^n$ and $p \in \operatorname{con} X$, then $p \in \operatorname{con} Y$ for some $Y \subset X$ with $\operatorname{card}(Y) \leq n+1$. Thus, any point in $\operatorname{con}\{k\}$ can be obtained with $m+1$ different values of the piecewise constant input, where m is the number of transition rates; therefore, $r_{\min} = m+1$. Any k_{eff} attainable with fast periodic forcing can be obtained with a fast periodic input in which each period consists of $m+1$ piecewise constant segments.

5 Two illustrative examples

To demonstrate the effect of fast periodic forcing we consider two transition rate models. Each consists of $m = 3$ transition rates and $q = 2$ inputs. The functional forms of the transition rates determine the extent to which the effective rates differ from the actual rates. The inputs u_1 and u_2 correspond to the physical variables of flux and temperature, respectively. We place the following constraints on the inputs which apply to both constant and periodic inputs: $u_{1,\min} = 0$ s^{-1}, $u_{1,\max} = 1$ s^{-1}, $u_{2,\min} = 300$ K, and $u_{2,\max} = 800$ K.

We first consider a model in which atoms attach to the surface with a rate proportional to the incoming flux of material. If an atom has no side bonds, it may also diffuse along the surface or desorb back into the gas with rates dependent only on the surface temperature. The second model differs only slightly from the first; k_1 and k_2 are unchanged, but the desorption process k_3 differs by a factor of u_1. In Model 2, a species in the gas adsorbs at high temperature and subsequently etches away atoms on the surface; the rate is a function of *both* inputs. The transition rates for Models 1 and 2 are given in Table 1, where E denotes an activation energy for a thermally-activated process, k_b is Boltzmann's constant, and ν refers to a vibrational frequency.

We first focus on Model 1. The surface in Figure 3 is the set of transition rates achievable with constant inputs within the input constraints and for the physical parameters given in Table 2. Because there are two inputs, this set of constant rates is a two-dimensional surface. Note that k_1 is only a function of u_1 while k_2 and k_3 are functions only of u_2. Thus, k_1 may be set to any desired value within the allowable limits, independent from the other rates. Rates k_2 and k_3 depend on the single input u_2 and cannot be independently selected.

Table 2: Parameters for Models 1 and 2.

Parameter	Model 1	Model 2
$\frac{E_2}{k_b}$(K)	17000	16000
$\frac{E_3}{k_b}$(K)	8000	14000
ν_2(s^{-1})	1.6934×10^{12}	4.8517×10^{11}
ν_3(s^{-1})	2.2026×10^{7}	3.9825×10^{11}

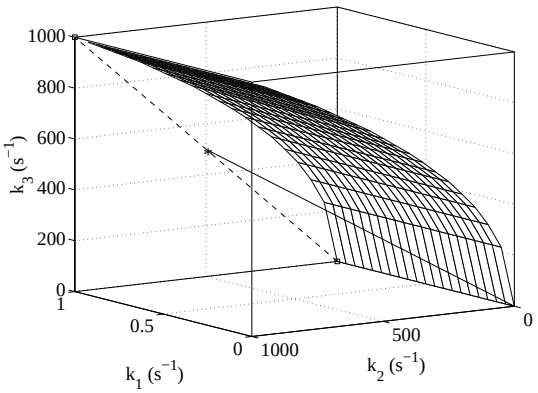

Figure 3: Model 1: Surface of transition rates attainable with constant inputs. '$*$' denotes an effective set of rates achievable only with fast periodic inputs.

Fast periodic inputs may be used to achieve a combination of effective rates which is not attainable with constant inputs. Consider the point $(1, 500, 500)$ in rate space, shown in Figure 3. This point is denoted with an asterisk and represents a periodic piecewise continuous input in which the period is divided evenly between $(u_{1,\max}, u_{2,\max})$ and $(u_{1,\max}, u_{2,\min})$. This effective input yields a direction in rate space which cannot be achieved with constant rates, and represents a decrease in desorption relative to diffusion. This might be desired to reduce the material lost during desorption while retaining a smooth film. However, these effective rates do not necessarily yield a system output y which is unattainable with constant inputs. The ultimate reachability of the system also de-

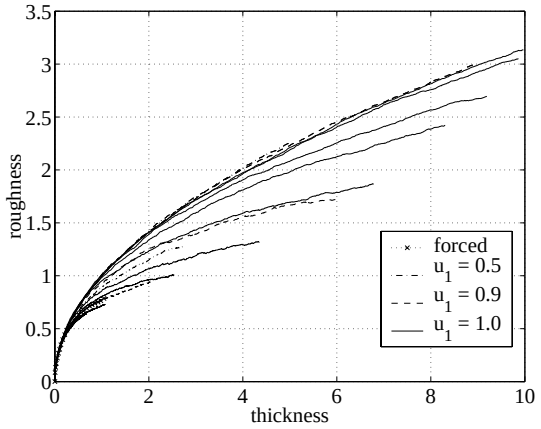

Figure 4: KMC simulations on a one-dimensional 5000-site surface for the transition rates of Model 1, 10 s of growth, and a representative range of constant inputs.

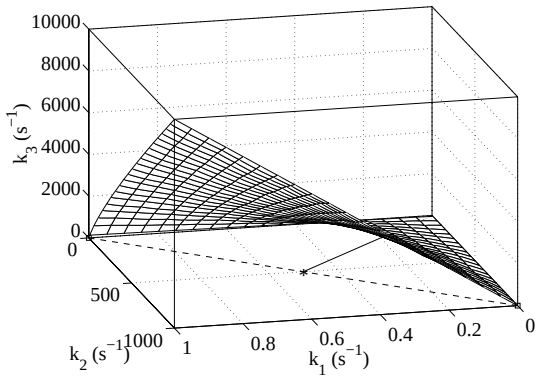

Figure 5: Model 2: Surface of transition rates attainable with constant inputs. '$*$' denotes an effective set of rates achievable only with fast periodic inputs.

pends on the system and output matrices N_i and C. We choose as our output the film thickness and root-mean-square (RMS) roughness. Because both diffusion and desorption smooth the surface, and because the new point in rate space is close to the constant surface, the thickness and RMS roughness associated with the effective rates may also be obtained with constant inputs, as shown in Figure 4 with KMC simulations. The output produced by the periodic inputs is inside the finite-time reachability set for constant inputs and 10 s of growth.

The effective rate vectors in the convex hull of the constant rate surface of Model 1 do not differ greatly from the constant rates themselves. Unlike Model 1, the functional form of the rates in Model 2 enables effective rate vectors in a significantly different direction. The surface in rate space associated with constant input for Model 2 is pictured in Figure 5. Because desorption k_3 is a monotonic function of both inputs, the surface remains near zero along both the k_1 and k_2 axes. As a result, we may connect a point on the k_1 axis to a point on the k_2 axis to get a point in the convex hull at which adsorption and diffusion are significant but desorption is suppressed. We consider a point associated with a piecewise constant function which spends half the period at $(u_{1,\max}, u_{2,\min})$ and the remaining half at $(u_{1,\min}, u_{2,\max})$, and yields an effective rate of $(0.5, 500, 0)$, as shown in Figure 5.

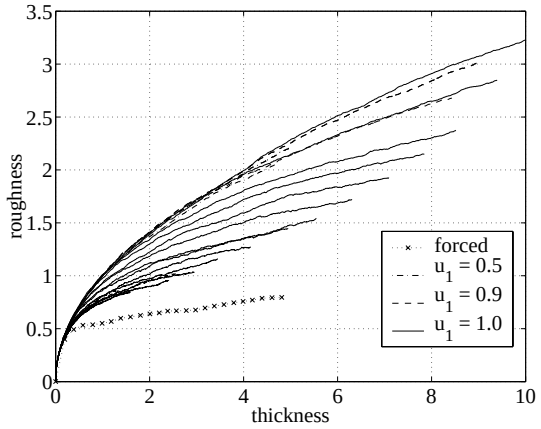

Figure 6: KMC simulations on a one-dimensional 5000-site surface for the transition rates of Model 2, 10 s of growth, and a representative range of constant inputs.

This effective rate enables an expansion of the reachable set over the constant input case, as pictured in Figure 6. Under constant growth conditions, the flux must be high to obtain a high growth rate. This flux causes the surface to roughen, unless the temperature is raised to allow smoothing through desorption and diffusion. However, at high temperature, material is lost back to the gas via etching, which decreases the growth rate. Under periodic conditions, the situation is different. Material is deposited at low temperature, which limits etching and diffusion. The flux is then turned off and the temperature raised. At this point the atoms diffuse, but cannot be etched due to lack of flux. The surface is ultimately able to reach a point after 10 seconds of growth in which the surface is less rough for a particular thickness, shown in Figure 6. Equivalently, the point in output space associated with a fast periodic input is not in the finite-time reachability set for constant inputs.

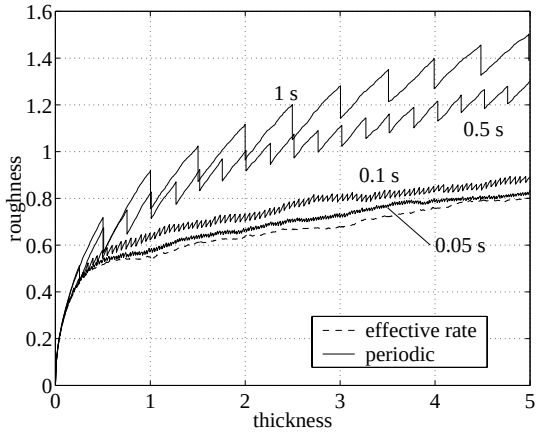

Figure 7: KMC simulations of thin film growth on a one-dimensional 5000-site surface for the transition rates for Model 2 and for 10 s of growth.

We have shown the reachable set associated with constant inputs may be expanded with fast periodic inputs. We have not, however, defined "fast". The input period must be faster than the response time of the system to achieve the limit of averaging theory. This response time depends on the particular system. We consider again as an example the system associated with Model 2 and vary the input period in KMC simulations. The results are shown in Figure 7. For a period of $\tau = 0.1$ s the response is near the response associated with the effective rates, and for a period of $\tau = 0.05$ s, the responses are indistinguishable within the noise level of the simulations. For this particular system, the required input frequency is only 10-20 Hz.

6 Conclusions

We have formulated a model of thin film growth in the input-output framework of control theory. To demonstrate the utility of this formulation, we apply the results of averaging and convex analysis to study the effects of fast periodic forcing on film morphology. We have shown that all of the new behavior produced by fast periodic forcing can also be produced by a periodic function with only a finite number of piecewise constant segments. The maximum number of segments is equal to one plus the number of transition rates.

An analysis of the transition rates may indicate new directions in which the system can evolve, and does not require knowledge of the high-dimensional state matrices. Effective transition rates obtained with fast periodic forcing are defined by the convex hull of the set of transition rates achievable with constant input. These effective rates may enable combinations of rates not possible with constant input. If no new directions in rate space are possible, then the infinite-time reachability set cannot be expanded. If in addition the length of this vector is never greater than the parallel vector associated with constant inputs, then the finite-time reachability set cannot be expanded either. Conversely, new directions in rate space available through periodic forcing may suggest periodic inputs that expand the reachability set.

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