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# Evolution of Thin Film Morphology

## Modeling and Simulations

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# Preface

Thin film deposition is the most ubiquitous and critical of the processes used to manufacture high-tech devices such as microprocessors, memories, solar cells, microelectromechanical systems (MEMS), lasers, solid-state lighting, and photovoltaics. The morphology and microstructure of thin films directly controls their optical, magnetic, and electrical properties, which are often significantly different from bulk material properties. Precise control of morphology and microstructure during thin film growth is paramount to producing the desired film quality for specific applications. To date, many thin film deposition techniques have been employed for manufacturing films, including thermal evaporation, sputter deposition, chemical vapor deposition, laser ablation, and electrochemical deposition.

The growth of films using these techniques often occurs under highly nonequilibrium conditions (sometimes referred to as far-from-equilibrium), which leads to a rough surface morphology and a complex temporal evolution. As atoms are deposited on a surface, atoms do not arrive at the surface at the same time uniformly across the surface. This random fluctuation, or noise, which is inherent to the deposition process, may create surface growth front roughness. The noise competes with surface smoothing processes, such as surface diffusion, to form a rough morphology if the experiment is performed at a sufficiently low temperature and / or at a high growth rate. In addition, growth front roughness can also be enhanced by growth processes such as geometrical shadowing. Due to the nature of the deposition process, atoms approaching the surface do not always approach in parallel; very often atoms arrive at the surface with an angular distribution. Therefore, some of the incident atoms will be captured at high points on a corrugated surface and may not reach the lower valleys of the surface, resulting in an enhancement of the growth front roughness. A conventional statistical mechanics treatment cannot be used to describe this complex growth phenomenon and as a result, the basic understanding of the dynamics of these systems relies very much on mathematical modeling and simulations.

The present monograph focuses on the modeling techniques used in research on morphology evolution during thin film growth. We emphasize the mathematical formulation of the problem in some detail both through numerical calculations based on Langevin continuum equations, and through Monte Carlo simulations based on discrete surface growth models when an analytical formulation is not convenient. In doing so, we follow the conceptual advancements made in understanding the morphological evolution of films during the last two and half decades. As such, we do not intend to include a comprehensive survey of the vast experimental works that have been reported in the literature.

An important milestone in the mathematical formulation used to describe the evolution of a growth front was presented more than two decades ago. This concept is based on a dynamic scaling hypothesis that utilizes an elegant model called self-affine scaling. Since then, numerous modeling, simulation, and experimental works have been reported based on dynamic scaling. Several books published recently have thoroughly discussed this subject, including *Fractal Concepts in Surface Growth* by A.-L. Barabási and H. E. Stanley (Cambridge University Press, 1995); and *Fractals, Scaling, and Growth Far from Equilibrium* by P. Meakin (Cambridge University Press, 1998). After the publication of these books, the field has grown considerably and the scope has broadened substantially. One of the salient developments is the recognition that films produced by common deposition techniques such as sputter deposition and chemical vapor deposition may not be self-affine, and have characteristics that have not been previously realized. Shadowing through a nonuniform flux distribution, for example, can profoundly affect the film morphology and lead to a breakdown of dynamic scaling. In addition to the common lateral correlation length scale, another length scale emerges called the wavelength that describes the distance between "mounds" that are formed under the shadowing effect. Also, the reemission effect, where incident atoms can "bounce around" before settling on the surface, can significantly change the surface morphology. Reemission is modeled with a sticking coefficient, which describes the probability that an atom "sticks" to the surface on impact. Depending on the value of the sticking coefficient, the morphology can change from a self-affine topology to a markedly different topology where the dynamic scaling hypothesis is no longer valid.

While following these conceptual developments on morphology evolution, the present monograph outlines the mathematical tools used to model these growth effects. The monograph is divided into three parts: Part I: Description of Thin Film Morphology, Part II: Continuum Surface Growth Models, and Part III: Discrete Surface Growth Models. In Part I, we introduce a set of useful statistics and correlation functions that have been utilized extensively in the literature to describe rough surfaces, including the root-mean-square roughness (interface width), lateral correlation length, autocorrelation function, height–height correlation function, and power spectral density function. Self-affine and non self-affine (mounded) surfaces are also introduced, as well as a discussion of the dynamic scaling hypothesis. In Part II, we outline how stochastic continuum equations are constructed to describe the evolution of growth front morphology, and explain the numerical methods that are used to solve these equations. We discuss both local models such as the random deposition model, Edwards–Wilkinson model, Mullins surface diffusion model, and the Kardar–Parisi–Zhang (KPZ) model, in addition to nonlocal models that include effects of shadowing and reemission. In particular, a connection between surface growth models with shadowing and reemission and a small world network model is discussed in detail. In Part III, discrete surface growth models based on Monte Carlo simulation techniques are introduced to describe the morphology evolution of thin films. Various aggregation strategies are described, including solid-on-solid techniques which are often used for relatively thin films, and ballistic aggregation techniques which are used to model thicker films. As an example, we use the results of these models, along with experimental results, to show the breakdown of dynamic scaling under common deposition conditions. Finally, the origin of a particular film impaction called "nodular defects" is discussed based on a ballistic aggregation model.

This monograph is useful for university researchers and industrial scientists working in the areas of semiconductor processing, optical coating, plasma etching, patterning, micromachining, polishing, tribology, and any discipline that requires an understanding of thin film growth processes. In particular, the reader is introduced to the mathematical tools that are available to describe such a complex problem, and lead to appreciate the utility of the various modeling methods through numerous example discussions. For beginners in the field, the text is written assuming a minimal background in mathematics and computer programming, which enables the readers to set up a computational program themselves to investigate specific topics of their interest in thin film deposition. Several of the simulations discussed in the text are implemented in the appendices to aid readers in creating their own growth models, and are also available on the Web at `http://www.stanford.edu/~pellim`.

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Troy, NY
July, 2007

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## Part II Continuum Surface Growth Models

## Part III Discrete Surface Growth Models

# 1

# Introduction

The natural world is filled with rough surfaces. Roughness is, however, a relative term. One may describe a sheet of paper as being smooth to the touch, whereas on an atomic scale one would observe deep valleys and tall mountains in the landscape. Of particular scientific interest in the past few decades have been surfaces that exhibit this rough behavior on a nanometer scale, often referred to as thin film surfaces. Numerous studies have been carried out investigating processes to create thin films, characterize them, and test their physical properties [187]. The physics behind the growth and structure of these surfaces has been shown to be very interesting and challenging due to the complexities of the growth processes and surface structures [8, 40, 104, 112]. Specifically, surface and interface roughness controls many important physical and chemical properties of films. For example, the electrical conductivity of thin metal films depends very much on surface and interface roughness [135], and the reliability of a Si MOSFET (metal-oxide-semiconductor field-effect transistor) channel depends on the roughness of the gate oxide–silicon interface [82]. Also, interface roughness has a profound effect on the magnetic hysteresis of a magnetic film [115], and controls optical losses in optical waveguides [130]. Rough surfaces can increase the effective area for advanced charge storage devices [19], as well as promote capillary forces through wicking in modern heat pipe design [51]. These properties of thin films are exploited in a number of applications, including semiconductor devices [153], solar cells [127], and thin-film transistor (TFT) displays [73].

There are many different experimental methods for growing thin films in the lab, depending on the desired properties of the film. However, all methods accomplish the same general goal; to deposit matter on a substrate. Many deposition methods aim to deposit a specific type of material on a substrate, such as silicon, silicon dioxide, germanium, copper, or tantalum, but other compounds such as organic molecules can also be deposited. In order to create surfaces with nanometer scale roughness, the thickness of the deposited film is generally on the order of micrometers or nanometers, which means the surface must be grown layers of atoms at a time. To accomplish this, the material to

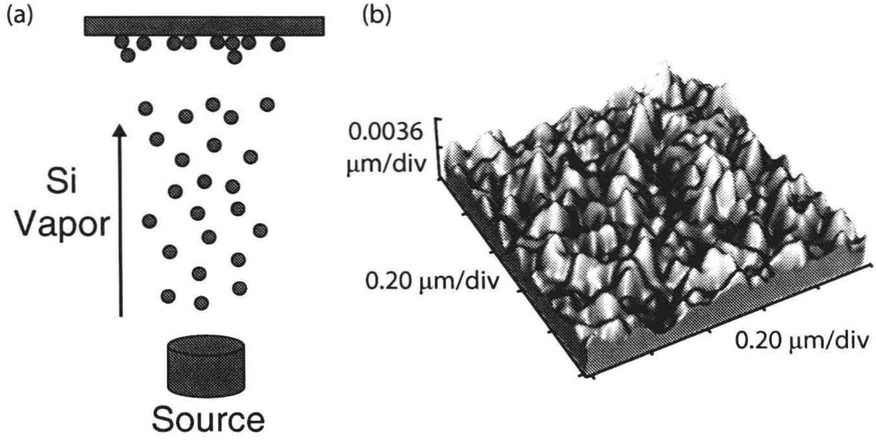

**Fig. 1.1.** (**a**) A schematic showing a thermal evaporation deposition experiment with a Si source. (**b**) An atomic force microscopy image of the surface morphology of a 2 μm thick amorphous Si film grown by thermal evaporation.

be deposited is often changed into a gaseous form in a vacuum to allow for atom-by-atom deposition on the surface.

The simplest deposition method is thermal evaporation [95], where the source material is placed in a crucible and then heated until it evaporates and condenses on a substrate located above the crucible. Figure 1.1a is a schematic drawing showing a thermal evaporation deposition experiment setup with a Si source. Figure 1.1b is an atomic force microscopy image of the surface morphology of a 2 μm thick amorphous Si film grown by the thermal evaporation technique at room temperature. As we can see from the image, the surface contains mountains and valleys over a certain length scale. The topology is obviously quite complex and it cannot be predicted deterministically. It belongs to a class of "complex phenomena" that has been pursued actively by scientists.

Once a thin film has been deposited, we need some way of quantitatively characterizing the surface. To this end, various mathematical tools have been developed that measure the most important properties of a surface, such as the mean height, roughness, and correlation length [187]. In addition, it has been found that many thin film surfaces obey certain common scaling properties that allow for a significant simplification of the description of the surface morphology. The most common such type of scaling is referred to as "self-affine" scaling, in which one can rescale the horizontal and vertical directions of the surface to obtain a new surface that is statistically identical to the original surface [100]. This definition of scaling is reminiscent of a fractal, and the mathematical concepts associated with fractals are used to describe self-affine surfaces. In particular, a self-affine surface is mainly characterized by a roughness exponent, which is related to the local roughness of the surface, but also the fractal dimension of the surface. A similar argument can be made about the scaling behavior of the surface profile in time, which is called "dynamic" scaling [8, 40, 41, 104]. Scaling arguments work quite well when the important growth effects in a deposition are "local", or only affect nearby surface heights, an example of which is surface diffusion, where atoms can diffuse to nearby locations depending on deposition conditions such as activation energy and temperature.

A problem arises when attempting to use self-affine scaling and dynamic scaling to describe thin film surfaces grown under the influence of nonlocal growth effects such as shadowing [123]. By definition, nonlocal growth effects are of much longer range than local effects, and as such are capable of defining a long-range length scale on the surface, often referred to as mound formation [122]. Mounds disrupt the self-affine behavior of the surface because they define a characteristic long-range length scale on the surface. When attempting to rescale the dimensions of the surface as in self-affine scaling, this characteristic length scale changes, and the rescaled surface is no longer statistically identical to the original surface. However, it has been shown that in growth processes that include only local growth effects mounded surfaces can be formed, as evidenced by surfaces created during molecular beam epitaxy [112, 166].

## 1.1 Growth Front Roughness

Many factors contribute to the formation of such a complex landscape on the surface of a film. First, there is always random noise that exists naturally during the deposition process because atoms do not arrive at the surface uniformly. These random fluctuations, which are inherent in the deposition process, can create growth front roughness. Noise competes with surface smoothing processes, such as surface diffusion, to form a rough morphology if the experiment is performed at either a sufficiently low temperature and / or at a high growth rate. In addition, growth front roughness can also be enhanced by growth processes such as geometrical shadowing. Shadowing is a result of deposition by a nonnormal incident flux [11, 62, 92, 106]. In many commonly employed deposition techniques such as sputter deposition [97, 144] and chemical vapor deposition [6, 31], atoms do not always approach the surface in parallel; very often they arrive at the surface with a distribution of trajectories. Figure 1.2 shows schematically the geometries of several commonly employed deposition techniques [92]. The angle $\theta$ is defined as the angle between the incident atomic flux and the surface normal. For conventional thermal evaporation or e-beam evaporation, if the substrate is sufficiently far away from the source and if the substrate dimensions are not too large, the flux arrives at the substrate with $\theta \approx 0°$, which is referred to as normal incidence. Oblique angle deposition can be achieved by tilting the substrate with respect to the particle flux in evaporation, and angles as large as $\theta \approx 85°$ are often used

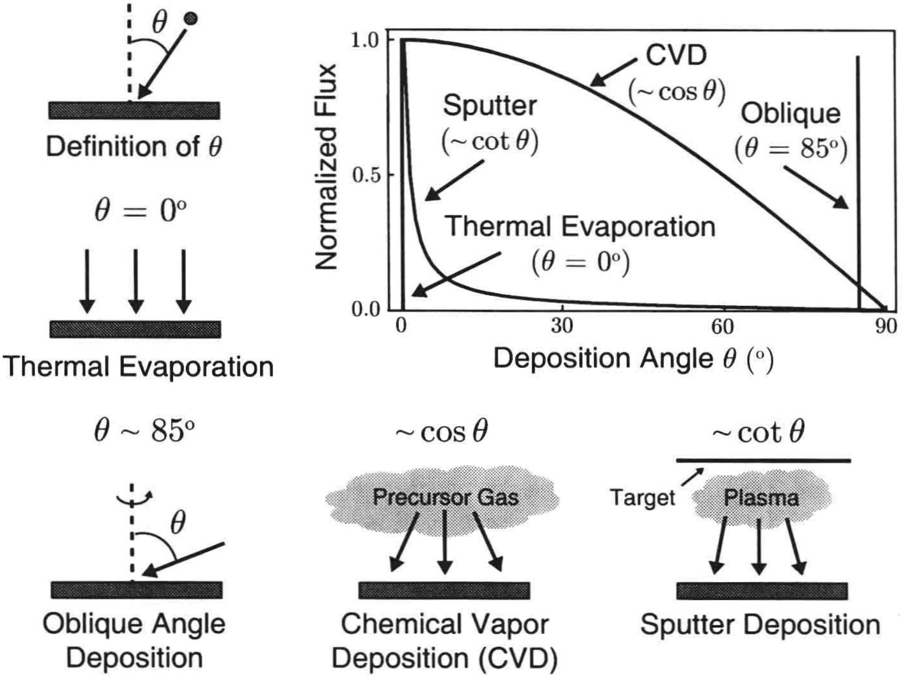

**Fig. 1.2.** Schematic diagrams showing the geometries of several commonly employed deposition techniques. The graph is a plot of the incident flux distribution of atoms arriving at the substrate for different deposition techniques. Depending on the geometry, sputter deposition can also be modeled with a cosine flux distribution [92].

experimentally [57, 76]. For chemical vapor deposition, precursor molecules may bounce around the deposition chamber numerous times before they undergo a reaction at the substrate. Therefore, the substrate experiences a molecular flux coming from a wide range of angles and can be represented by a cosine distribution. For sputter deposition, the distribution can be somewhat narrower (a ratio between cosine and sine functions) but, depending on the separation between the substrate and the source, can also be modeled by a cosine flux distribution. These nonnormal incident fluxes can lead to a shadowing effect during growth, as some of the incident atoms will be captured at high points on a corrugated surface at the expense of lower valleys on the surface, resulting in a dramatic enhancement of the growth front roughness.

Another important effect to consider is the value of the sticking coefficient [92, 184]. The sticking coefficient is defined as the probability that a particle will stick to the surface when it strikes. In both sputter deposition and chemical vapor deposition, the sticking coefficient may not be equal to unity. A nonunity sticking coefficient would allow the particle to be reemitted from

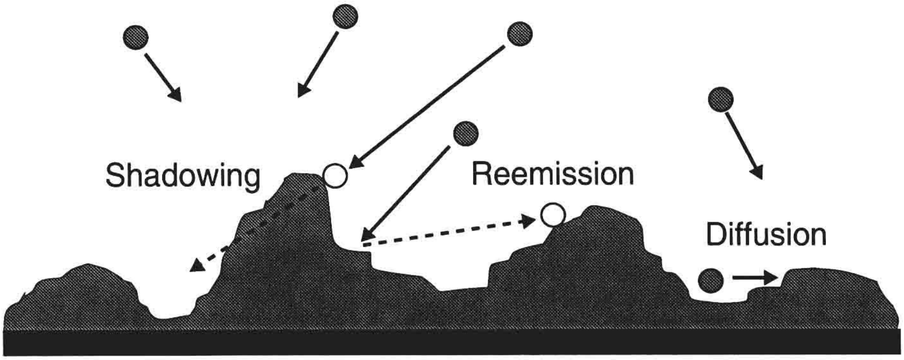

**Fig. 1.3.** Diagram of growth effects including diffusion, shadowing, and reemission that may affect surface morphology during thin film growth. The incident particle flux may arrive at the surface with a wide angular distribution depending on the deposition methods and parameters.

the surface upon impact. The particle may then deposit on the surface at a different location, or it may bounce around the surface more before it settles, which leads to a smoothing effect. Both shadowing and reemission effects are inherently nonlocal because an event that occurs at one place on the surface can affect the surface profile a far distance away. A summary of common growth effects is illustrated in Fig. 1.3.

## 1.2 Measurement Techniques

Before any analysis can be carried out regarding the roughness evolution of a surface, we must utilize measurement techniques that can reliably provide important information about a growth front. There are two classes of techniques that allow for a collection of quantitative information about the morphology of a growth front: real-space imaging techniques, and diffraction techniques [187]. Examples of real-space imaging techniques include atomic force microscopy (AFM), scanning tunneling microscopy (STM), scanning electron microscopy (SEM), and stylus profilometry. Real-space imaging techniques have the advantage of providing a direct visual interpretation of the surface morphology. From the surface profiles, one can extract all surface statistics relating to surface roughness. Examples of diffraction techniques include high-resolution low-energy electron diffraction (HRLEED), reflection high-energy electron diffraction (RHEED), atom diffraction, X-ray diffraction, and light scattering. For diffraction techniques, all surface roughness information can be extracted from the angular distribution of the diffracted radiation. Diffraction techniques have the advantages of providing noncontact measurements and the ability to obtain a statistical average of a large surface area in a short time. Also, some diffraction techniques are capable of, and many have the potential

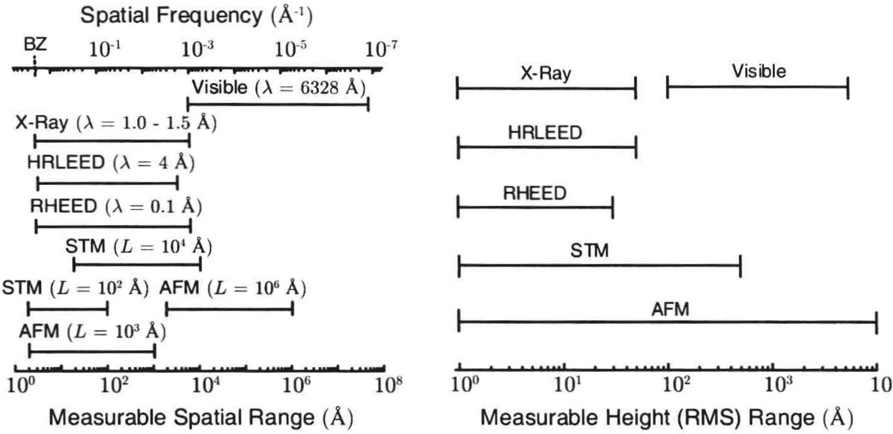

**Fig. 1.4.** Spatial length scale and frequency ranges for different imaging and diffraction techniques. For real-space imaging techniques, $L$ represents the scan size, and for diffraction techniques, $\lambda$ represents the wavelength of radiation used. The location of the Brillouin zone (BZ) is given for a lattice constant of approximately 2 Å, a length characteristic of experimental surfaces.

of, performing real-time measurements during growth or etching. Many of the real-space imaging and diffraction techniques are complementary to each other in the sense that they cover different length scales. Figure 1.4 shows a summary of the range of measurements each technique can cover in the lateral direction in terms of a spatial range, and the vertical direction in terms of the root-mean-square (RMS) roughness. More recently, it was shown that in situ spectroscopic ellipsometry can also provide useful information about the local surface roughness evolution [148]. Because the experimental characterization of growth front roughness is not the focus of this monograph, interested readers are referred to a recent book dedicated to this subject, *Characterization of Amorphous and Crystalline Rough Surface: Principles and Applications* by Y.-P. Zhao, G.-C. Wang, and T.-M. Lu (Academic Press, 2001).

## 1.3 Modeling

The main focus of this monograph is the modeling of thin film surface growth. Thin film growth models can be separated into two main categories: models that are based on continuum mathematics, and models that are based on discrete mathematics. In the past few decades, a number of models of both types have been proposed and have been shown to successfully predict properties of certain types of thin film growth, each with their own advantages and disadvantages. The ultimate utility of any of these theoretical models can be traced back to the core assumptions used to construct the model, which can be quite