From Initial to Late Stages of Epitaxial Thin Film Growth: STM Analysis and Atomistic or CoarseGrained Modeling

James W. Evans
*Iowa State University*, evans@ameslab.gov

Yong Han
*Iowa State University*, y27h@iastate.edu

Barış Ünal
*Iowa State University*

Maozhi Li
*Iowa State University*

K. J. Caspersen
*Iowa State University*

See next page for additional authors

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Evans, James W.; Han, Yong; Ünal, Barış; Li, Maozhi; Caspersen, K. J.; Jing, Dapeng; Layson, Anthony R.; Stoldt, C. R.; Duguet, T.; and Thiel, Patricia A., "From Initial to Late Stages of Epitaxial Thin Film Growth: STM Analysis and Atomistic or CoarseGrained Modeling" (2010). *Chemistry Conference Papers, Posters and Presentations*. 1.

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Abstract
Epitaxial thin film growth by vapor deposition or molecular beam epitaxy under ultra-high vacuum conditions generally occurs in two stages: (i) nucleation and growth of well-separated islands on the substrate; (ii) subsequent formation of a thicker continuous film with possible kinetic roughening. For homoepitaxial growth, two-dimensional (2D) monolayer islands are formed during submonolayer deposition. Typically, the presence of a step-edge barrier inhibits downward transport and leads to the formation of mounds (multilayer stacks of 2D islands) during multilayer growth. For heteroepitaxial growth, islands formed in the initial stages of deposition sometimes have a 2D monolayer structure. However, they may instead exhibit bilayer or 3D multilayer structure due to, e.g., a high film surface energy, strain, or quantum size effects. Various growth modes are possible for thicker films. Atomistic modeling provides the most detailed picture of film growth. For coherent (defect-free) epitaxial films, lattice-gas modeling analyzed by kinetic Monte Carlo simulation (KMC) is particularly successful in describing film growth on the appropriate time and length scales. For large islands or complex systems, another effective and instructive approach is laterally coarse-grained step-dynamics modeling which tracks only the evolution of step edges in each layer. However, fully coarse-grained 3D continuum modeling for the evolution of a film height function does not yet have predictive capability. Examples are provided for: Ag homoepitaxy on (100), (111) and (110) surfaces; Ag heteroepitaxy on lattice-matched substrates including NiAl(110), NiAl(100), and Fe(100); and Ag heteroepitaxy on 5-fold icosahedral Al-Pd-Mn and 2-fold decagonal Al-Cu-Co quasicrystalline surfaces.

Keywords
Physics and Astronomy, Institute of Physical Research and Technology, Mathematics, Ames Laboratory, Materials Science and Engineering, thin films, vapour phase epitaxial growth, nucleation, diffusion, surface energy

Disciplines
Biological and Chemical Physics | Materials Science and Engineering | Mathematics | Physical Chemistry

Comments
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Authors
James W. Evans, Yong Han, Barış Ünal, Maozhi Li, K. J. Caspersen, Dapeng Jing, Anthony R. Layson, C. R. Stoldt, T. Duguet, and Patricia A. Thiel

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J.W. Evans, a,b,c,f Yong Han, a Barış Ünal, d,f,h Maozhi Li, g K.J. Caspersen, a,e,i Dapeng Jing, c,f A.R. Layson, a,e,j C.R. Stoldt, a,e,k T. Duguet, c,f and P.A. Thiel d,e,f

a Institute of Physical Research and Technology, Departments of b Physics & Astronomy, c Mathematics, d Materials Science & Engineering, and e Chemistry, and f Ames Laboratory - USDOE, Iowa State University, Ames, Iowa 50011 USA

g Department of Physics, Renmin University, Beijing, People’s Republic of China

h Current address: Department of Chemical Engineering, Massachusetts Institute of Technology, Cambridge, MA 02139 USA

i Current address: Lawrence Livermore National Laboratories, Livermore, California 94550 USA

j Current address: Lawrence Livermore National Laboratories, Livermore, California 94550 USA

k Current address: Department of Mechanical Engineering, University of Colorado, Boulder, Colorado 80309 USA

Abstract. Epitaxial thin film growth by vapor deposition or molecular beam epitaxy under ultra-high vacuum conditions generally occurs in two stages: (i) nucleation and growth of well-separated islands on the substrate; (ii) subsequent formation of a thicker continuous film with possible kinetic roughening. For homoepitaxial growth, two-dimensional (2D) monolayer islands are formed during submonolayer deposition. Typically, the presence of a step-edge barrier inhibits downward transport and leads to the formation of mounds (multilayer stacks of 2D islands) during multilayer growth. For heteroepitaxial growth, islands formed in the initial stages of deposition sometimes have a 2D monolayer structure. However, they may instead exhibit bilayer or 3D multilayer structure due to, e.g., a high film surface energy, strain, or quantum size effects. Various growth modes are possible for thicker films. Atomistic modeling provides the most detailed picture of film growth. For coherent (defect-free) epitaxial films, lattice-gas modeling analyzed by kinetic Monte Carlo simulation (KMC) is particularly successful in describing film growth on the appropriate time and length scales. For large islands or complex systems, another effective and instructive approach is laterally coarse-grained step-dynamics modeling which tracks only the evolution of step edges in each layer. However, fully coarse-grained 3D continuum modeling for the evolution of a film height function does not yet have predictive capability. Examples are provided for: Ag homoepitaxy on (100), (111) and (110) surfaces; Ag heteroepitaxy on lattice-matched substrates including NiAl(110), NiAl(100), and Fe(100); and Ag heteroepitaxy on 5-fold icosahedral Al-Pd-Mn and 2-fold decagonal Al-Cu-Co quasicrystalline surfaces.

Keywords: Homoepitaxy, heteroepitaxy, Scanning Tunneling Microscopy (STM), Density Functional Theory (DFT) analysis, atomistic lattice-gas models, KMC, step-dynamics, island formation, kinetic roughening

PACS: 68.55.-a, 61.44.Br, 68.37.Ef, 81.15.Aa

INTRODUCTION

Film growth typically occurs initially via island formation on the substrate, and subsequently by development of thicker continuous films [1-4]. In defect-free coherent epitaxial growth on crystalline substrates, adsorbed atoms reside at a periodic array of adsorption sites on the substrate and in higher layers. Below is a more detailed enumeration of the key processes underlying film growth behavior in such epitaxial systems (see Fig. 1):

(i) “Random” deposition on the substrate at rate F (in units of atoms per adsorption site per unit time). Generally, atoms adsorb at the closest adsorption site. However, the deposition process usually incorporates downward funneling (DF) of atoms deposited at step edges to adsorption sites in lower layers [4,5]. There may also be steering towards any steep sides of multilayer nanoprotrusions [6,7] or shadowing for oblique incidence [8].

(ii) Terrace diffusion of deposited atoms, most often by hopping between neighboring adsorption sites. Hop rates are often 6-9 orders of magnitude above the deposition rate [1-4].

(iii) Island nucleation by aggregation of multiple diffusing adatoms on terraces (homogeneous nucleation) or at defects or traps (heterogeneous nucleation). A critical size, i, is often introduced or prescribed such that islands
containing more than \( i \) atoms are regarded as stable. Islands containing \( i \) or less atoms can dissociate and are regarded as being in quasi-equilibrium with diffusing adatoms [3,4,9].

(iv) Island growth due to aggregation of diffusing atoms. Island growth shapes often have far-from-equilibrium forms controlled by the details of edge or periphery diffusion and also by possible detachment [1-4].

(v) Possible upward diffusive transport of adatoms from lower to higher layers. This process is sometimes assisted by strain effects, at least in systems with a thermodynamic preference for bilayer or 3D multilayer islands. The upward transport process requires that adatoms “break bonds” with the edges of lower layer islands [1].

(vi) Downward diffusive transport of atoms deposited in higher layers to lower layers. This process is often inhibited by the presence of an additional barrier to diffusion over the step edge, which is called the Ehrlich-Schwoebel (ES) barrier [2-4].

For homoepitaxy (A on A), all adatoms are aggregated into a single large island in the thermodynamically preferred submonolayer state [4]. However, non-equilibrium growth morphologies have many “smaller” single-atom high 2D islands. If edge diffusion is inhibited, these can have far-from-equilibrium, irregular (e.g., fractal or dendritic) shapes, rather than equilibrated, geometric shapes [3,4]. The thermodynamically preferred state for multilayer films is a smooth film, but non-equilibrium growth morphologies are typically kinetically roughened [3,4]. Specifically, inhibition of downward transport of atoms deposited in higher layers leads to “unstable growth” characterized by the development of mounds, i.e., multilayer stacks of 2D islands [2-4]. For strongly inhibited downward transport, these mounds may look like “wedding cakes” [3,4] (This characterization of submonolayer and multilayer thermodynamic equilibrium states assumes typical growth temperatures which are below the critical temperatures for 2D phase separation and thermal roughening.)

For heteroepitaxy (A on B, etc.), classic growth modes include Frank van der Merwe (layer-by-layer 2D growth), Stranski-Krastanov (wetting layer + 3D islands), or Volmer-Weber (3D islands) [1,10]. A preference for 2D (3D) islands occurs when twice the surface energy of the overlayer film A is lower (higher) than the adhesion energy of the film A to the substrate B [1]. (Note that twice the surface energy of A corresponds to the adhesion energy of A to A, so this criterion amounts to a direct comparison of different adhesion energies.) Note that the A to B adhesion energy includes a possible strain contribution [1]. It should also be emphasized that there are other possible growth modes, e.g., height selection due to quantum size effects (QSE) will be described below [11]. There is relatively little detailed analysis of the growth kinetics in systems exhibiting bilayer or 3D growth [12]. In particular, we note that to form bilayer or 3D multilayer islands, upward mass transport is essential.

![FIGURE 1. Schematic of the key atomistic processes involved in epitaxial thin film growth: deposition including downward funneling; terrace diffusion; island nucleation; aggregation with or attachment to islands; edge diffusion at and detachment from or dissociation of islands; upward and downward interlayer diffusive transport.](image)

Different possible modeling strategies are summarized in Fig. 2 [3,4,13]. Atomistic modeling tracks the positions of all atoms and thus provides the most detailed description of growth. For coherent defect-free epitaxial films, lattice-gas (LG) models are utilized where atoms reside at a discrete periodic array of crystalline sites (as indicated above). Model input includes rates for all atomistic processes (deposition, terrace and numerous edge and interlayer diffusion processes, attachment and detachment processes). 2D continuum (or step dynamics) modeling does not...
resolve individual atoms but only the positions of step edges within each layer. Model input specifies the local propagation velocity of these step edges. This velocity is determined from the net flux of attachment of atoms diffusing across the adjacent terraces (together with a contribution reflecting edge diffusion). This flux is obtained by solving deposition-diffusion equations, \( \partial n/\partial t = F + D \nabla^2 n \), for the adatom density, \( n \), on terraces adjacent to the step, using appropriate Burton-Cabrera-Frank (BCF) type boundary conditions. Here, \( F \) is the deposition flux and \( D \) the terrace diffusion coefficient. 3D continuum modeling describes the growing film by a continuous height function, \( h \), which depends on the (continuous) lateral position on the substrate. A partial differential equation of the simple form \( \partial h/\partial t = F - \nabla \cdot J + \text{noise} \) describes the evolution of this height function. However, the complication lies in prescribing the lateral surface diffusion flux, \( J \), which depends on the local slope, curvature, etc., of the film surface, and for which the appropriate form is still a subject of debate. The first and second approaches have predictive capability. The third at present offers more a qualitative description.

In this contribution, we consider three different classes of systems. The next section treats homoepitaxial growth of Ag films. The following section treats lateral strain-free heteroepitaxial growth of Ag films on laterally lattice-matched substrates. We place particular emphasis on QSE in the case of the binary alloy substrate NiAl(110). The last main section treats strained heteroepitaxial growth of Ag films on complex quasicrystalline substrates. In these complex systems, the goal has been to propagate quasicrystalline order from the substrate into the overlayer film.

FIGURE 2. Schematic of different modeling strategies [13]: atomistic LG model; 2D continuum step-dynamic model; 3D continuum partial differential equation (PDE) model. (Copyright American Institute of Physics 2007)

HOMOEPITAXIAL GROWTH OF AG ON AG(100), AG(111), AND AG(110)

Here, we consider “simple” strain-free homoepitaxial growth on fcc(100), fcc(111), and fcc(110) surfaces. Fig. 3 provides a schematic of the arrangement of atoms in the corresponding surface layers, and of the expected 0 K geometric equilibrium island shapes on these surfaces.

Ag/Ag(100): “False Prototype” for Smooth Quasi-Layer-by-Layer Growth

This system [14-19], and other homoepitaxial metal(100) systems [20-24], constitute perhaps the simplest of all examples of thin film growth. For Ag/Ag(100) below about 300 K, island formation is effectively irreversible (\( i=1 \)), i.e., if a pair of diffusing adatoms meet, they nucleate a new stable island [14-15]. Periphery diffusion is facile, so once terrace adatoms reach the island and convert to edge atoms, they effectively instantaneously reach kink sites. This feature ensures square or rectangular shapes for individual separated growing islands, and simple growth coalescence shapes. See Fig. 4. Note that the equilibrium island shapes are also square (with somewhat rounded corners) reflecting the 4-fold symmetry of the surface. Thus, there is not a dramatic difference between growth and equilibrium shapes. In this system, there are just two key energetic parameters which can be adjusted to match experimental behavior: (i) a terrace diffusion barrier of \( E_d \approx 0.43 \) eV which controls the submonolayer island density [15]; (ii) a small ES barrier of \( E_{ES} \approx 0.07\pm0.01 \) eV at close packed steps (but no barrier at kinked steps).
which induces mound formation and controls kinetic roughening [18]. Downward funneling at step edges to lower 4-fold hollow (4fh) ensures that mound slope (or shape) selection is achieved after a mound steepening regime. An atomistic LG model incorporating 4fh adsorption sites and a fcc(100) crystal structure can be tailored to incorporate the above key features. This tailored model allows efficient KMC simulation for film thickness up to $\theta \sim 10^4$ monolayers (ML). All key aspects of the experimentally observed morphological evolution of the growing film are captured by the model [17,18]. See Fig. 5 and Fig. 6.

**FIGURE 3.** Schematic of the arrangement of atoms in the top layer of fcc crystal surfaces with various orientations, and of the 0 K equilibrium island shapes (white atoms) on those surfaces: (a) fcc(100) surface with square islands having close-packed $<110>$ step edges; also shown is a kinked $<100>$ step edge; (b) fcc(111) surfaces where hexagonal islands have alternating A- and B-type step edges; (c) anisotropic fcc(110) surface with a channel-row structure where 0 K islands are elongated rectangles.

Deposition of Ag on Ag(100) at 300 K was traditionally regarded as the prototype of smooth growth [25]. In fact, initial growth is quasi-layer-by-layer. However, after deposition of $\theta \sim 30$ ML of Ag, the presence of the small ES barrier produces mound formation with slope steepening and rapid roughening. This regime, as observed by STM [18] and surface-sensitive x-ray scattering [26], persists for the next $\sim 1000$ ML until slope (or shape) selection is achieved. See Fig.6. Once this selection is achieved, our simulation studies indicate that mound coarsening occurs as a random or stochastic process at least in the subsequent regime up to $\theta \sim 10^4$ ML [18].

**FIGURE 4.** Submonolayer deposition of $\sim 0.25$ ML Ag on Ag(100) at 300 K forming near-square or rectangular islands with examples of growth coalescence shapes: (a) STM (80\times80 nm$^2$) $F=2.2\times10^{-3}$ ML/s; (b) KMC (47\times47 nm$^2$) $F=5.5\times10^{-2}$ ML/s. Different image sizes are selected to match the total number of islands, given different $F$ and $F^{1/3}$-scaling of the island density.

In the simplest picture for slope selection in very large mounds, there is a net lateral surface diffusion flux on mound sides in the uphill direction due to “reflection” from descending steps and irreversible incorporation at ascending steps (a result of the ES barrier). This flux is counterbalanced by a non-diffusive lateral flux in the downhill direction due to downward funneling [3,4]. However, the situation is complicated by finite mound sizes.
The atomically flat terrace constituting the top layer of a mound can be quite broad. (This case arises when the “threshold size” of this top terrace, i.e., the minimum size required for formation of new layers, is large.) Then, for smaller mounds, where the threshold size is significant compared to the mound lateral diameter, the flat mound top limits the side slope for selected mound shapes. Specifically, the slope for the selected shape will be lower than the selected slope for very large mounds. This mound slope will slowly increase during coarsening as the mound diameter grows. Eventually, this mound lateral diameter will become much larger than the threshold size, and the “true” selected slope for large mound sizes will be achieved. Further coarsening then occurs with fixed mound slope [27,28]. See Fig. 6, which characterizes schematically the different stages of mound evolution.

Continuum theory can describe at least qualitatively the possibly delayed formation of mounds, as well as their subsequent steepening and slope (or shape) selection [21,29,30]. There are some quirks associated with a 4-fold symmetry of the metal(100) surface. However, the basic picture is that once slope selection is achieved, the continuum theory predicts an essentially deterministic mound coarsening process, i.e., the average mound diameter and valley-to-peak height increase, while the slope of the sides is maintained. In contrast, KMC reveals stochastic coarsening up to at least ~10^5 ML, as noted above, although asymptotic evolution should be deterministic [18].
Ag/Ag(111): Prototype for Rough Poisson-like Growth

In this system, terrace diffusion is rapid with a barrier no higher than $E_d \approx 0.1$ eV [3,4,31]. The onset of reversibility in island formation occurs at around 150 K, and is controlled by a dimer bond strength of $E_b \approx 0.2$ eV [31,32]. However, it suffices to use a model with $i=2$ (i.e., dimers can dissociate, but no detachment from larger islands) in the temperature range from 100-200 K, where one finds the most dramatic transitions in film morphology [32]. For submonolayer deposition, inhibited edge diffusion including “corner diffusion anisotropy” (see below) results in the formation of: 3-fold symmetric dendritic islands for 120-135 K; isotropic fat fractals for 150-165 K; and distorted hexagons for 180-200 K [33]. See Fig. 7. A large ES barrier in this system ensures that the signature of these submonolayer island shapes is propagating into higher layers. This feature determines the lateral morphology of observed “wedding cake” like mounds [34,35]. Development of a predictive atomistic model for film growth necessarily incorporates several factors: fcc(111) 3-fold hollow (3fh) adsorption sites for a fcc(111) crystal geometry; reversible island nucleation described by $i=2$; appropriate edge diffusion barriers for a large variety of local edge configurations, where these are selected with guidance from semi-empirical (EAM) energetics; and a precise treatment of interlayer transport (see below) [32]. Such a model can describe the observed complex features of islands and wedding cake mounds, even the dramatic variations with temperature. See Fig.7 and Fig. 8.

FIGURE 7. Images (280×280 nm$^2$) of distorted hexagonal island shapes at 180 K [32]: (a) STM data; (b) KMC simulation with a uniform ES barrier of 0.1 eV and corner diffusion anisotropy producing longer B-steps; (c) KMC simulation, also including a non-uniform ES barrier (0.08 eV at B steps and 0.16 eV at A-steps). (Copyright American Physical Society 2008) Images (200×200 nm$^2$) of fat isotropic fractals islands at 150 K [33]: (d) STM data; (e) KMC. Images of dendritic islands at 135 K [33]: (f) STM data (150×150 nm$^2$); (g) KMC (135×135 nm$^2$) (Copyright American Physical Society 2005)

To explain the meaning of corner diffusion anisotropy [33,36,37], we note that hexagonal islands on an fcc(111) surface are bordered by two types of alternating steps: (100)-microfaceted A-type steps, and (111)-microfaceted B-type steps. See Fig. 3. Corner diffusion anisotropy means that adatoms which have attached in a singly-coordinated or singly-bonded configuration at corners between A- and B-steps are preferentially directed to A-steps. This effect alone would cause A-steps to grow faster and thus “grow out”, so A-steps would become shorter than B-steps. In fact, shorter B-steps are observed for islands grown at 180-200 K, a feature explained below. In contrast, equilibrium island shapes are near-perfect 6-fold symmetric hexagons due to near-equality of step energies for A- and B-type steps. The same features of the potential energy surface for adatoms moving along island edges which contribute to
corner diffusion anisotropy are also responsible for the formation of dendrites with 3-fold symmetric, triangular envelopes at lower temperature. See Ref. [33,37] for a detailed discussion.

Deposition of Ag on Ag(111) at 300 K and below is regarded as the prototype of rough Poisson-like growth where the root-mean-square film roughness, W, increases like $W \sim 0^{1/2}$, a feature confirmed in many experiments [26,34,35]. However, there has been considerable debate regarding the magnitude of the ES barrier [38]. In fact, we find that this barrier is non-uniform, being lower at B-steps (where downward transport by exchange with step edge atoms is facile) [32]. Thus, more adatoms descend the B-steps, causing them to grow faster. This effect on island shape of the non-uniform ES barrier dominates the opposite effect of corner diffusion anisotropy, thus explaining why observed submonolayer islands at 180-200 K have shorter B-steps. See Fig. 7. We also find that the mean size of terraces at the top of wedding cakes is extremely sensitive to precise magnitude of the ES barrier. Comparison of experimental and simulated sizes indicates that the ES barrier equals $E_{\text{ES}} \approx 0.08 (0.16)$ eV at B-steps (A-steps) [32].

![Image](80x340 to 516x625)

**FIGURE 8.** Wedding cake morphologies of 3ML Ag films on Ag(111) [39]: (a) STM data versus (b) KMC simulation at 150 K; (c) STM data versus (d) KMC at 180 K. Simulation results [32] for the mean size of the top terrace as a function of ES-barrier for the B-step (ES barrier for the A-step is chosen twice as large). Experimental values are matched for B-step barrier of 0.08 eV.

To provide more direct elucidation [39] of such basic issues as mound steepening and the height (above the substrate) of valleys between mounds, it is instructive to implement much simpler step-dynamics modeling [3,4]. In this approach, we treat mounds as concentric stacks of circular islands, but with a step-edge region in which deposited atoms funnel down to the lower layer where they immediately attach at the step edge. Diffusive downward transport from higher to lower terraces of atoms deposited on terraces on the side of the mound is prohibited (at, say, 150 K) due to the high ES barrier. Thus, such atoms invariably attach to ascending steps. This attachment flux, together with the DF component, determines the velocity with which the steps advance. All atoms landing on the terrace at the top of the mound hop down, causing that terrace to grow until it reaches a threshold size (determined from experiment) for nucleation of a new top layer island. This threshold size is incorporated into the step-dynamics model. Then, model analysis indicates a valley height of &sim;12 layers for 20 ML films [39]. See Fig. 9. This result is in good agreement with the average height extracted from detailed atomistic simulations. (We should emphasize that this valley height is very sensitive to the degree of downward funneling, and is zero with no DF.)
Ag(110): Prototype for Anisotropic Growth

An fcc(110) surface has an anisotropic channel-row structure as shown in Fig. 3. For Ag on Ag(110), there are strong attractions ($E_b \approx 0.18$ eV) between nearest-neighbor (NN) adatoms in the $<110>$ direction along the rows, and weak attractions ($E_b \approx 0.02$ eV) between further separated NN adatoms in the orthogonal $<001>$ direction across the rows [40]. See again Fig. 3. There is also anisotropy in diffusion, the in-channel direction being preferred ($E_d \approx 0.28$ eV) over the cross channel direction ($E_d \approx 0.38$ eV) [40]. Island growth shapes are highly-elongated quasi-1D in the direction of the channels at lower $T \sim 175$ K, partly reflecting the strongly anisotropic interactions [4,41,42]. A transition towards 2D growth shapes, which are still elongated but presumably closer to equilibrium, occurs above 200 K. KMC simulations of an appropriate anisotropic lattice-gas model have been performed by the Genova group [40,41], reproducing the above behavior (and also showing small, irregular, somewhat isotropic islands at low $T \sim 140$ K). Interestingly these studies show “intermixing” of aggregated atoms at higher $T$, i.e., the more-recently-deposited atoms can end up embedded in the interior of elongated islands. This implies significant detachment and reattachment [40]. Note that in isotropic systems, most edge atoms are multiply coordinated with at least 2 strong bonds and are thus effectively irreversibly incorporated. In contrast, atoms at the ends of elongated islands have only a single strong bond and thus more readily detach during growth.

FIGURE 9. KMC simulation results [39] for the morphology of a 20 ML Ag film on Ag(111) deposited at 150 K showing a top view (a) and a line-profile (b) of length 140 nm. (c) Step-dynamics simulations [39] of the profile of one side of a single mound for different coverages reasonably recovering the simulated the peak and valley heights above the substrate for the 20 ML film. (Copyright Institute of Physics UK 2009)

FIGURE 10. Elongated islands formed by Ag deposition on Ag(110). STM images (60×60 nm$^2$) for ~0.3 ML Ag with $F = 0.045$ ML/s: (a) 195 K; (b) 220 K; (c) 240 K [4,42]. KMC simulation of an anisotropic model (images 100 atoms or ~30 nm across) with $F = 0.01$ ML/min.: (a) 190 K; (c) 220 K (f) 240 K [40]. (Copyright American Institute of Physics 1997)
For multilayer growth at higher T ~ 175 K, the elongated submonolayer islands evolve into elongated mounds or “ripples” [43]. Interestingly, the orientation of these ripples was observed to rotate by 90° below 150 K, a feature captured in simulations of an anisotropic LG model [44] and also incorporated into a phenomenological continuum theory description [45].

HETEROEPITAXIAL GROWTH OF AG ON LATTICE-MATCHED SUBSTRATES

Deposition of Ag upon lattice-matched substrates allows us to neglect effects of lateral-mismatch strain, and thus enables development of particularly high-level realistic atomistic modeling of the kinetics of film growth. This, in turn, allows sophisticated and detailed exploration of such issues as the impact of QSE on the kinetics of film growth. We should mention that there have been recent substantial advances in atomistic modeling of “strongly strained” heteroepitaxial growth systems [46], but to date these include idealized or generic descriptions of kinetics.

Ag/NiAl(110): Prototype for Bilayer Island Formation driven by QSE

First, we note the almost perfect lateral lattice-match for Ag(110) [with a rectangular unit cell of 0.4086×0.2889 nm² as determined the bulk lattice constant aAg = 0.4086 nm] on NiAl(110) [with a rectangular unit cell of 0.4083×0.2887 nm² determined by the bulk lattice constant aNiAl = 0.2887 nm]. The NiAl(110) surface is bulk-terminated with a checker-board arrangement of alternating near-in-plane surface Al and Ni [47,48]. Here we describe our recent extensive experimental and modeling analyses of the growth of Ag films on NiAl(110) [12,49,50]. In these Ag(110) films, the lowest-layer Ag adatoms sit at short Ni-bridge (Ni-br) sites on the substrate (see Fig. 11) and in higher layers, Ag sits in fcc(110) 4fh sites [12,50]. A key feature of this system (as quantified by DFT) is that the relative surface energy of supported Ag(110) films with an even number of layers is lower than that for an odd number of layers [49,51]. The bilayer oscillation in surface energetics is also apparent in behavior of the adsorption energy for isolated Ag adatoms on films of different thickness: the adsorption energy is stronger for adatoms on films of odd thickness [12,50,51]. Note, e.g., that adatoms accumulating on top of a 1-layer film create a more stable 2-layer film. The origin of the bilayer oscillations lies in the feature [52] that the half Fermi wavelength of electrons confined vertically in the Ag film roughly matches the ~3 nm height of a bilayer Ag(110) island [49,51]. The lateral lattice-match between Ag(110) and NiAl(110) and this bilayer energetic variation together explain: (i) the observed formation of bilayer Ag(110) islands on the NiAl(110) substrate during the initial stages of deposition; and (ii) the subsequent bilayer-by-bilayer growth occurring at least up to 6 monolayers (or 3 bilayers) [12,49,50].

We now present a realistic atomistic LG model [12,50] which can describe bilayer island formation in this system. In particular, we obtain insight into why bilayer island formation is facile even at low T ~ 130 K, despite the need for upward mass transport (which is typically strongly inhibited). Extensive DFT analysis is utilized to provide energetic input for the model, thereby enabling accurate description of terrace and edge diffusion, attachment-detachment kinetics, and also of QSE. The latter impact key interlayer transport processes. In our “single-site” LG model, Ag adatoms on the NiAl(110) substrate reside only at Ni-br sites as shown in Fig. 11. (The single-site simplification is discussed further in the following subsection.) Ag adatoms hop diagonally between such Ni-br sites with a barrier of Ec ≈ 0.27 eV. The barrier for hopping along straight [001] edges of rectangular Ag(110) islands is Ec ≈ 0.13 eV, and that for hopping along straight [110] edges is Ec ≈ 0.29 eV. Note that [100] edges are in the vertical y-direction in our figures, and [110] edges are in the horizontal x-direction. Transport around corners occurs via detachment-reattachment rather than direct hopping. NN attractions are strongly anisotropic: Eby ≈ 0.09 eV (Ebn ≈ 0.04 eV) for the [001] ([110]) directions. For 2nd layer Ag, adsorption is stronger by ΔEads ≈ 0.05 eV due to QSE, and we increase Eby ≈ 0.18 eV to around the value for Ag(110) homoepitaxy. Other parameters, which do not significantly affect growth behavior, adopt 1st layer values. See Fig. 11. Population of the 3rd layer is excluded due to weaker Ag adsorption, again reflecting QSE. The specification of general activation barriers for hopping, Ecst, is built around these DFT results (i.e., it recovers the above values for terrace diffusion and diffusion along straight edges). However, this specification must also prescribe edge diffusion and detachment barriers for a large number of other edge configurations in a way which is consistent with detailed-balance [12,50]. We choose a common prefactor or attempt frequency for all types of diffusive hops.

For intralayer terrace diffusion and attachment-detachment processes, we select the activation barrier as Ecst = Ed + Eyini where Eyini is the total lateral interaction (attraction). For edge diffusion, we select the barrier as Eedge = Ecst +
for hopping in the x-direction, and $E_{\text{edge}} = E_{\text{ex}} + [E_b^{\text{init}} - E_{\text{bx}}]$ for hopping in the y-direction, thereby recovering DFT values for straight edges. For upward transport, we select the barrier as $E_{\text{up}} = E_d + E_b^{\text{init}}$ equal to that for intralayer detachment. This choice means that we do not include any additional step climbing barrier (as simulation analysis indicates that any such barrier of significant size impedes bilayer island formation). For downward transport, we select the barrier as $E_{\text{down}} = E_d + E_b^{\text{init}} + \Delta E_{\text{ads}}$, noting that enhanced adsorption in the 2\textsuperscript{nd} layer inhibits downward transport and thus enhances 2\textsuperscript{nd} layer population. This choice is consistent with the lack of an additional step climbing barrier (and corresponds to the lack of an ES barrier in the case where $\Delta E_{\text{ads}} = 0$).

Results from the above atomistic LG model, implemented for simplicity with a simple-cubic crystal geometry rather than a fcc(110) geometry, describe well the observed bilayer Ag(110) island formation even at low T [12,50]. Fig. 12 shows that deposition at a “very low” T=130K with a high F=$2 \times 10^{-2}$ bilayers per sec. (BL/s) produces only partial bilayer islands (essentially because upward transport in the system is kinetically-limited for these extreme deposition conditions). However, almost complete bilayer islands are formed in less than 10 min. after deposition due to the thermodynamic preference for such structures (due to QSE). For T=140 K and above, the model predicts that almost complete bilayer islands form during deposition, consistent with experimental observations [12,50]. To explain the efficiency of the associated upward transport even at low T, we note that diffusing adatoms can easily ascend the vertical [001] sides of islands due to weak interaction $E_{\text{bx}}$. In contrast, for isotropic systems, edge atoms interact strongly with all sides of an island. Perhaps more significantly, adatoms at 1\textsuperscript{st} layer kink sites for Ag/NiAl(110) islands are only bound by a single strong bond, and can readily detach to the terrace or straight edge and climb to the 2\textsuperscript{nd} layer either during or after deposition. In contrast, for isotropic systems, adatoms at kink sites are bound by at least two strong bonds, and thus are typically “frozen” during growth.

Another striking aspect of this system is that island growth shapes become progressively more elongated up to at least 300 K [50]. This feature stands in contrast to Ag/Ag(110) as described above, where shapes become less elongated above ~200 K as shown in Fig. 10 [4,40-42]. One could imagine that the bilayer structure more effectively “locks in” growth shapes, thus preventing shape equilibration. To test this idea, we have simulated the evolving morphology of growing bilayer Ag(110) islands at 175 K, and compared this with the evolution of monolayer islands simulated using the same model and energetic parameters except excluding 2\textsuperscript{nd} layer population. Indeed, we...
find that bilayer islands exhibit near-frozen morphologies during growth, but that monolayer islands display significant fluctuations (which would facilitate shape equilibration) and are much less elongated. See Fig. 13.

FIGURE 12. Deposition of Ag on NiAl(110) at 130 K with $F=3 \times 10^{-2}$ BL/s [12]. Images are $14 \times 19$ nm$^2$. STM data (a). KMC simulation for 0.07 BL (b); 0.14 BL (c); and 8 min. after deposition (d). Grey (white) denotes 1$^{st}$ (2$^{nd}$) layer Ag. (Copyright American Physical Society 2008)

FIGURE 13. Images ($50 \times 30$ nm$^2$) of Ag deposition on NiAl(110) at 175K with $F=3.3 \times 10^{-3}$ BL/s: (a) STM data; (b-c) KMC simulation images taken 1/6 sec apart revealing negligible fluctuations in island shape. (d) KMC simulation images ($15 \times 15$ nm$^2$) of a model for monolayer island formation with the same energetic parameters but prohibiting 2$^{nd}$ layer population showing significant fluctuations in island shape over the same time period. Grey (white) denotes first (second) layer adatoms.

Ag/NiAl(110): Multi-site Lattice-Gas Picture for Initial Island Formation

Detailed analysis of the Potential Energy Surface (PES) for Ag on NiAl(110) reveals that in addition to the very stable Ni-br adsorption sites, there are also a weakly stable quasi-threefold NiAl$_2$ sites [12,50]. See Fig. 14. The NiAl$_2$ site is a distance ~0.07 nm from the short Al-bridge (Al-br) site, with just slightly stronger adsorption than the Al-br site by ~0.01 eV. There is an adsorption energy penalty of ~0.23 eV for populating this NiAl$_2$ site relative to the Ni-br site. However, this can be offset by enhanced attractive interactions with in-plane Ag atoms for higher
local coverages, as discussed below. Thus, it is natural to consider a multi-site (two-site) LG model for Ag on NiAl(110) allowing population of both these sites. As an aside, multi-site LG models have been used to describe deposition of Ni and Al on NiAl(110) [53], and also to describe chemisorption and catalytic reaction on metal surfaces [54]. Some motivation for use of multi-site LG models comes from DFT analysis, which reveals that certain non-fcc(110) structures for monolayer Ag films have low energies comparable to that of the bilayer fcc(110) structure [50]. These non-fcc(110) structures include arrangements that we call double-chain and square-hex structures with adjacent [100]-oriented rows of atoms on both Ni-br and NiAl₂ sites [50]. See Fig. 15b,c.

Further motivation for multi-site models comes from additional DFT studies of edge diffusion of Ag adatoms along the vertical [100] sides of rectangular Ag(110) islands. These find that the NiAl₂ site adjacent to the island edge has a slightly lower energy than the Ni-br site. (For simplicity, the vertical [100] side of the island was represented by a frozen row of Ag atoms at Ni-br sites in these analyses.) Similar studies reveal that a vertical Ag ad-dimer near a [100] island edge prefers to reside at NiAl₂ sites rather than Ni-br sites. See Fig. 15e,f.

**FIGURE 14.** (a) Schematic of the adsorption sites and the most relevant interaction energies for the multi-site LG model for Ag on NiAl(110). (b) PES for Ag adatom diffusion. Note the stable Ni-br sites (A,B,C,D) and the stable 3-fold NiAl₂ sites (E,G). (Copyright American Physical Society 2010)

**FIGURE 15.** Schematic of: (a) the “standard” monolayer Ag(110) structure; (b) the double-chain structure; (c) the square-hex structure (which has the lowest energy of the three cases); (b) and (c) can be described as a combination of vertical rows of Ag on Ni-br sites and on NiAl₂ sites. (Copyright American Physical Society 2010) Schematic of: (d) a vertical row of Ag on Ni-br sites representing the edge of an Ag(110) island; (e) a single edge atom at the most favorable NiAl₂ site adjacent to this vertical row; (f) a vertical edge dimer with both atoms at the most favorable NiAl₂ sites adjacent to this vertical row.
Knowledge of the energies for the configurations shown in Fig. 15d-f allows estimation of the interaction between Ag adatoms at the Ni-br and NiAl₂ sites described above. We find an attraction between Ag on Ni-br and NiAl₂ sites “diagonally” separated by 0.31 nm of $E_{\text{diag}} \approx 0.15$ eV, and between Ag on NiAl₂ sites on opposite sides of a surface Ni atom “horizontally” separated by 0.28 nm of $E_{\text{h3f}} \approx 0.2$ eV. See Fig. 14. These “strong” values are reasonable considering the fairly optimal separation of the Ag adatoms. Comparing the energy of a monolayer Ag(110) structure in Fig.15a with those for double-chain and square-hex structures shown in Fig.15b,c provides independent estimates for these interactions. The results are reasonably consistent with the above values obtained from analysis of energetics associated with Fig. 15d-f.

Within a multi-site LG model incorporating these additional interactions, we can assess the relative energies of small clusters of Ag with various configurations. Allowing just Ni-br single-site population on the NiAl(110) substrate, the most stable islands are [100]-oriented linear chains up to 7 Ag adatoms. For 8 adatoms, one can form a more stable bilayer island with a rectangle of 6 atoms in the 1st layer and a dimer in the 2nd layer. However, in the multi-site model, one naturally also considers monolayer (n,m)-2D clusters with n adatoms populating Ni-br sites and m adatoms populating NiAl₂ sites in an adjacent row. These can be energetically competitive with linear (n+m)-1D chains of n+m atoms on Ni-br sites. For example, one finds that a (2,1)-2D cluster and a (3)-1D chain are comparable in energy; a (3,3)-2D cluster and a (6)-1D chain are comparable; but a (3,2)-2D cluster is more stable than a (5)-1D chain. Thus, some of these (n,m)-2D clusters could be involved in the pathway to formation of stable bilayer Ag(110) islands.

Another issue which can be afforded a higher-level analysis in the multi-site LG model is that of upward interlayer transport. In the simpler Ni-br single-site LG model, an adatom is attracted weakly to a straight vertical [100] edge of a Ag(110) island with an attraction of $E_a \approx 0.04$ eV. In the multi-site model, this adatom prefers to sit at a NiAl₂ site thus interacts with the step edge atoms with two “strong diagonal” interactions each of $E_{\text{diag}} \approx 0.15$ eV. However, this attraction is off-set by an adsorption site penalty of 0.23 eV, so that the effective attraction to the step edge is only $E_{\text{eff}} \approx (2 \times 0.15 - 0.23)$ eV $\approx 0.07$ eV. Thus, upward transport is still viable.

**Ag/Fe(100) and Ag/NiAl(100): Lattice-Matched Systems and QSE**

The near-perfect lateral lattice-match for Ag(110) on NiAl(110), noted above, carries over to a near-perfect match for Ag(100) on NiAl(100): the square unit cell of side-length 0.2887 nm for Ag(100) matches well 0.2889 nm for NiAl(100) [51]. We note that Ag(100) also matches well to Fe(100) with unit cell side-length of 0.287 nm. In fact, formation of Ag(100) films by deposition of Ag on Fe(100) provides another classic example of QSE in film growth [55-57]. For Ag/Fe(100), experimental studies reveal strongly preferred stable heights of 2 and 5 layers. In addition, there is a weaker variation in surface properties with a period of ~5 layers. The latter is consistent with a simple analysis of electron confinement in Ag(100) films [58,59]. For Ag(100) on Al-terminated NiAl(100), 3 ML and 8 ML films are somewhat more stable than multilayer films of other thicknesses [51], presumably a weak signature of the same ~5 layer periodicity [58,59].

Initial quasi-layer-by-layer growth at 300 K is observed for Ag on Fe(100) [55-57], and predicted for Ag on NiAl(100) [51]. Indeed, growth behavior should be similar to that for Ag/Ag(100) homoepitaxy [4]. Post-deposition annealing studies for Ag/Fe(100) reveal that 2 ML and 5 ML films are particularly stable. However, 3 ML and 4 ML films bifurcate to the more stable 2 ML and 5 ML films upon heating. This bifurcation process has been monitored with Low Energy Electron Microscopy [60,61]. There are kinetic limitations to bifurcation which can be in part circumvented by the presence of pin-hole defects running through the film to the Fe substrate [61].

**HETEROEPITAXIAL GROWTH OF AG ON AL-RICH QUASICRYSTALS**

Deposition of thin metal films upon quasicrystalline substrates may offer the possibility to create new types of materials and nanostructures by propagation of features of the aperiodic substrate structure into the overlayer [62-67]. This motivates pursuit of the major challenge of understanding and controlling growth in these complex systems. Strong adspecies-substrate interactions can generate initial pseudomorphic quasiperiodic 2D near-layer-by-layer growth. However, strain effects are generally expected to lead to either Stranski-Krastanov (SK) or Volmer-Weber (VW) type 3D growth weakening the influence of substrate structure.
Ag on 5-f i-Al-Pd-Mn: Partial Propagation of Quasicrystalline Order and QSE

We have performed detailed studies of the deposition of Ag on the 5-fold surface of an Al-rich icosahedral Al-Pd-Mn quasicrystal [68-72]. This surface [73-75] provides a quasiperiodic arrangement of local adsorption sites for Ag [68,69,71,73]. No two sites are exactly equivalent although similar motifs repeat quasiperiodically. Of particular significance is the occurrence of so-called “dark star” sites, arranged on a pentagrid pattern on the surface. These appear to provide particularly strong binding sites for Ag, and thus constitute traps for heterogeneous nucleation of Ag islands [75]. See Fig. 16. Furthermore, a small “starfish cluster” with a single Ag atom in the center of the dark-star site surrounded by 5 Ag atoms is likely to be particularly stable (corresponding to i=6 at higher T) [71]. Limited STM images indicate that Ag adatoms within larger growing islands are arranged pseudomorphic to the substrate [72]. See Fig. 17a. Our strategy for detailed atomistic-level modeling of the initial stages of film growth in this type of system [76,77] requires mapping out a “disordered-bond-network” identifying a “random lattice” of adsorption sites with neighbors “linked” or identified by “bonds”. Adsorption energies must be assigned for each site (e.g., based on a semi-empirical potential) and for the transition state between each neighboring pair. Then, hopping between neighboring sites can be implemented with the appropriate Arrhenius rates. These rates differ for each pair of sites and each direction (since the two adsorption sites generally have different adsorption energies). After assigning NN adatom interactions, one can perform KMC simulation of deposition, diffusion, and aggregation into starfish and larger clusters [77]. In this way, one can provide a reasonably high-level description not just of adlayer ordering but also the kinetics of deposition processes on these complex surfaces.

Experimental observations of subsequent growth at 365 K indicate that the facile conversion of isolated 2D islands into 3D islands which quickly grow to a “selected height” and then spread laterally [70,72]. See Fig. 17. This height selection is again attributed to QSE, as discussed further below. In the quasicrystalline substrate, different “layers” of atoms actually consist of a few vertically-closely-spaced planes of atoms, with the top layer being Al-rich [73,74]. Likewise, the 3D Ag islands presumably consist of such “layers” with a selected height of 3 layers. However, the height of each layer varies somewhat from island to island on average being 0.26, 0.27, 0.29 nm for layers 1, 2, 3, respectively. This decreases to 0.24±0.03 nm for layer 4, which is finally populated after significant merging of 3rd layer islands [72]. This suggests that a strongly non-fcc(111) pseudo-quasicrystalline structure for the first 3 layers converts to more fcc(111) like structure for higher layers (with a ~0.24 nm step height).

Atomistic-level modeling of 3D island formation in this system would be extremely challenging. Thus, we opt for simpler step-dynamics modeling to assess the key factors guiding height selection and lateral spreading (i.e., inhibition of 4th layer island formation) [72]. As for Ag/Ag(111) mounds, 3D islands are represented by a concentric stack of circular 2D islands. The rate of advance of each island edge comes from solving the standard deposition-diffusion equations for the adatom density, n, on different terraces. However, key non-trivial physics is incorporated
in the boundary conditions [72]. These involve an equilibrium adatom vapor density, $n_{eq}$, at step edges, where $n_{eq}$ will control the rate of adatom detachment from steps [3,4]. The key is to correctly assign the complex form of $n_{eq}$.

Next, we discuss the form of $n_{eq}$. The presence of QSE means that adatom adsorption energies, $E_{ads}^\pm > 0$, can differ above (+) and below (-) a step. This induces corresponding difference in the equilibrium densities $n_{eq}^\pm$. These are determined by equating the chemical potential, $\mu_{step}$, for the step to that for the ideal 2D adlayer gas with these densities, i.e., $\mu_{step} = -E_{ads}^\pm + kT \ln(n_{eq}^\pm)$. As discussed below, $\mu_{step}$ includes a contribution $-E_{ads}$, so $E_{ads}$ above (below) $E_{ads}$ forces $n_{eq}^+$ to be above (below) $n_{ads}^-$. See Fig. 18. The boundary conditions applied to the deposition-diffusion equation in this BCF-type formulation are as follows: $n = n_{eq}^+$ for ascending steps (corresponding to facile attachment), and an attachment flux satisfying $J_{attach} = D(n - n_{eq}^-)/L_{ES}$ for descending steps [3,4]. Here, $L_{ES} = \exp(\beta E_{ES}) - 1$ with $\beta = 1/(k_BT)$ is the ES length for ES barrier $E_{ES} \approx 0.1$ eV inhibiting attachment similar.

**FIGURE 17.** (a) STM image ($23 \times 20$ nm$^2$) of a pseudomorphic 1-layer Ag island on the 5-fold surface of icosohedral Al-Pd-Mn. (b) Total areal occupancy vs. layer: 0.26 ML (white bars) and 0.70 ML (dark bars showing height selection). (c) STM image ($243 \times 243$ nm$^2$) of 3-layer Ag mesas on Al-Pd-Mn. (d) Line profiles for typical individual Ag islands showing a sequence of shapes during growth and highlighting height selection. [72] (Copyright American Physical Society 2009)

**FIGURE 18.** (a) Schematic of our step-dynamics modeling, the lower panel showing the variation of the adatom density across the terraces on the side of a 3D island. The flux across any terrace includes an "equilibrium component" $J_{eq}$ due to detachment from steps and controlled by the equilibrium step densities. There is also a component due to deposition. (b) Step-dynamics modeling of the evolution of the radii of various layers of the 3D island: height selection followed by lateral spreading. [72] (Copyright American Physical Society 2009)
The key remaining requirement is to specify $\mu_{\text{step}}$. As indicated above, $\mu_{\text{step}}$ involves: (i) a negative contribution from bonding to the lower terrace, $-E_{\text{ads}}$; (ii) a negative contribution from lateral bonding (which could increase for higher layers which relax to fcc(111) structure, but which also decreases due to strain buildup as islands grow); and (iii) a positive contribution due to repulsion of any nearby steps. We assume that selection of 3-layer high islands results from QSE-mediated stronger adsorption on top of 2-layer islands. This reduces $n_{\text{eq}}^+\text{on top of the steps of 3rd layer islands (inhibiting 4th layer formation) and boosts } n_{\text{eq}}^+\text{ on top of the steps of 2nd layer islands. See Fig. 18. The latter together with larger } n_{\text{eq}}\text{ for lower larger islands enhances the flow of atoms to the edge of the 3rd layer island producing the lateral expansion of 3-layer high mesas mentioned above [72]. See Fig. 18.}

**Ag on 2-fold Al-Cu-Co: Dependence of Growth Mode on Terrace Chemical Termination**

The 2-fold surface of the decagonal Al-Cu-Co quasicrystal has the remarkable feature of periodicity in one direction and quasiperiodicity in the orthogonal direction [78]. A model for bulk structure by Deloudi and Steuer [79] is available for Al-Cu-Co. This model, together with STM observations of surface structure [78], indicate the predominance of two classes of terraces on the surface, one transition-metal (TM) rich and the other Al-rich in the top layer. Both display rows of atoms in the periodic direction with a separation of 0.4 nm, although STM images indicate a periodicity of $2 \times 0.4$ nm. The spacing between rows follows a Fibonacci sequence built up of long and short separations of $L=0.77$ nm and $S=0.47$ nm. See Fig. 19. Interestingly, the periodicity matches the larger dimension of the rectangular Ag(110) unit cell. One could imagine quasi-periodicity may facilitate an epitaxial match [80] with some multiple of the smaller unit cell dimension in the orthogonal direction (although we do not currently have experimental evidence to support this claim).

**FIGURE 19.** STM images of different types of terraces on 2-fold decagonal Al-Cu-Co [78]: (a) Pure-Al termination; (b) TM-rich termination. Inset shows surface structure deduced from bulk structure model of Deloudi and Steurer. Dark (blue) dots are Al and light (yellow) dots are TM. Periodicity is in the [00001] direction and quasiperiodicity in the [001-10] direction. (Copyright American Physical Society 2009)

Our key experimental observation for Ag deposition at 365 K [81] is that growth is smoother on the TM-rich terrace compared to the Al rich terrace. Further, upon annealing to 420 K, the film of the TM-rich terrace becomes smoother, and that on the Al-rich terrace becomes rougher [81]. See Fig. 20. The annealed morphologies presumably are closer to the thermodynamic equilibrium morphologies. The film on the TM-rich terrace consists of highly elongated 3D Ag islands (cf. elongated islands or ripples for Ag/Ag(110) homoepitaxy). Since this film on the TM-rich terrace evolves to the smoother equilibrium state upon annealing, it must be kinetically roughened during growth, presumably due to the presence of a significant ES barrier. In contrast, the film on the Al-rich terrace consists of less elongated 3D islands which de-wet and coarsen upon annealing.
Insight into this terrace-dependent thermodynamics can be extracted from a benchmark DFT analysis of related simple heteroepitaxial systems [81]. Specifically, we consider the adhesion of fcc Ag slabs to strained fcc Al, Cu, and Co slabs with various orientations. (We also assess adhesion to unstrained Ag slabs where the adhesion energy corresponds to twice the Ag surface energy.) Ag adhesion to Al is weaker than to Cu or Co (or to Ag). This is consistent with the rougher growth and the annealed 3D VW structures for Ag on Al-rich terraces. DFT analysis is too simplistic to confirm stronger adhesion of Ag to TM-rich terraces than to Ag (the traditional signature of smooth growth), but the trend in adhesion of Ag to Al versus TM surfaces is consistent with experimental observations.

SUMMARY

In this contribution, we have attempted to show that remarkably detailed insight into epitaxial film growth can be achieved by an integrated combination of STM experiments and predictive atomistic LG or coarse-grained modeling. Even for the simplest possible case of homoepitaxial growth, remarkably complex behavior emerges which is elucidated by the modeling. However, even here uncertainties still remain, e.g., regarding the nature of the long-time mound evolution dynamics in metal(100) homoepitaxy (arguably the simplest of all epitaxial growth systems). With extensive input from DFT analysis incorporated into atomistic LG modeling, it is now possible to model rather complex lattice-matched heteroepitaxial growth systems, here taking advantage of the feature that strain effects do not dominate behavior. For deposition on quasicrystal surfaces, atomistic-level modeling is viable for the initial stages of deposition (island nucleation and growth). However, at present it is necessary to revert to coarse-grained approaches to provide some insight into multilayer growth behavior.

ACKNOWLEDGMENTS

JE, YH, ML, and KC were supported (for the modeling work on all systems) and DJ, AL, and CS (for the experimental studies on Ag/Ag and Ag/NiAl) by NSF grant CHE-0809472 and by previous NSF grants. BU, TD, PT were supported (for experimental studies of Ag/Al-Pd-Mn and Ag/Al-Cu-Co) by the Division of Materials Sciences, USDOE – BES. ML was also supported by NSF China grant 10704088. The work was performed at Ames Laboratory which is operated for the USDOE by Iowa State University under Contract No. DE-AC02-07CH11358.

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