Fluctuations and Thin Film Growth

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Abstract

Two-dimensional mound coarsening is studied using a continuum model in which the effects of deposition noise have been included but in which coalescence due to mass transfer is assumed to be negligible as expected at low temperature. In agreement with scaling arguments for the case of fluctuation-dominated mound coarsening, we find $n = \beta = 1/3$, where n is the mound coarsening exponent and β is the surface roughening exponent. Our technique for including the effects of deposition noise in a continuum model may have applications in future three-dimensional simulations of polycrystalline growth.

Introduction

Thin film growth refers to a variety of processes in which atoms of a given element are individually deposited on a surface to create a thin film. After growth is complete, these films typically range from a thickness of ten atoms, or approximately 30 Angstroms, to 100 Microns, or 0.1 Millimeters. One deposition procedure is molecular-beam epitaxy (MBE), in which single atoms are deposited in a vacuum and fall vertically towards the surface. This precision process is used to make ultra thin films of high quality, such as semi-conductors used in the lasers of CD players.

However, under certain conditions during MBE, mounds of atoms form, and these mounds coarsen with time.[1] An energy barrier, termed the Ehrlich-Schwoebel barrier, which limits the ability of atoms to diffuse to a lower layer of the film, prompts mound formation.[2-4] At lower temperatures newly deposited atoms often lack the energy to overcome this barrier, and mounds begin to form. A number of experiments have observed that in a given system the mounds coarsen with time and share a common angle with respect to the horizontal.[5-11] Figure 1 shows experimental results of mound formation in a CdS system after MBE, and the results of a three-dimensional CdS epitaxal growth simulation. Figure 2 shows experimental results of Cu mound formation in several systems grown with MBE.

The mechanisms responsible for film coarsening are not well understood. However, two phenomena appear to cause roughening. At high temperatures, most coarsening is caused by mound coalescence due to mass transfer. Neighboring mounds coalesce in order lower their potential energy (Figure 3). In systems at lower temperatures, atoms typically lack the energy to move towards their neighbors, and roughening is mainly due to fluctuations during the MBE process.[12] During MBE, atoms are deposited at random locations in the system, and as time progresses the disparity between the heights of different mounds increases. Consequently, many smaller mounds are eventually covered by their larger neighbors. The number of mounds in a system decreases with time, and average mound height increases, causing the system to coarsen.

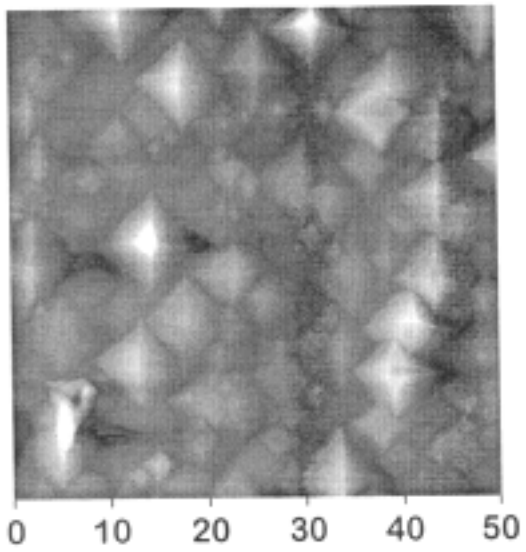
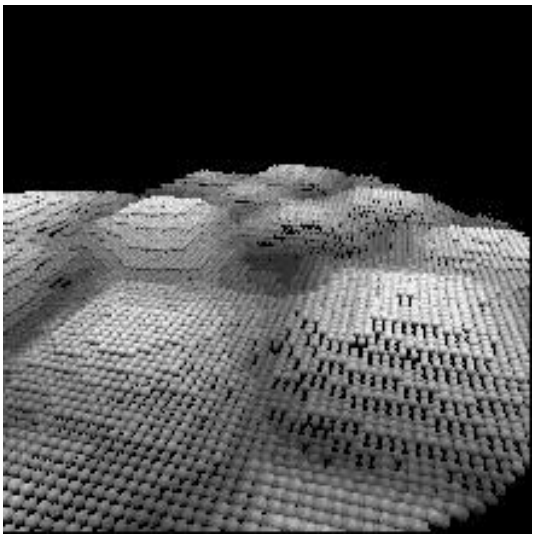


FIG. 1. CdS epitaxial growth experimental results showing mound formation.



Three-dimensional CdS growth simulation.

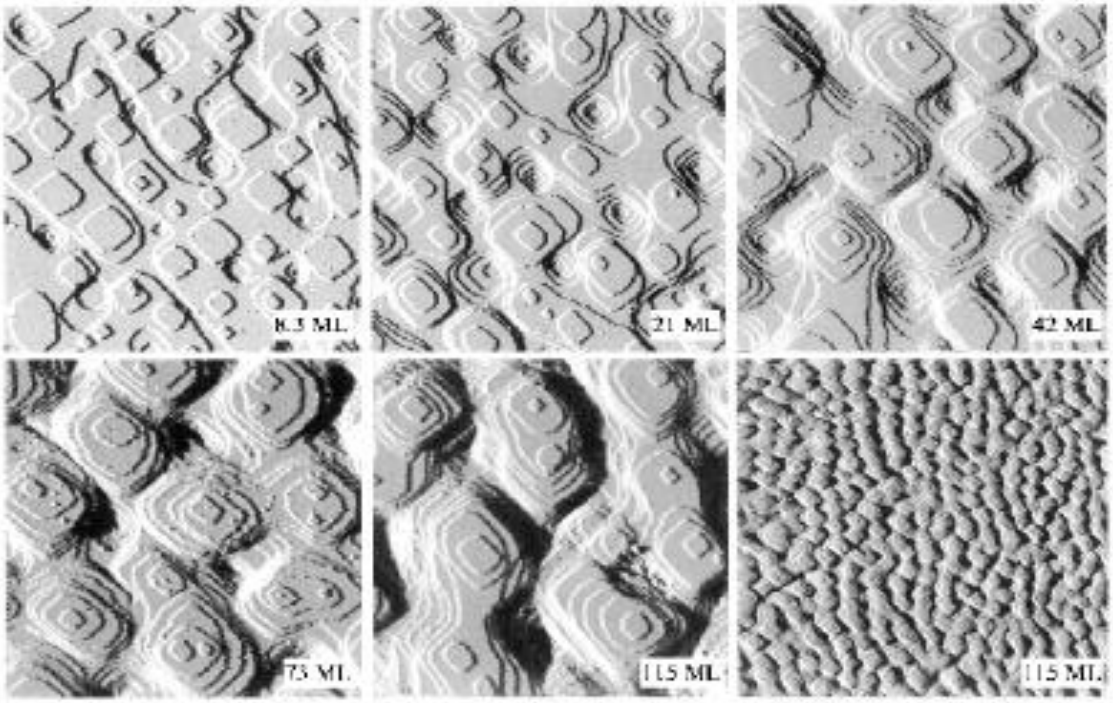


FIG. 2. Experimental results of epitaxial Cu film growth showing mound formation.

A quantitative method of describing the coarseness of a system as a function of system size and time is to compute the system's width. Width is defined as

$$W^2(L, t) = \langle (h - \langle h \rangle)^2 \rangle \quad (1)$$

where L is system size, t is time, and h is height.[13] Width has been found to vary with time during atom deposition according to a power law,

$$W(L, t) \sim t^{\beta}. \quad (2)$$

The constant exponent β is known as the growth exponent, and β "characterizes the time-dependant dynamics of the roughening process."[13]

Thin films are often sandwiched together to make semi-conductors, and for such applications the minimization of film roughness is often sought. Knowledge of the β value of different systems may illuminate the physics responsible for mound coarsening, and improve the design of devices utilizing thin films. Accordingly, methods to predict β for different systems, computational simulations of these models, and the examination of β values of experimentally grown films have all become topics of much interest.

One recent theory proposes that in the case which deposition fluctuations dominate and mound slope is constant,

$$\beta = 1 / (d + 1) \quad (3)$$

where d is the dimension of the system.[14] This result has been derived using a scaling argument, and to our knowledge has never previously been tested in a simulation of MBE which exclusively utilizes fluctuations as the means of coarsening, and neglects mass transfer. Therefore, we sought to test this equation by developing an epitaxial growth simulation that simulates random deposition but does not model mass transfer.

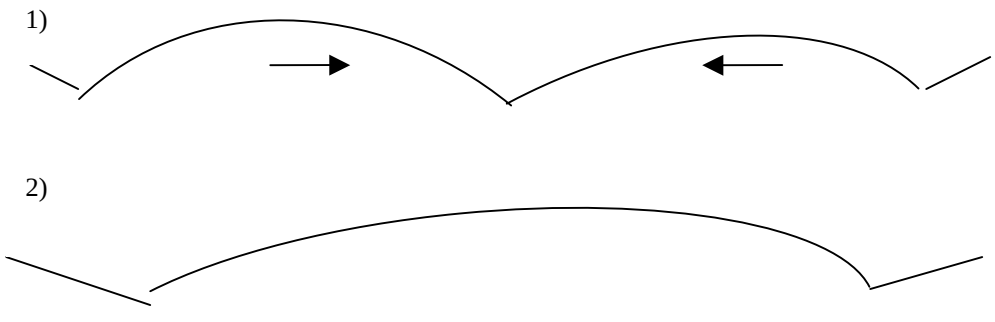


FIG. 3. Two neighboring mounds coalesce to reduce their potential energy.

Methods

Our simulation has several design goals. Perhaps the most basic is to simulate theoretical *two*-dimensional mound growth to reduce development time and programming complexity. We utilize a continuum model to reduce the computer memory needed to simulate large systems, and our model represents mounds as triangles, and uses three unique x-y coordinates to describe each mound. For our simulation to produce meaningful results representing fluctuation-driven coarsening, practical deposition must be truly random. Also, if a mound is overrun by the growth of another, the smaller mound must be incorporated into the larger and cease all independent growth. In other words, small mounds once obliterated must not be allowed to reemerge from under larger mounds. Mound angle must stay constant to follow the constraints of Eq. (3). Lastly, the system must have periodic boundary conditions.

To ensure deposition randomness, we deposit what we like to call 'ghost particles.' Particles are deposited singly in random locations, chosen by a random number generator to lie within the system. Once a deposition location is chosen, the program locates first the mound occupying this location, and then determines on which side of the mound the particle will land. The chosen mound gains an area of two, and this area is added to the side of the mound on which the particle was deposited. Figure 4a shows the results of a typical particle deposition, while Figure 4b shows a deposition in which a neighboring mound is overrun by its larger neighbor. The arrows indicate the chosen deposition locations of the ghost particles, and the added area gained by the mounds is equal to two in both cases.

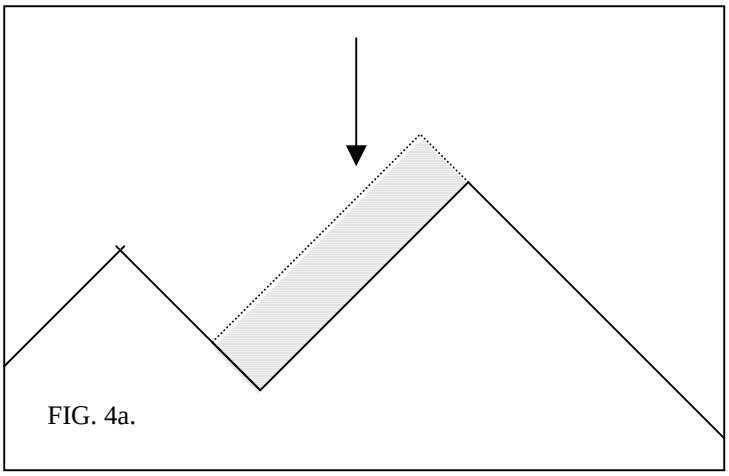


FIG. 4a.

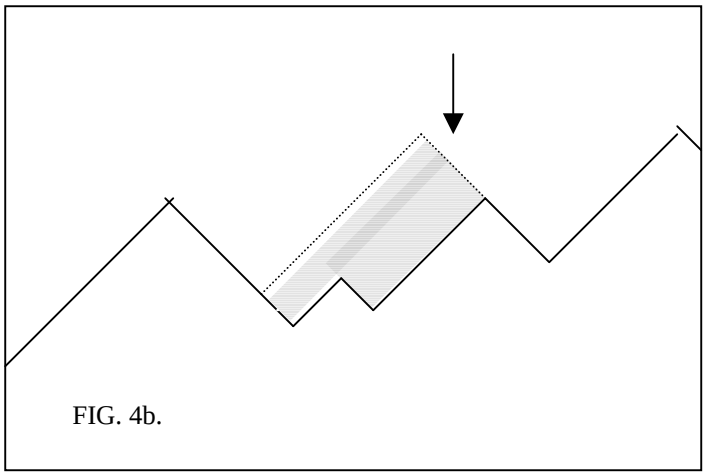


FIG. 4b.

The initial configuration of our system consists of equally sized mounds, with a base of 2 units, and a side angle of 45 degrees that remains fixed throughout the simulation. The initial condition of our system is analogous to that of the discrete single-step model.[15] Figure 5 illustrates an initial configuration of 20 mounds using a screen shot taken while the simulation was running. Figure 6 shows the state of this system after 25 ghost particle depositions, and Figure 7 illustrates a similar system after 75 depositions. In Figure 6, some mounds have been obliterated while others have gained in size. One mound however remains in its initial configuration. The system in Figure 7 has still fewer mounds, yet these mounds are more massive then those in Figure 6. Qualitatively, one can observe that our simulation produces mounds that coarsen with time.

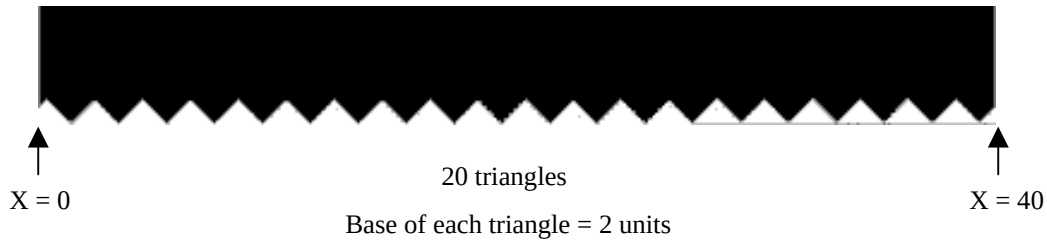


FIG. 5.

FIG. 6.

FIG. 7

Results and Discussion

After each ghost particle deposition, our simulation computes the average height of the system, the system's width (Eq. 1), and the average length of the mound sides. We present graphs and curve fits for data obtained for a system consisting of 2000 mounds upon which 21000 ghost partials were deposited. Figure 9 shows a fit for curves of width and average side length plotted against average mound height, or film thickness. As time progresses the plot of width quickly approaches a state where $W \sim t^{1/3}$ (4), and average mound size length S soon enters a state where $S \sim t^{1/3}$ (5). For the remainder of the simulation, W continues to vary with time as described in (4), and S continues the behavior shown in (5). Figure 10 shows an enlargement of the region where W and S increase according to (4) and (5) respectively.

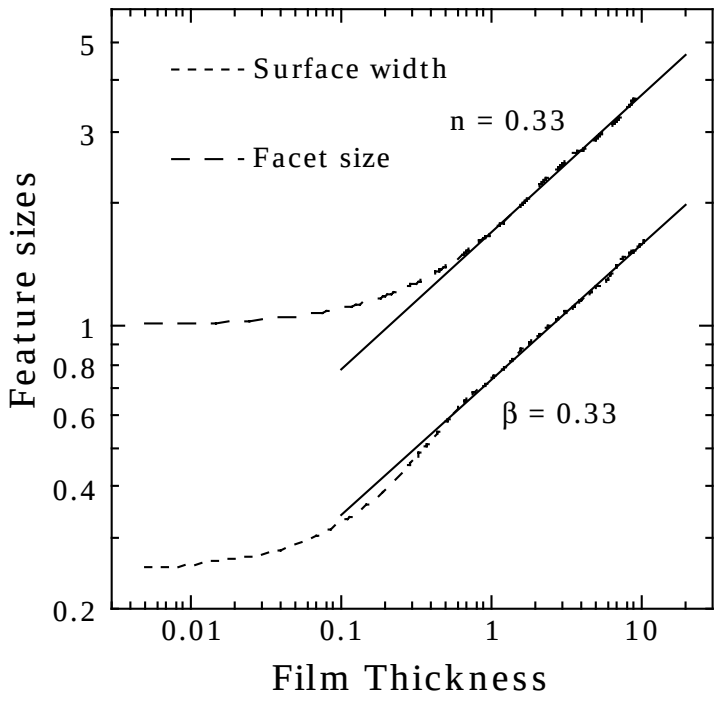


FIG. 9.

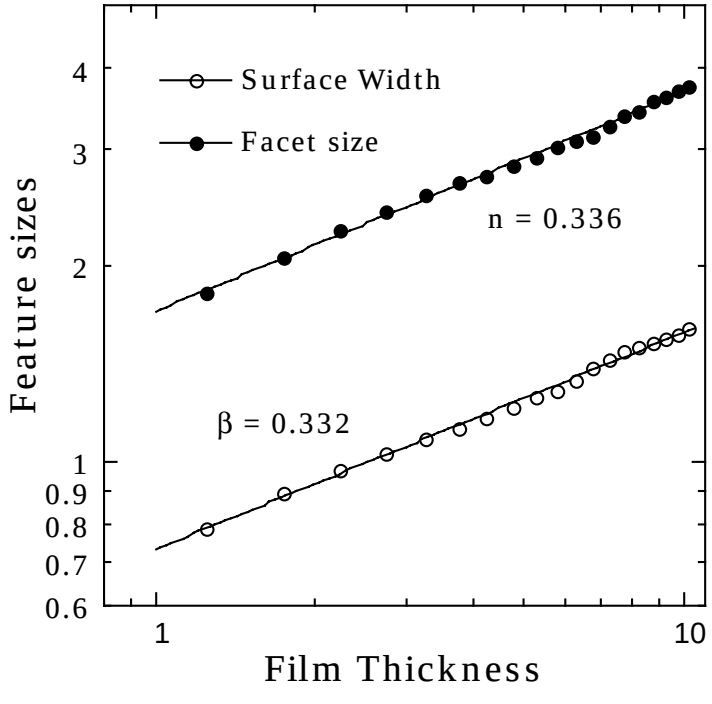


FIG. 10.

Equation (3) predicts that for a two-dimensional system such as ours, $W \sim t^{\beta}$, with $\beta = 1/3$. Again, (3) is supposed to give correct predictions for a system of mounds of a fixed angle undergoing epitaxial growth and dominated by noise driven coarsening. Our model obeys these constraints and moreover eliminates *any* mass transfer driven coarsening. We find $W \sim t^{1/3}$, giving $\beta = 1/3$ for our simulation. Our results show that (3) is correct for d = 2, and the accuracy of the equation's prediction suggests that (3) may hold for higher dimensions. We have found that $S \sim t^{n}$, where $n = 1/3$, suggesting that (3) has implications beyond the definition of width given in (1). Our model produces these results without using mass transfer, showing that coarsening due to mass transfer during epitaxial growth is not necessary to produce a growth exponent of 1/3 in two dimensions.

Our method of depositing ghost particles may have applications in future three-dimensional simulations of polycrystalline growth. Experiments of polycrystalline growth have yielded a growth exponent of 0.5,[16] while theoretical three-dimensional polycrystalline growth computer simulations yield a different growth exponent of 0.4.[17] We believe that the discrepancy may be due to simulations not including noise. Our ghost particle method could be used to model noise in a future three-dimensional polycrystalline growth simulation. However, the crystal upon which a ghost particle lands would have to increase by a fixed volume, rather than the set area gain of our two-dimensional mounds.

Conclusion and Acknowledgements

Our simulation suggests that fluctuations alone in 2D mound growth yield $\beta = 1/3$. Also, the 'ghost particle' noise deposition method we developed for our 2D mound simulation may be useful in future 3D simulations of polycrystalline growth.

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Notes to potential future users of this simulation program at UT

There are five files required to compile the program:

triangleII.cpp

triangle_classII.h

data_classII.h

DBglut.h

triangleIIMakefile.

The program is written in C++, and these five files should be in their own directory somewhere on Dr. Amar's computer Jette. Constants such as system size, triangle number, mound angle, and ghost particle area are globally declared at the top of triangleII.cpp, so it is necessary to recompile the program to change these. The program can be compiled using the triangleIIMakefile. The source code is decently commented. However the program is not super fast, and its speed could be increased by changing the implementation of the triangle class from an array to a linked list. Currently because of inactive triangles the program must perform a sequential search of the triangle array near the beginning of each deposition, and changing to a linked list would enable the outright elimination of inactive triangles, and the implementation of a speedy binary search algorithm. If there are any questions about an aspect of this code I can be reached at baxterd@carleton.edu.