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Abstract

Metal(100) homoepitaxial systems constitute perhaps the simplest class of systems in which to study thin film growth. Yet, our Variable-Temperature Scanning Tunneling Microscopy (VTSTM) analysis of Ag/Ag(100) homoepitaxy reveals that the variation of roughness with temperature is extraordinarily complex. As the deposition temperature is reduced from 300K to 50K, the roughness of 25 monolayer films first increases, then decreases, and then increases again. Furthermore, a transition from mound formation to self-affine (semi-fractal) growth occurs at around 135K. We postulate that the following the atomistic mechanisms underly this behavior: the existence of a small step-edge barrier inhibiting diffusive downward transport; “downward funneling” of atoms deposited at step edges and microprotrusions towards lower four-fold hollow adsorption sites; and statistically significant deviations from “complete” downward funneling at lower temperatures, where deposited atoms instead become trapped on the sides of (the more prevalent) small steep microprotrusions. To support these postulates, we employ kinetic Monte Carlo simulations to show that atomistic (lattice-gas) models for epitaxial growth, which incorporate these mechanisms, reproduce the experimental data quantitatively.

Comments

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KINETIC ROUGHENING OF MULTILAYER Ag/Ag(100) FILMS: COMPLEX TEMPERATURE-DEPENDENCE IN A SIMPLE SYSTEM

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ABSTRACT

Metal(100) homoepitaxial systems constitute perhaps the simplest class of systems in which to study thin film growth. Yet, our Variable-Temperature Scanning Tunneling Microscopy (VTSTM) analysis of Ag/Ag(100) homoepitaxy reveals that the variation of roughness with temperature is extraordinarily complex. As the deposition temperature is reduced from 300K to 50K, the roughness of 25 monolayer films first increases, then decreases, and then increases again. Furthermore, a transition from mound formation to self-affine (semi-fractal) growth occurs at around 135K. We postulate that the following atomistic mechanisms underly this behavior: the existence of a small step-edge barrier inhibiting diffusive downward transport; "downward funneling" of atoms deposited at step edges and microprotrusions towards lower four-fold hollow adsorption sites; and statistically significant deviations from "complete" downward funneling at lower temperatures, where deposited atoms instead become trapped on the sides of (the more prevalent) small steep microprotrusions. To support these postulates, we employ kinetic Monte Carlo simulations to show that atomistic (lattice-gas) models for epitaxial growth, which incorporate these mechanisms, reproduce the experimental data quantitatively.

INTRODUCTION

Even if their equilibrium structure is simple, films grown by deposition on perfect substrates can develop surprisingly complex surface morphologies. This is because deposition can drive the system into far-from-equilibrium forms, particularly at lower temperatures, T , where equilibration is more limited [1]. A variety of important physical properties of such films depend quite sensitively on their surface morphologies, often the dependence on roughness being of primary importance. Prominent examples include transitions in conductivity or superconductivity, and related localization behavior, in quench condensed metal films on insulating substrates, the magnetic properties of ultra-thin metal films, and the catalytic properties of bimetallic films [1,2]. An important goal is to develop a fundamental and comprehensive understanding of the atomic-scale processes that underlie film growth, and to assess their relationship to film morphology. Such an understanding would better enable control of film structure. However, this goal has yet to be achieved even for simple growth systems.

Homoeptaxy (or self-growth) provides the most simple case to study, because thermodynamics requires that such films grows as smoothly as possible within entropic limits, with each layer filling in sequence (for typical T). Furthermore, by focusing on metal films, one can usually avoid the complexity associated with reconstruction of the substrate surface, a typical scenario for semiconductor systems. Naturally, one expects to see (quasi-) layer-by-layer growth at high T , where kinetic barriers—especially those to interlayer diffusion—can be overcome. It is also the case that deposition at lower T is expected to produce substantially rougher non-equilibrium structures - a growth process referred to as "kinetic roughening" [1,3].

Indeed, traditionally, the most common expectation was that the roughness of deposited films (of a given thickness) should increase monotonically as the deposition temperature is lowered, and the system deviates farther from equilibrium. This view was prompted in part by a highly idealized "rain model" for deposition at very low T , where randomly deposited atoms stick irreversibly at on-top adsorption sites in a simple cubic (SC) crystal geometry [3]. This model exhibits rough growth characterized by a Poisson height distribution (as is easily seen by noting that the height of any column equals the number of times that column is "hit" by depositing atoms). Furthermore, it is clear that this height distribution will persist whenever interlayer diffusion is inoperative (in an SC geometry). As an aside, we note a recently promoted contrasting view that films should grow smoother at lower T , due to the presence of smaller islands [1]. The idea is that atoms deposited on top of islands can more easily reach their edges and hop down, but this neglects the fact that terrace diffusion is reduced at lower T . In fact, we shall see that neither of the above pictures is supported by our results reported below.

Experimental studies, mostly over the last decade, have demonstrated that metal homoepitaxial film growth is far more complex and challenging to understand than suggested by the simple pictures above. One class of examples is the non-monotonic variation of roughness with temperature observed on substrates with threefold symmetry. One finds "re-entrant" smooth growth for Pt/Pt(111) wherein the roughness first increases as T is lowered (traditional behavior), but then decreases again for lower T [4]. Exactly the opposite T -dependence of the roughness is observed for Rh/Rh(111), i.e., the roughness first decreases, then increases again, for decreasing T [5]. In both cases, this anomalous behavior has been related to a transition from compact to irregular shapes of the large 2D islands formed, which occurs at the temperature of extremal roughness. For Pt/Pt(111), the increase in kink sites at island edges below the transition temperature is believed to facilitate interlayer transport, producing re-entrant smooth growth [4].

On substrates with fourfold symmetry, such as Ag(100), there is no *comparable* island shape transition. Non-compact islands are formed only at very low T , where terrace diffusion and corner/kink rounding of adatoms diffusing along island edges are strongly inhibited [1]. Despite this fact, smooth growth has been observed in metal(100) homoepitaxy at low (liquid nitrogen) temperatures where terrace diffusion is inoperative. This is reflected by persistent oscillations in the Bragg diffraction intensity at the out-of-phase condition of destructive interference from consecutive layers [6,7]. More recently, roughness was observed to increase from 300K to 200K for 15-20ML Ag films deposited on Ag(100) [8], but to decrease from 200K to 160K for 50-100ML Cu films deposited on Cu(100) [9]. In these higher T regimes, where terrace diffusion is significant, the formation of "mounds" has also been observed during multilayer metal(100) homoepitaxy, where the mound sides often have selected slope. This phenomenon was first observed in He-atom scattering studies of Cu/Cu(100) growth at lower T [9], and later in STM studies of Fe/Fe(100) growth at 300K [10,11]. Further discussion is provided below.

While these fragments of experimental evidence for the T -dependence of growth in systems with fourfold symmetry have been tantalizing, they have been insufficient to provide a comprehensive picture for growth. As a result, in this study, we provide the broad experimental picture that has been absent, including an assessment of behavior down to extremely low temperatures—where new surprises emerge [12]. Our analysis is naturally divided into two regimes: a mounding regime which is observed above about 135K, and self-affine growth observed at lower T . Kinetic Monte Carlo simulations of tailored atomistic lattice-gas (LG) growth models are applied to test our interpretation of experimental observations. In this paper, we report in detail simulation results only for the T -dependence of growth in the mounding regime. Extensive modeling of self-affine growth at lower T is reported in a separate paper in this Proceedings volume [13]. The result of combining VTSTM experiments with LG modeling is to provide a detailed understanding of the atomistic processes controlling film growth.

EXPERIMENTAL DETAILS

The experiments reported here examining the temperature dependence of multilayer homoepitaxy on Ag(100) were performed using an Omicron Variable-Temperature Scanning Tunneling Microscope (VTSTM). The microscope is housed in an ultra-high vacuum (UHV) chamber with base pressure below 1×10^{-10} Torr. Films were formed by evaporation of pure Ag from an Omicron EFM3 UHV evaporator. Cooling was achieved using liquid nitrogen down to 135K, and using liquid He for lower T. During and after deposition, the substrate temperature, T, remained fixed to within ± 5 K. Data on film morphology was obtained from central portions of broad terraces (of the original substrate) in order to minimize the effect of step edges.

GROWTH MODES IN HOMOEPITAXY

To facilitate analysis and characterization of VTSTM data for the film morphology, we first provide some brief background on possible modes for film growth. For metal homoepitaxy, roughening is often accompanied by the development of three-dimensional mounds with a well-defined characteristic lateral size [1,3,4,5,9-12]. Although other possibilities exist [14], this behavior is usually associated with the existence of an additional potential energy barrier, the Ehrlich-Schwoebel or step-edge barrier, which inhibits downward diffusion at step edges [15]. Further explanation is provided below. In many systems [3], an alternative to mounding is observed known as self-affine (semi-fractal) growth, which is characterized by a continuous spectrum of lateral lengths. A single system usually exhibits just one of these two types of behavior, the distinction between them being provided by the presence or absence of oscillations in the height-height correlation function, $H(r)$. This correlation function simply gives the mean-square height difference for two points on the film surface with lateral separation, r [3].

VTSTM ANALYSIS OF THE TEMPERATURE-DEPENDENCE OF GROWTH

In Fig. 1, we show STM images of 25 monolayer (ML) Ag films deposited on Ag(100) at various T. In order to characterize the vertical morphology precisely, we "discretize" the continuous film height distribution obtained from STM by peak fitting using multiple Gaussian functions with a separation equal to the atomic interlayer spacing, $b=2.04\text{\AA}$. The resulting surface roughness, W (i.e., the root-mean-square width, W , of the height distribution in units of b), versus T, is shown in Fig. 2. The temperature variation of W for 25ML Ag films is remarkably complex: W first *increases* as T is reduced from 300K to about 205K, then *decreases* as T is reduced further to 140K, and finally *increases* again for lower T (at least down to 50K).

Next, we examine $H(r)$ vs. r for the STM data shown in Fig.1. Behavior at 62K (230K) is plotted in Fig. 3a (Fig. 3b). The presence of oscillations at 230K indicates formation of somewhat ordered arrays of mounds. The first maximum (minimum) corresponds roughly to the average mound radius, R_{av} (separation or diameter, $D_{av} \approx 2R_{av}$). These quantities decrease rapidly with decreasing T to about 205K, after which they remain roughly constant. An Arrhenius analysis of this behavior for $T \geq 190$ K (see Fig.4) yields an energy of $E_c \approx 0.074$ eV, which we shall interpret below. Oscillations in $H(r)$ are apparent down to 175 K, but are gone by 135 K, suggesting a transition to self-affine growth. Further evidence for this transition comes from examining the roughness exponent, α , determined from the roughness, W_L , for a range of short STM scans of length L, and using $W_L \sim L^\alpha$ [3]. We find that $\alpha \approx 1$ for $T > 135$ K (reflecting "regular" mounded morphologies), decreasing to $\alpha < 0.5$ below 135 K (reflecting self-affine morphologies). We also find that the average step density (or mean local slope), and the slope of mound sides,

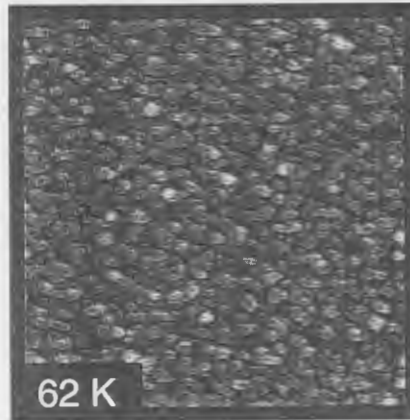
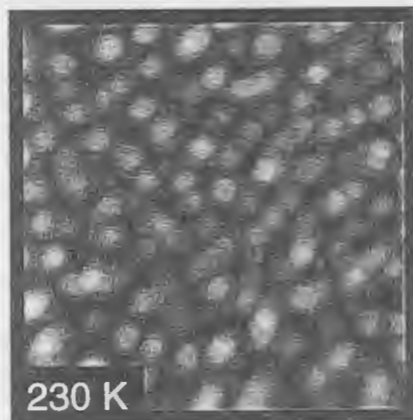
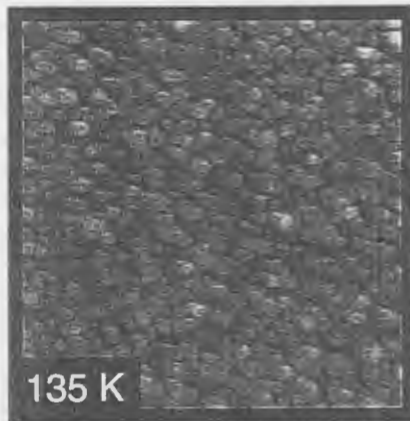
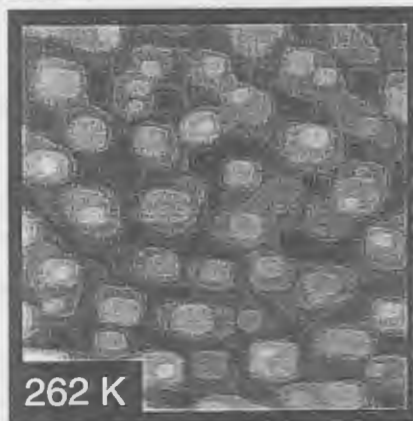
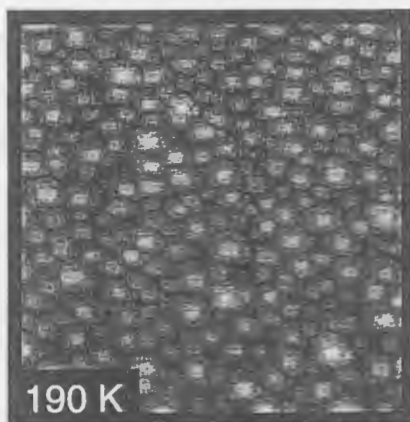
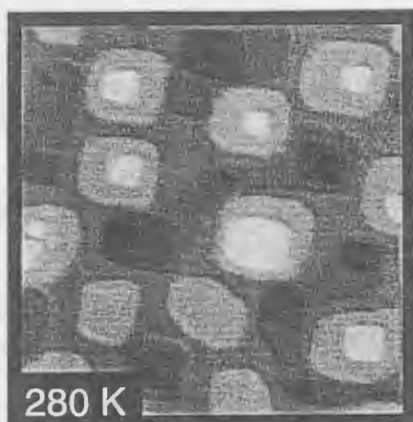


Fig.1 Differentiated $100 \times 100 \text{ nm}^2$ STM images of 25 ML Ag/Ag(100) films deposited with $F \approx 0.02 \text{ ML/s}$ at various temperatures (shown). Darker (brighter) regions are lower (higher).

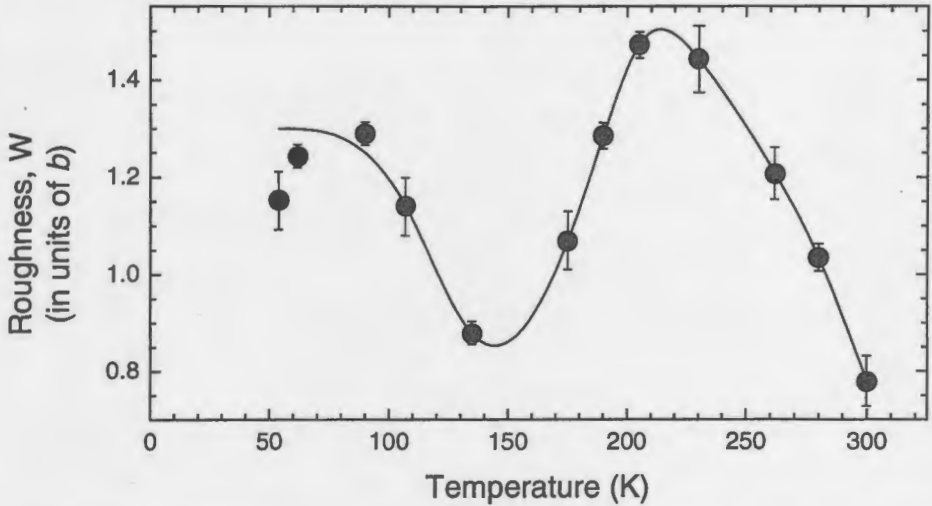


Fig.2 Roughness W (solid circles) in units of interlayer spacing, $b=2.05\text{\AA}$ of 25 ML Ag/Ag(100) films deposited with $F\approx 0.02\text{ML/s}$ for temperatures between 50K and 300K. Solid curve guides the eye; error bars indicate statistical uncertainty (excluding systematic tip effects, expected at lower T where the features of the film morphology are smaller).

increase monotonically with decreasing T , observations which we shall exploit below in interpreting low T growth. The former quantity displays "non-analytic" variation with T at around 135K, consistent with the existence of a transition in growth mode at that point.

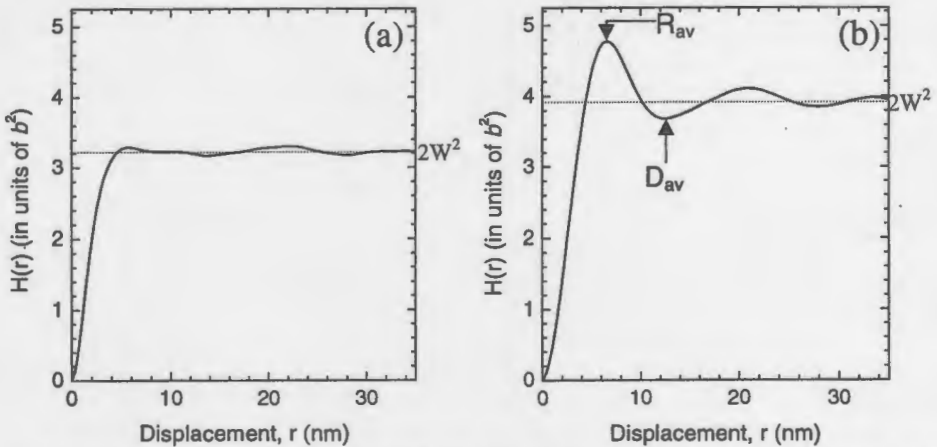


Fig.3 $H(r)$ versus r at 62K (a) and 230K (b) for 25 ML Ag/Ag(100) films deposited with $F\approx 0.02\text{ML/s}$; arrows in Fig3b indicate R_{av} and D_{av} .

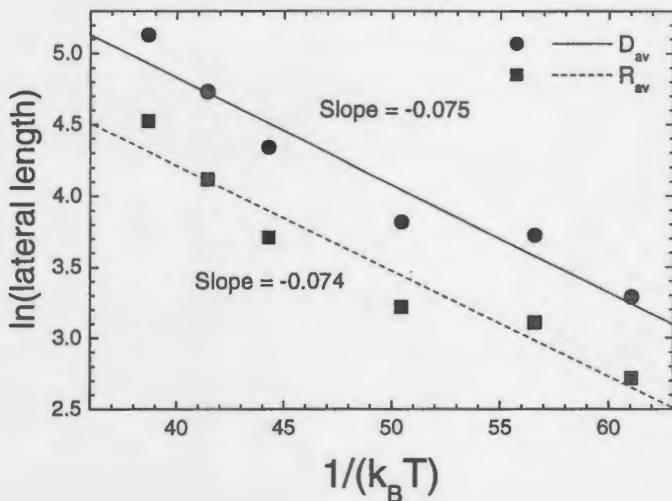


Fig.4 Arrhenius plot for the average mound separation, D_{av} (circles) and radius, R_{av} (squares) for 25 ML Ag/Ag(100) films deposited with $F \approx 0.02$ ML/s. Lengths are in units of lattice constants.

DISCUSSION OF UNDERLYING ATOMISTIC GROWTH PROCESSES

The behavior shown in Figs. 1-4 warrants a detailed discussion regarding the possible atomic processes responsible for the observed growth characteristics. This discussion below is split into the two temperature regimes for mounding and self-affine growth, respectively.

Mounding Regime at higher T

Based on the VTSTM data described above, we propose that film structure above about 135K is determined by three main processes: (i) intralayer terrace diffusion (with barrier E_d) leading to the irreversible formation [16] of near-square islands in each layer; (ii) downward interlayer diffusion inhibited by a small (additional) step edge barrier (of magnitude E_{se}); and (iii) "downward funneling" (DF) or deflection of depositing atoms from step edges and from larger microprotrusions to lower four-fold hollow (4FH) adsorption sites [17]. See Fig.5a. Villain made the key observation that the step edge barrier causes diffusing atoms to be reflected from descending step edges, thus enhancing their probability of capture at ascending step edges. This produces a lateral mass current in the uphill direction (j^{up}) resulting in a growth instability (mounding) [15]. Downward funneling produces stabilizing downhill current (j^{down}) which for sufficiently large slopes of mound sides can counterbalance the uphill current (i.e., $j^{up} + j^{down} \approx 0$), resulting in selection of the slopes of the mound sides [11,18]. See Fig.5a.

At 300K, the two diffusional processes are most important (for films up to ~25ML). Terrace diffusion is highly active producing large lateral structures, and (downward) intralayer diffusion is also efficient, perpetuating smooth growth. As the deposition temperature is reduced from 300 K to about 200 K, the main effect on roughness comes from increasing inhibition of interlayer diffusion due to the presence of the small step-edge barrier. Atoms become less likely

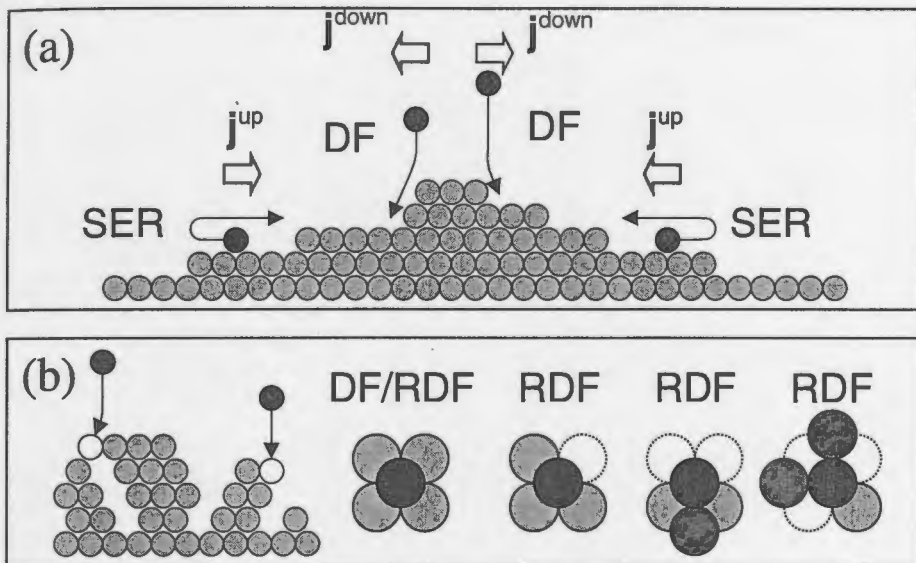


Fig.5 Schematic for: (a) mound formation – step edge reflection (SER), downward funneling (DF), and associated lateral mass currents; (b) restricted downward funneling (RDF) together with a bird's eye view of some trap sites which can capture depositing atoms.

to reach lower layers and, as a result, mounds with nearly-square with broad, flat summits (see Fig. 1) become more pronounced even by 25ML. This is reflected in an increase in W , which peaks between 230K and 205 K (see Fig 2).

Another feature, which is evident with the reduction of temperature, is a strong inhibition of terrace diffusion, leading to a significant decrease in feature size (see Fig.1 and Fig.4), and a corresponding increase in step density. Below about 210 K, we propose that this increase in step edge density becomes a determining factor: it enhances the effect of the DF mechanism, which is to smooth the surface, as is evidenced by the decrease in W below about 205-230K (Fig. 2). Furthermore, DF ultimately to induce a transition out of the mounding regime [18]. Below 230K, we also find that DF significantly influences mound morphology, inhibiting the growth of mound slopes already by 25ML.

Self-Affine Growth Regime at Lower T

From Fig.2, one see that below 135 K, W increases yet again! Such a phenomenon has not been observed previously. Why does this occur? A key point is that the surface becomes increasingly irregular and local slopes become steeper at lower T. Molecular Dynamics simulations of metal(100) homoepitaxy indicate that in such situations, DF can breakdown [19,20], deposited atoms becoming captured on the sides of microprotrusions rather than reaching lower 4FH sites, leading to formation during growth of overhangs and internal defects or voids [20]. This behavior is illustrated schematically in Fig.5b.

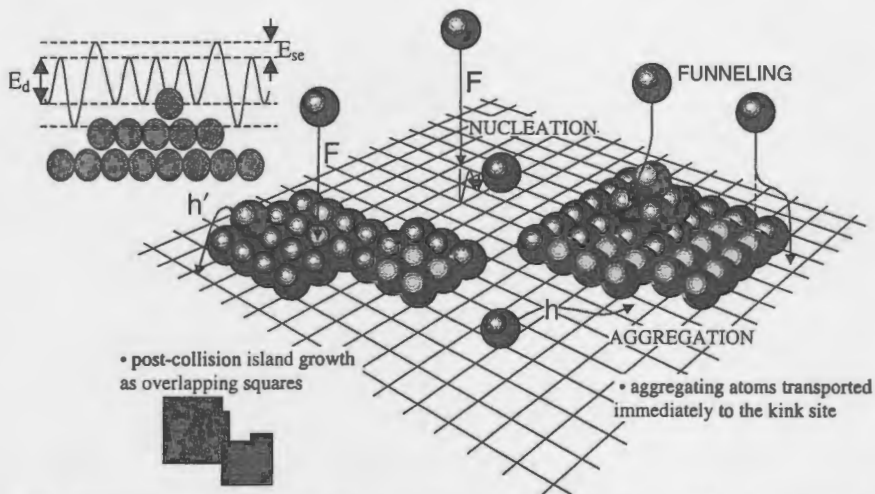


Fig.6 Schematic of our model for mounding incorporating irreversible formation of square islands in each layer, no restructuring of pairs of islands (in the same layer) which collide due to growth, and a uniform step-edge barrier. Here $h = \nu \exp[-E_d/(k_B T)]$ and $h' = h \exp[-E_{se}/(k_B T)]$.

MODELING AND SIMULATIONS OF MOUND FORMATION ABOVE 135K

To test our understanding of the evolution of film structure in the mounding regime, we employ kinetic Monte Carlo (KMC) simulations of a "canonical" atomistic model for homoeptitaxial growth with an face-centered-cubic(100) or FCC(100) crystal geometry [11] that incorporates the following features: irreversible nucleation and growth of near-square islands in each layer, mediated by terrace diffusion; downward thermal transport inhibited by a *uniform* step-edge barrier; and downward funneling from step edges. Also growing islands in the same layer do not restructure upon collision, but continue to grow as overlapping squares. See Fig.6. This model was first developed and successfully applied to describe behavior observed in STM studies of Fe/Fe(100) homoeptitaxy [11]. It was also applied to examine the temperature-dependence of Ag/Ag(100), Cu/Cu(100), and Fe/Fe(100) growth [18], although at that time detailed experimental data did not exist for comparison. From previous Arrhenius analyses of submonolayer island separation [16,21], and from *ab-initio* theory [22], we know that $E_d = 0.38-0.45$ eV. Interestingly, a consistent estimate of $E_d \approx 6E_c = 0.44$ eV follows from an Arrhenius analysis of the mound separation data in Fig.2b (recalling that characteristic lengths scale like the sixth power of the terrace diffusion rate [3,11]). Here, we use $E_d = 0.40$ eV. We estimate the step edge barrier to be 30 ± 5 meV from a fit to the value of W at 230 K reported above. All attempt frequencies were set to the value $10^{13}/s$ (cf. Ref.[16,21]). Hence, there remain no adjustable parameters in the model, and the key question is then how well it describes the observed T -dependence of W ?

As shown in Fig.4a, this kinetic model does in fact reproduce very well the experimental W 's between 300 and 135 K. It also reproduces qualitatively (but not necessarily quantitatively) the monotonic decrease in lateral characteristic lengths and the monotonic increase in mean local slope with decreasing T , as well as a transition out of the mounding regime for lower T . These trends were seen previously in "generic" simulations studies using this canonical model [18].

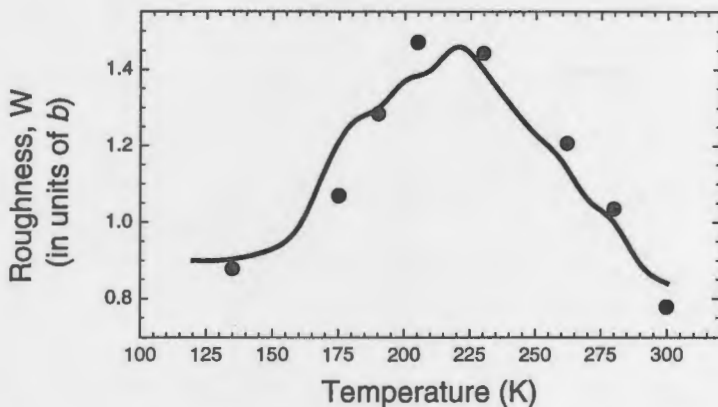


Fig.7 KMC simulation results for the roughness of 25ML Ag/Ag(100) films (solid curve) deposited between 135K and 300K. Experimental data (from Fig.2) are shown as solid circles.

As a further test of our claim that DF produces the observed decrease in W below about 200K, we have repeated these simulations with the exactly same values of the parameters ν , E_d and E_{sc} , but for a SC crystal geometry, where DF is naturally excluded due to the on-top adsorption sites. Indeed, the results of these simulations reveal a monotonic increase in W for 25ML films as T decreases below 300K, achieving a "large" Poisson value of $W=5b$ for 25ML films at low T where all diffusion processes are effectively inoperative.

Despite the success of our canonical modeling, we emphasize that it contains rather severe simplifying assumptions: the idealized treatment of step-edge diffusion leads to near-square isolated islands, and we arbitrarily prescribe that islands do not restructure upon collision. This results in film morphologies with only close-packed [110] step edges, and (consistently) the step edge barrier is assumed uniform. In reality, there can be considerable restructuring of islands, particularly for corner-to-corner collisions [23]. This restructuring, and (perhaps more significantly) subsequent growth of pairs of islands which have coalesced in a corner-to-corner arrangement, leads to the creation of a significant population of open step edges (e.g., of the [100]-type) even for higher temperatures. We anticipate that E_{sc} is likely to be far lower for open step edges, which have a significant density of kinks, than for [110] edges [24]. Thus, our estimate of $E_{sc} \approx 30\text{meV}$ should be regarded as an effective or averaged value. Another observation is that the introduction of island restructuring should increase the lateral correlation lengths in multilayer films. In future work, we shall report simulation results from more sophisticated model which incorporates these features, and which better matches the detailed features of the observed film morphologies [25].

MODELING AND SIMULATIONS OF SELF-AFFINE GROWTH BELOW 135K

We have modeled growth in this low T regime starting from a "restricted downward funneling" (RDF) model, which should apply at 0 K. In this model, deposited atoms funnel downward, but can get stuck when they reach special types of "trapping sites" which do not contain complete quartets of four supporting atoms. This produces films which are much rougher than for the conventional DF model, and which contain a significant concentration of overhangs and internal defects or voids. The next challenge is to extend this model to describe the T -dependence of W in the range up to 135K, recalling that terrace diffusion is still inoperative in

this range. The key point here is that on the irregular structures formed during film growth at low T, there are many other thermally activated interlayer hopping processes (e.g., for three-coordinated atoms) with low barriers which can be operative and affect film morphology. With this in mind, we augmented the above RDF model by incorporating various interlayer hopping processes for atoms. Our model recovers the general experimental trend between 50K and 135K. See Ref.s [12] and [13] for a more detailed discussion. Thus, a picture emerges that idealized DF provides a reasonable description of deposition dynamics at temperatures above 100K, because either the film morphology is locally smooth enough to make breakdown rare, or when breakdown occurs, low barrier interlayer diffusion processes are active which can bring deposited atoms to lower 4FH sites.

We wish to emphasize that our modeling of growth in this low T regime has also incorporated a number of simplifying assumptions, which could significantly impact the predicted film morphology. It is likely that the actual "restricted downward funneling" dynamics of depositing atoms differs from the assumed form. Probably, deposited atoms which directly impinge at trap sites with less than three supporting atoms (e.g., three-fold hollow sites on the side of {111} microfacets) do not remain there, but funnel down to lower trap sites. The "knockdown" of overhanging adatoms at such weakly supported trap sites by subsequent depositing atoms also seems likely. Both these effects would likely reduce the density of internal defects in the growing film from that predicted by our model [13]. Another simplification in our modeling is that while we do incorporate low-barrier interlayer diffusion processes, we completely neglect various intralayer step-edge diffusion processes, which are also known to have low barriers. While the neglect of these processes may not significantly affect the predicted W value, it will likely limit the development of lateral correlations, which are known to be surprisingly large in low-T metal(100) homoepitaxial deposition processes [6].

CONCLUSIONS

In summary, we have shown that the simple Ag/Ag(100) homoepitaxial system exhibits the most complex variation of W versus T yet observed in metal homoepitaxy. Atomistic simulations support the following characterization of the deposition and diffusion processes controlling observed behavior: a small step edge barrier leads to mound formation at the higher temperatures, and an increase in W (for 25ML films) as T decreases from 300K to about 200K; downward funneling at step edges triggers smoother growth and ultimately a transition to self-affine morphologies at lower T (where step edges are more prevalent); and, finally, the breakdown of funneling on small steep microprotrusions leads to rougher growth at very low T. In future work, we shall focus on characterization of kinetic roughening in this system, and on associated coarsening of mounds in the higher temperature regime. There is intense current interest in this topic. However, often experimental data is available only for a single (or perhaps two) temperatures, perhaps giving a misimpression of general behavior. This is problematic since it is typically not possible to obtain true asymptotic values for dynamic exponents describing growth, and the effective values obtained will depend strongly on temperature. Thus development of a fundamental understanding of these growth phenomena is limited by a lack of data for different temperatures, a situation which our study will attempt to remedy.

ACKNOWLEDGEMENTS

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