Simulations of Epitaxial Growth With Shadowing in Three Dimensions

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ABSTRACT

Compared to simulations done in the past, relatively little attention has been given to models which include shadowing and diffusion. These cases have been for the most part ignored because they are harder to simulate than less realistic, easier to program models simulating vertical deposition in which particles do not move around once they land on the model. While studying the more realistic case of ballistic deposition with shadowing and diffusion, values of $\beta = 1$ and $n = 1$ were found when a uniform distribution was used, indicating an unstable system. Using a cosine distribution, values of $\beta = 1$ and $n = 0.70$ were discovered.

Introduction

In this project, we simulated the epitaxial growth of thin film layers on a flat substrate in order to study the development and coarsening of mounds in three dimensions. There is a large amount of interest in this field today, but not many realistic simulations have been done. Ours is one of the first to add diffusion and shadowing to the basic model that has been used up until now, consisting of simple vertical deposition.

More and more attention has been given to the study of thin film growth over the years. This interest can be attributed to the many uses which extremely thin films (with thicknesses from a few atoms to a tenth of a millimeter) can be applied. Applications for these thin films range from solar cells to semi-conductors to protective coatings. However, as in any area, problems do arise. In most applications, it is desirous to have a perfectly (or nearly so) flat surface of atoms. However, due to mechanisms not fully understood as of yet, these perfectly flat surfaces are still unattainable.

The reason for this is easy enough to explain: as time goes by, and more and more layers of atoms are deposited on the substrate, mounds begin to form, which shadow the rest of the sample. In this way, mountains and valleys are produced before too long, which render the sample less useful than it could be. It is hoped that by modeling this process of mound coarsening and studying the resulting data, the process of formation can be better understood, and, accordingly, eliminated--or at least minimized.

Before we talk about the program used to model the growth of these mounds, we should explain a little bit about what, exactly, it is that we will be simulating. Thin film growth refers to the process of depositing atoms onto a substrate. This substrate is usually made up of a different element than the one being deposited, and the atoms that are deposited on it immediately begin to defuse around very rapidly until they form bonds with enough of their fellows to stop moving around on the substrate. The atoms being dropped on the substrate constitute a gas of such low pressure that the process is the equivalent of depositing one atom at a time onto the substrate, since each atom has time to defuse and stick to a neighboring particle before another atom is dropped in its vicinity. As time goes by, more and more atoms are deposited on the substrate, with the end result being a sample that is a number of angstroms thick, with a surface constituted

of a number of hills and hollows. Now that the mechanism has been explained, we shall move on to describe the actual process.

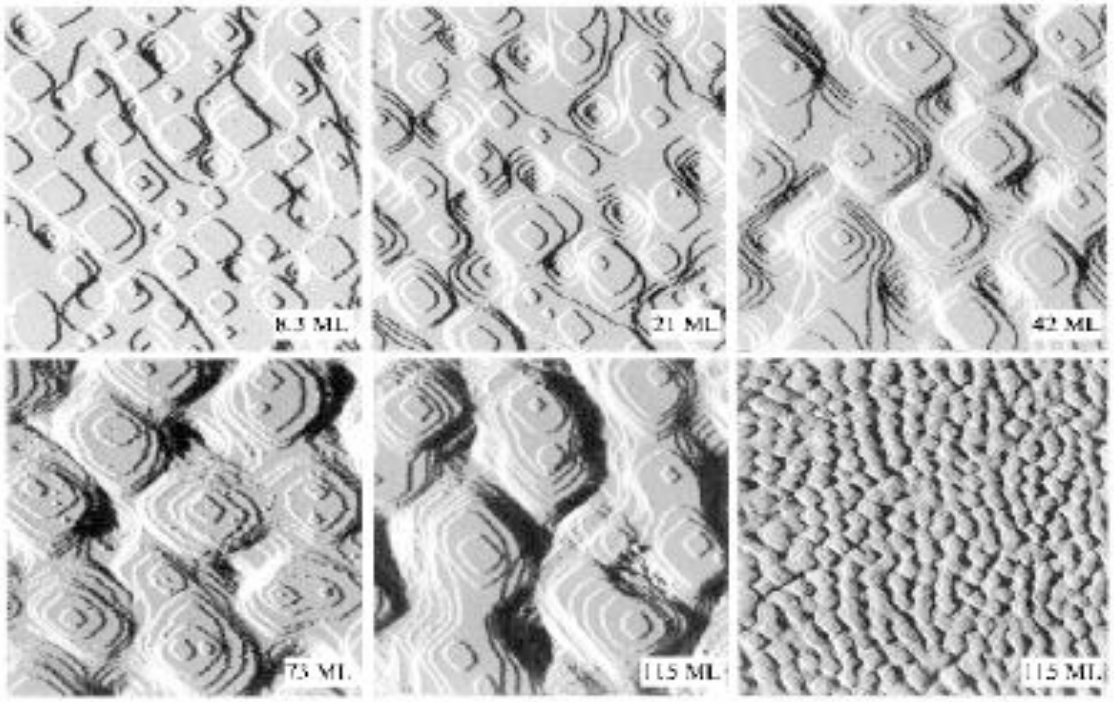


Fig. 1: Experimental results of epitaxial Cu film growth showing mound formation.

Methods

After a number of false starts and a long development period, we finally settled on our current project, which involved epitaxial growth with shadowing and diffusion in three dimensions, as stated above. By setting various options in our program, we were able to simulate a large range of conditions, which allowed us to see how the system developed with different restraints.

It all starts simply enough: with a flat substrate, generated when the program first starts to run. Our model simulates body-centered lattices, which means that each atom "sits" in the hollow created by the four atoms below it. Following, you'll find an illustration that depicts this model. You should be able to clearly see what this type of lattice looks like on an atomic scale.

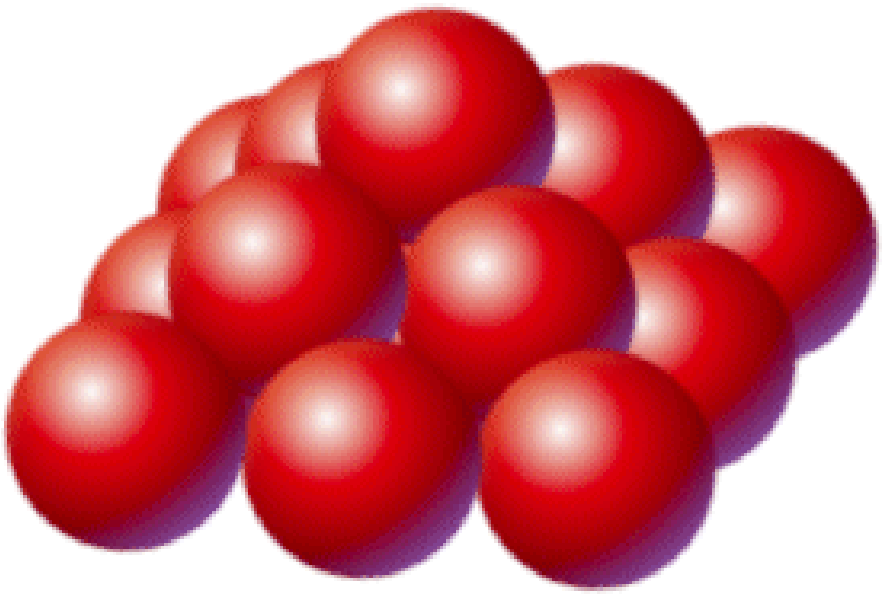


Fig. 2: A diagram showing atoms resting in the hollows created by the layer below them.

The user has the capability to adjust a number of different parameters. The system size, which was essentially the number of atoms in a row, is one such parameter. For instance, the system depicted above has a size of three, since this is the number of atoms in a row on the first layer (the substrate). In addition, the speed of the falling particles has two settings from which to choose: slow and fast. There are a number of barriers to be set, as well. The first is a barrier that determines how difficult it is for atoms to travel down a layer. The atoms will never go up layers, but depending on how large this barrier is made, they can move down layers. We did trials ranging from a moderate barrier of 0.07 to a large, practically insurmountable barrier of 0.6. The barrier inhibiting edge diffusion can also be changed. For the most part, we used a moderate barrier of 0.1, but we did make a couple runs with a very large barrier of 0.6. Finally, the program includes the option to turn on and off corner diffusion. For the most part, we left this on, which means that atoms can diffuse around corners with no difficulties. However, when turned off, an insurmountable barrier is inserted at the corners, prohibiting atoms from moving around corners. Of course, the number of layers to be deposited can be set, as well,

allowing the user to deposit anywhere from a fraction of a layer to a large number of layers. This makes it possible to study the system at many different times.

Finally, we come to one of our most important parameters: the type of deposition. The choices are vertical, uniform, and cosine. In vertical deposition, the atoms fall at randomly selected points straight down on to the sample. Although this is the model that has been used the most in the past since it is the easiest to simulate, it is also the least realistic, and it was this need for realism that drove us to include the other two types of deposition in our model. Which brings up our next type of deposition: uniform. In this case, the atoms can fall from any angle that lies between the vertical and the ninety degree arc that reaches to the substrate, as shown in the illustration below. In the uniform

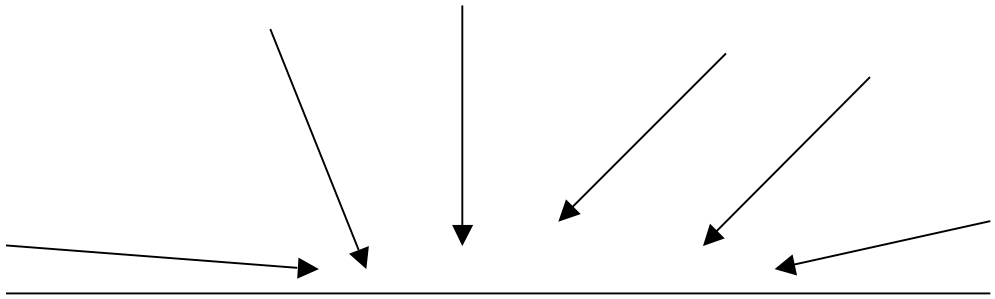


Fig. 3: The arrows represent the possible angles for an incoming atom for both the uniform and cosine distributions.

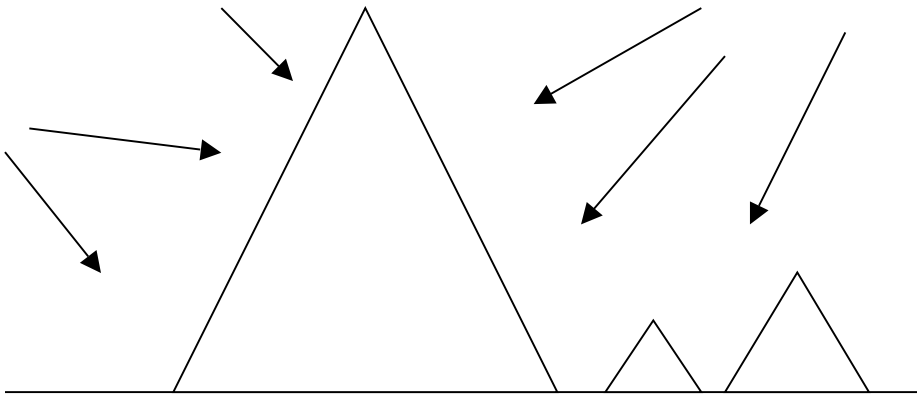


Fig. 4: This drawing shows the effect shadowing has on a system. As can be seen, only the particles that appear above the little mounds and fall practically straight down will avoid hitting the large mound.

case, every angle has an equal probability of being selected by the program. Because of this, shadowing by larger mounds quickly becomes a factor, causing one or two mounds to "blow up," totally dominating the system. This effect is depicted above. The final method of deposition employed by our program is the cosine distribution. This is the most interesting of all, since it corresponds to a uniform flux, which is the most realistic of all these cases. The cosine distribution is similar to the uniform distribution in that all angles are possible, as depicted in figure three. However, all angles are not equally probable in the cosine distribution. It is much more likely for an atom to fall with a vertical or near vertical trajectory than with an angled path, and the probability quickly falls to zero as the trajectory approaches ninety degrees to the vertical.

At this point, it is also worth noting that our program employs periodic boundary conditions. This means that if an atom would fall or diffuse off of one side of our sample, it instead appears in the same place on the other side of the model, as if the two edges were connected. This allows us to avoid oddities created by infinite barriers or other methods designed to stop the atoms from moving out of the sample.

So, to put it simply: one by one, atoms fall on our substrate. They diffuse around, following our initial conditions (barrier, deposition angle and speed, etc.), until they bond with other atoms that have been deposited on the substrate. Gradually, layers build up, one at a time, and before too long, mounds appear and coarsen. When, at last, we have deposited all the atoms we want, the program ends, and we analyze our data. So, now that it is clear how our program works, let us proceed to look at the results of our simulations.

Results

After enough false starts to fill a record book, followed by more errors than you could shake a stick at, our final program was finally running to our satisfaction, and producing results that were believable, if not exactly what we had initially expected. We used our model to simulate a number of different cases utilizing the uniform and cosine distributions, each of which I will present below. We didn't use our program to simulate any cases involving vertical deposition because these cases have all been studied extensively already, and we wanted to explore some territory that is not as well mapped out.

It has been found that equations describing some of the important parameters of the growing mounds are simple power law fits. These equations are: $W \sim t^{\beta}$ and $r_c \sim t^n$. The first equation describes how the root mean square of the width (W) of the mounds changes with time (t). The exponent (β) is referred to as the growth exponent. The second equation models the change in the lateral length scale (r_c) with time. Its exponent (n) is referred to as the coarsening exponent. Knowing these exponents provides us with valuable information, and that is where our simulation comes into play.

First on our list was the uniform distribution. Most of these ended up looking very similar, even though we used a number of different conditions to get our data. Following are some of these graphs, complete with an explanation of their parameters.

To start with, figure five shows some pictures of the development of the mounds. In the first picture, there are many mounds beginning to form. However, by the time 300 monolayers (ml) have been deposited, one mound is clearly overshadowing the rest and dominating the system. We did a number of different cases using a uniform distribution and depositing 300 ml, but they all yielded very similar results. By looking at the pictures (all of which look like the picture on the left in figure five), we can surmise that this is

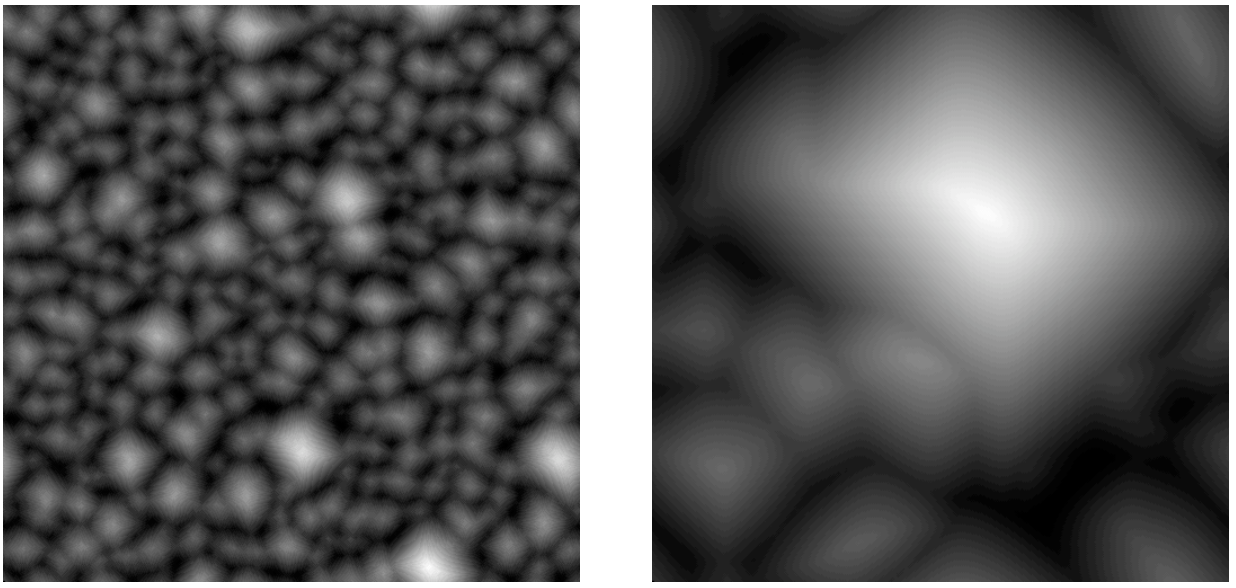
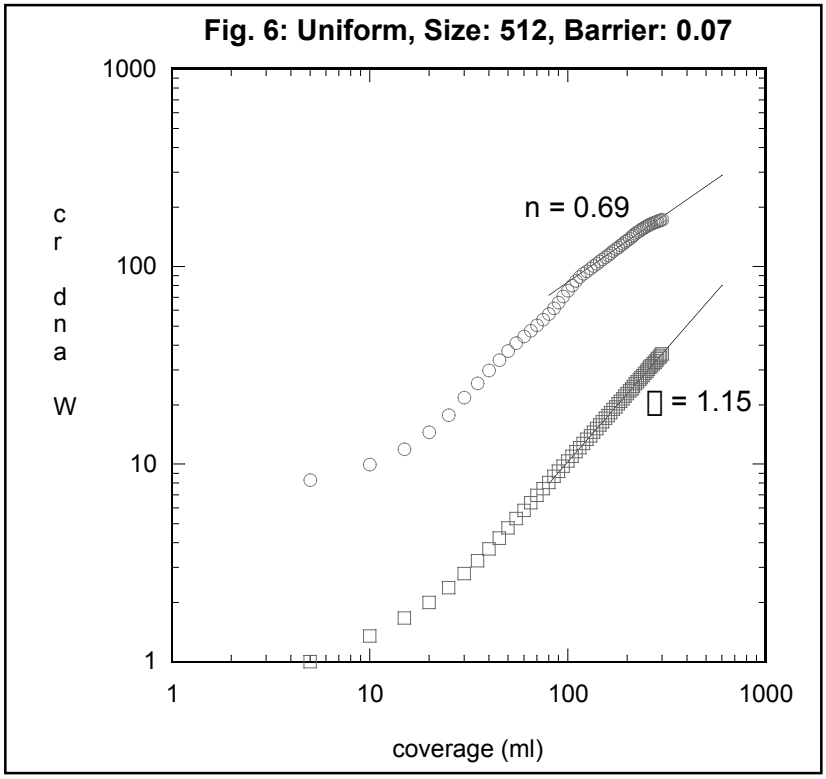
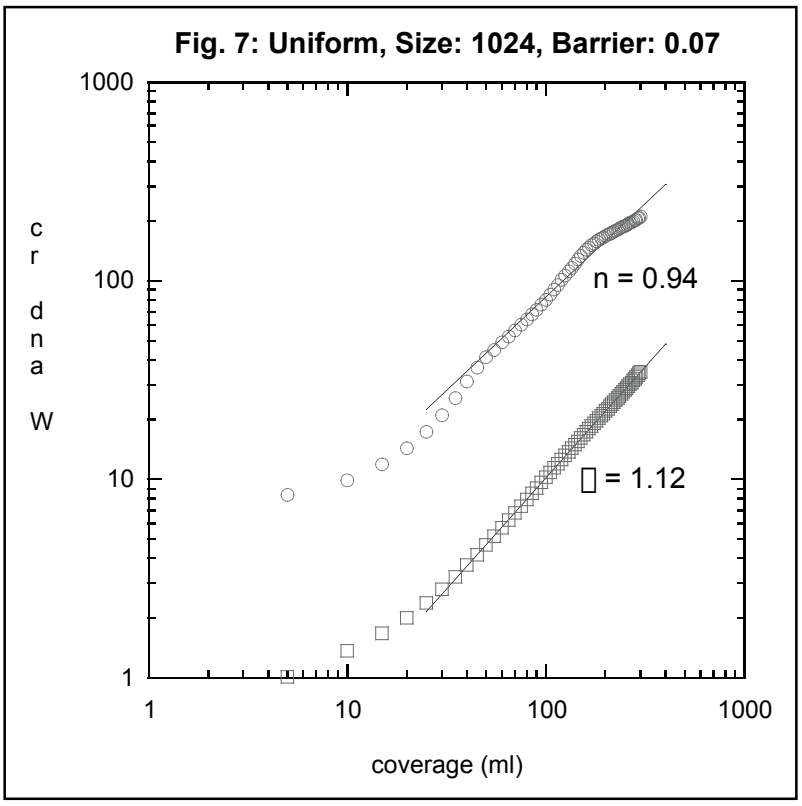


Fig. 5: The pictures here are of system sizes of 512, with fast deposition and a uniform distribution. There was no barrier to edge diffusion and a large barrier to prevent particles from traveling down layers. The picture on the left is of the sample at forty monolayers (ml), while the picture on the right is of the same system at 300 ml.

due to finite size effects, in which the system is giving inaccurate results because of inherent limitations in the size of our model. A representative graph, made of a run that had a system size of 512 and used fast deposition with a moderate barrier to particles moving downward, is displayed in figure six. As in all such graphs, the number of monolayers deposited is displayed on the x-axis, with the rms width and lateral length scale plotted on the y-axis. It is clear that the growth exponent has already reached a value of one, which signifies instability. It also appears that the coarsening exponent was



on its way to one before the finite size effects started affecting the system. To assure ourselves that our results were accurate, and not being affected by the small size of our system, as we suspect, we made another run, similar in all ways to the run depicted in the graph, but with a larger system size of 1024. The results from this run are shown in figure seven. As can be seen, really is going to one and then staying there, and n is in fact approaching one. This means that, in all likelihood, this system is unstable, and therefore less than totally interesting. So, it is at this time that we would like to move on to our



numerous cosine simulations. It is our fervent hope that we will find some more interesting results there.

Our results from the cosine runs we made run the gauntlet from exactly what we expected to the downright bizarre. To start our analysis, we will begin with the one of the last graphs we made, since it is undoubtedly the most accurate. The parameters for this graph were: a system size of 512, with slow deposition and a large barrier preventing particles from diffusing downwards. We deposited one thousand monolayers and analyzed our results, which were some of the best we had gotten so far. We were able to make a power law fit to the last six hundred layers deposited and get a perfect line for both n and , indicating that our sample had stabilized and was likely going to keep the same exponents until so many layers had been piled on that finite size effects started to ruin the data. Making our results appear even more credible, our coarsening exponent of 0.70 was identical to the exponent that Ryan Snyder found when studying data from an actual experiment in 1998. Our growth exponent of 0.93 was a bit bigger than his, which was 0.82, but these two values aren't so far apart as to cause a real problem. Two pictures of the system, one while it had only five hundred monolayers deposited and the other

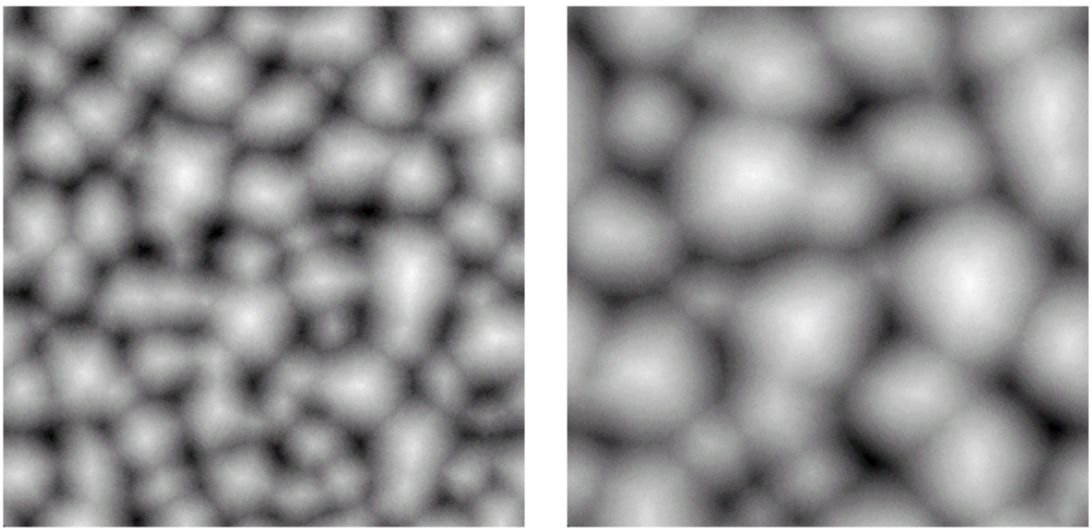
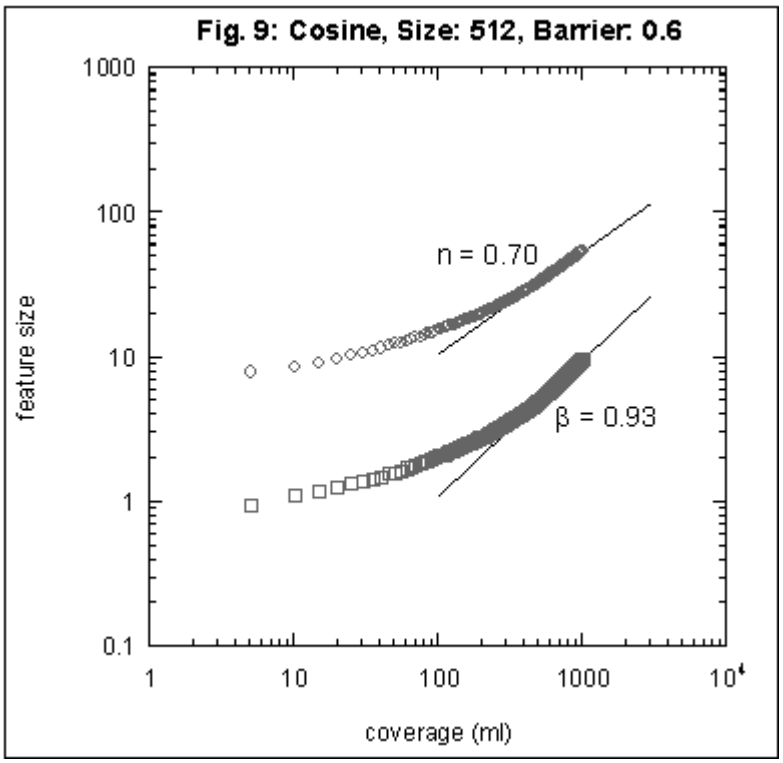
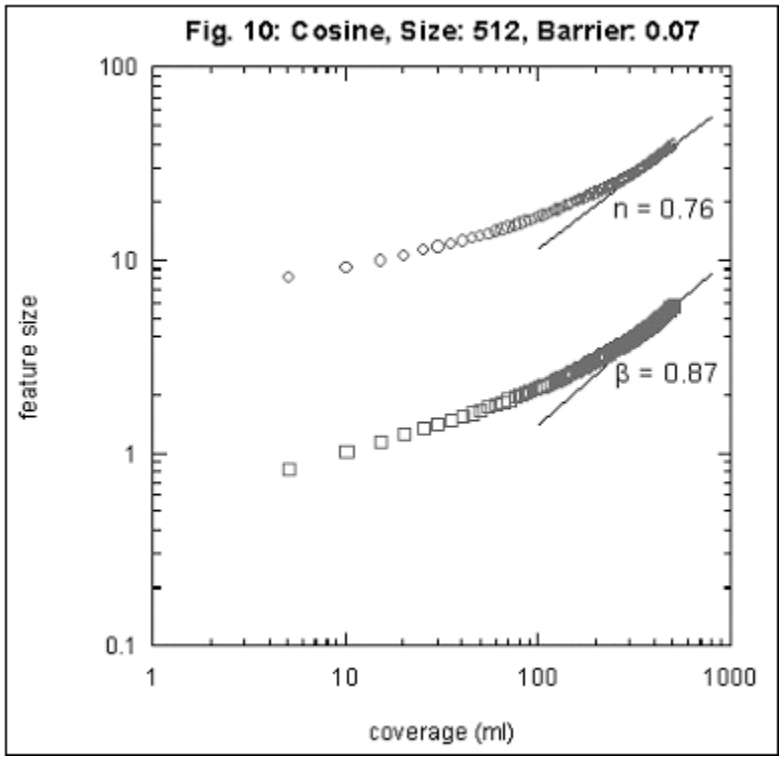


Fig. 8: This system's size is 512, and the particle's were deposited in a cosine distribution using slow deposition and a large barrier to prevent particles from traveling downwards. The picture on the left depicts the system after 500 ml have been deposited, whereas the picture on the right shows the sample after 1000 ml have been deposited.



when it had completed its run, at one thousand monolayers, are shown in figure eight. The graph of this system, which was mentioned above, is shown in figure nine.

However, we tried many different sets of parameters throughout the course of our investigation, with each system yielding different results. A representation of one of these other trials follows. The graph in figure ten is of a system with a size of 512, which uses fast deposition and a cosine distribution. It has a moderate barrier to downward diffusion and we deposited five hundred layers to get this graph. As can be seen, these exponents



aren't quite as good as the ones we achieved above. Still, values of n = 0.76 and = 0.87 aren't too bad. In fact, the width exponent is actually closer to Ryan Snyder's value.

Some of our runs gave us very interesting results. In one of our final systems, we made our sample size 512, as usual, used slow deposition with a moderate barrier for atoms that tried to move down a layer, and deposited five hundred monolayers. The interesting thing however, was that we only allowed atoms to be deposited at an angle of 75 degrees from the vertical. This led to a very structured system, as shown in figure

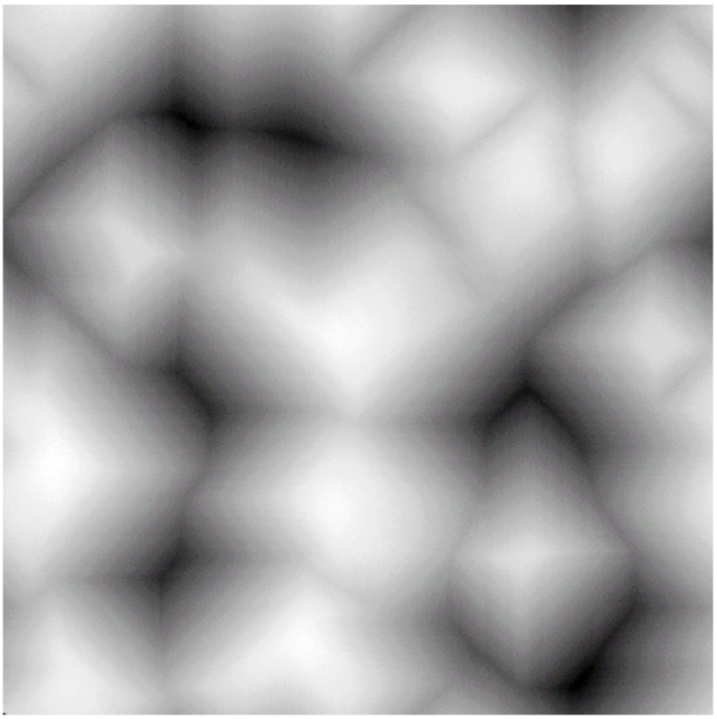


Fig. 11: This is a picture of a run in which atoms were deposited for 500 ml at an oblique angle of 75 degrees.

eleven, as well as an unstable system, with a coarsening exponent of 1.00 and a growth exponent of 1.08. This is clearly a system that merits more investigation.

Conclusion

After a number of simulations, we now know quite a bit more about the ways in which shadowing and diffusion affect thin film growth. It seems clear that in the uniform case, our values of one for both the coarsening and growth exponents indicate an unstable system. Our values for the cosine case, namely 0.70 for the coarsening exponent and about 0.90 for the growth exponent, indicate that the system doesn't appear to be unstable. However, with more simulations, it may be that the n and values go to one for the cosine distribution, as well. All in all, our results were quite satisfactory, and, although more simulations can certainly be run, we did a pretty good job of covering many general cases corresponding to real-life systems.

References and Acknowledgements

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[1] Snyder, Ryan. Atomic Force Microscopy Analysis of Microstructure Evolution and Morphology of Magnetron-Sputtered CdS Thin Films. 1998.