

Epitaxial Thin Film Growth

By *Jim W. Evans*

Thin film deposition on semiconductor surfaces underlies microelectronics fabrication technologies. Indeed, it was the promise of the solid-state electronics revolution in the early 1950s that in large part drove the development of modern ultra-high-vacuum (UHV) surface science in the 60s and 70s. Offering additional motivation was the potential of modern surface science to provide a fundamental understanding of catalytic processes on metal surfaces and films. These developments not only enabled the study of surfaces and thin films in well-controlled contaminant-free environments, but also led to a host of experimental probes, including surface-sensitive diffraction for characterizing thin film morphologies in reciprocal space. The 1980s saw additional dramatic advances, with the development of high-resolution scanning probe and electron microscopes capable of providing real-space images of film morphologies down to the nanometer or even atomic scale (for scanning tunneling microscopy).

Thin film deposition continues to be of interest today as an approach for creating functional surface nanostructures in various applications: semiconductor quantum dots for novel optoelectronic devices, and metallic nanostructures for applications to catalysis, plasmonics, magnetic thin films, among many others. Furthermore, the analysis of thin film growth on near-perfect atomically flat surfaces in pristine environments made possible by UHV surface science, combined with extraordinarily detailed images of film morphologies in these “ideal systems,” has driven major efforts and advances of the last 20 years in detailed and realistic modeling of film growth.

An invited talk and a three-part minisymposium at this year’s SIAM Conference on Mathematical Aspects of Materials Science specifically addressed the topic of epitaxial thin film growth. Complementing these sessions were several other minisymposia whose speakers explored mathematical modeling strategies related to epitaxial thin film growth.



In epitaxial growth, atoms are vapor-deposited at very low pressures onto crystalline surfaces that either are perfectly flat or have a simple stepped-terrace morphology. The adsorbed atoms reside on a periodic array or lattice of adsorption sites on the surface, hopping between sites as a result of surface diffusion.

The simplest case of homoepitaxy (A on A) corresponds to growing a single crystal of material A. The equilibrium state of the homoepitaxial film is just a flat surface (for a flat “initial condition”); deposition drives the system far from equilibrium, creating a rich variety of complex rough morphologies (see Figure 1). In fact, basic questions remain regarding morphological evolution even for these simplest of systems. Extensive recent investigations have been driven mainly by the desire to obtain a fundamental understanding of crystal growth [1,2]. With regard to technological applications, it is heteroepitaxy (A on B) that provides a route to the functional nanostructures mentioned earlier. Such systems present additional complications, e.g., strain resulting from lattice mismatch for crystals of A and B.

These well-defined epitaxial thin film systems are amenable to detailed atomistic-level lattice-gas modeling, in the spirit of the discrete lattices of adsorption sites mentioned earlier. Such models are also described by mathematical statisticians as (discrete) stochastic interacting particle systems. Their behavior can be assessed precisely on the appropriate time- and length-scales via kinetic Monte Carlo simulation. Indeed, modeling of this type has achieved remarkable predictive accuracy for selected systems.

At the same time, epitaxial growth systems also provide a natural testing ground for coarse-grained modeling strategies. A major motivation is the potential for enhanced computational efficiency. The process of developing appropriate coarse-grained models actually has the additional benefit of providing new insights into the fundamentals of film growth. However, substantial challenges remain in the development of reliable and rigorous coarse-grained models.

To provide some orientation for a discussion of coarse-grained modeling strategies, we note that deposited atoms often diffuse rapidly and aggregate into large two-dimensional single-atom-high islands within each layer of a growing film. Rather than tracking all atoms within an island or film, it sometimes suffices to track only the evolution of the steps at the island edges within each layer, describing them as continuous curves. This “step-dynamics” modeling strategy, then, uses coarse-graining to obtain a continuous treatment in the lateral direction, but retains vertical discreteness. Step velocities follow from analysis of continuum deposition–diffusion equations on each terrace with appropriate boundary conditions, often described as a Burton–Cabrera–Frank formulation. Such step-dynamics formulations can be implemented via either Lagrangian interface tracking or Eulerian level-set methods. Alternatively, we could artificially smear the sharp steps and use a phase-field formulation to describe their evolution. These different approaches were discussed at the conference in various minisymposia. In the end, basic questions remain—regarding the appropriate treatment of the diffusion of atoms along island edges, of the nucleation of new islands, and so forth.

The step-dynamics approach is natural for relatively smooth films in which just a few atomic layers are exposed, and in which the characteristic lateral dimension (i.e., island size) is large. For films that are quite rough, by contrast, further coarse-graining to a fully continuum formulation may be more effective; in such cases, film morphology is described by a continuous height function $h(x,t)$, which depends on a continuous lateral position x and time t . Evolution is then described by a partial differential equation with the natural form $\partial h/\partial t = F - \nabla \cdot \underline{J} + \eta$, where F denotes the deposition flux, \underline{J} denotes the surface diffusion flux, and the zero-mean noise term η can be included to account for fluctuation effects. The mean height

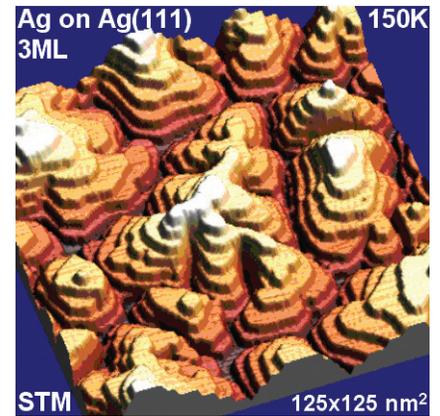


Figure 1. Scanning tunneling microscopy image of a film morphology created by depositing three layers of Ag atoms on a perfectly flat hexagonal close-packed Ag(111) crystalline surface at a temperature of 150 K [1]. Clearly visible are single-atom-high steps bordering 2D islands with fractal shapes, which together form multilayer wedding cake-like “mounds.”

thus grows as $\langle h \rangle = Ft$. Equations of this type have long been used to study surface evolution, often with $F = 0$ and with the form for \underline{J} motivated by quasi-equilibrium ideas of Mullins connecting the surface free energy to local curvature [3]. However, neither the form of \underline{J} nor the form of the noise term is clear for far-from-equilibrium growth. Reasonable phenomenological choices have been made, but a rigorous coarse-graining of atomistic or step-dynamics models to derive the above PDE is at best a partially solved problem.

There are additional challenges specific to heteroepitaxial systems. Strain effects can lead to the spontaneous organization of 3D islands or “quantum dots.” Quantum size effects, produced by the confinement of electrons within metal films, can lead to film height selection and the development of dramatic mesa-like film morphologies. Traditionally, continuum equations have been used to model strained-layer heteroepitaxy. Speakers at this year’s minisymposia offered glimpses into possibilities for more detailed atomistic-level modeling of thin film systems whose growth is impacted by strain and/or quantum size effects.

References

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[2] T. Michely and J. Krug, *Islands, Mounds, and Atoms*, Springer, Berlin, 2004.

[3] W.W. Mullins, *Flattening of a nearly planar solid surface due to capillarity*, J. Appl. Phys., 30 (1959), 77–83.

Jim W. Evans, a professor of mathematics at Iowa State University, gave an invited talk on thin film deposition at SIAM’s 2010 materials science conference and was an organizer, with Dionisios Margetis and Peter Smereka, of a three-part minisymposium on the subject.