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A Computer Simulation of Thin Film Nucleation and Growth—The Volmer-Weber Case

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A COMPUTER SIMULATION OF THIN FILM NUCLEATION AND GROWTH - THE VOLMER-WEBER CASE

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SUMMARY

The computer simulation of thin film nucleation and growth, which was previously performed for the case of single monolayer, was modified to include multilayer growth via the Volmer-Weber mechanism. The main conclusions drawn from the simulation results are:

1. The kinetics of multilayer film growth is nearly identical to that of monolayer growth.

2. When no reevaporation takes place, the cluster density resulting from multilayer growth is higher at high coverage than that resulting from monolayer growth.

3. When reevaporation does take place, the cluster density resulting from multilayer growth is nearly identical to that resulting from monolayer growth. This is not due, however, to similarity in microstructure.

INTRODUCTION

Most studies of thin film nucleation and growth by means of computer simulation reported in the literature (refs. 1 to 3) are limited to a single monolayer, i.e., to the early stages of growth by the Frank-van der Merwe and Stranski-Krastanov mechanisms. In these two mechanisms a monolayer of the deposited material is formed before multilayer growth takes place. Only one study (ref. 4) which deals with multilayer growth has been published, and no comparison of the growth behavior under these two different modes of growth has been reported.

Multilayer growth is, however, very common and has been observed in many cases (see for example ref. 5). In fact, it constitutes one of the three basic modes of film growth, namely the Volmer-Weber mechanism, whereby multilayer

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growth takes place from the very beginning. Thus, a study of the growth behavior via this mechanism, and especially comparison with the other mechanisms, seems to be warranted.

DESCRIPTION OF THE MODEL

In order to allow comparison between the results obtained earlier (ref. 3) for the case of monolayer growth and those of the present study, the model employed here was made as similar as possible to the one employed there. Thus, the sample lattice selected for this study was a square lattice, and most of the basic assumptions are identical, namely:

1. Only single atoms impinge on the surface

2. Only single atoms (i.e., atoms which do not have any nearest neighbors) are capable of migrating on the surface.

3. Mobile (single) atoms migrate on the surface by means of jumps to nearest-neighbor sites and the jump frequency is constant.

4. A cluster is formed by two or more atoms occupying adjacent lattice sites.

5. Only nearest-neighbor interactions are taken into account.

6. No decomposition of clusters takes place.

7. Only single atoms can reevaporate from the surface.

8. The substrate surface is free from sites of preferred nucleation, e.g., defects or impurities.

9. The boundary conditions taken are of an infinite potential barrier, i e., atoms cannot cross the boundary.

For a discussion of these assumptions see reference 3.

PROCEDURE

The simulations were carried out on a microcomputer. A sample lattice of 400 (20 by 20) points was used. Some test runs were also done on a 1600 (40 by 40) point lattice, in order to check the effect of sample size. The results obtained in these test runs fell within the error range of the results obtained with the 400 point lattice.

The x and y coordinates of the sites at which atoms impinge on the substrate surface were chosen by means of a random number generator with homogeneous distribution. An atom can jump in four possible directions on a two-dimensional square lattice. The actual direction of each jump was also chosen by means of a random number generator. In cases involving reevaporation, the following procedure was used to determine whether a given (single) atom reevaporates at a given time: a random number in the interval (0,1) was generated.

If this number was smaller than the preassigned reevaporation probability, that atom was "erased."

Eight simulation runs were carried out for each case studied, and the average as well as the standard deviation were calculated. In order to check the sufficiency of the number of runs, 16 simulation runs were performed for some cases. The results were within the error range obtained with eight runs and no reduction in standard deviation resulted.

RESULTS AND DISCUSSION

In order to study the effect of each of the processes involved on the overall growth behavior, the simulation was done for each of the following cases.

Impingement

First, the case when the only process taking place is impingement of atoms on the surface was studied. This corresponds to the physical situation that exists when a film is deposited on a very cold substrate. The three-dimensional microstructure resulting from this process on a 40 by 40 point lattice is represented in figure 1 for four values of the coverage. The percentage of atoms bound in clusters out of the total number of impinged atoms is given, as a function of coverage, in figure 2, where the results obtained for the monolayer growth are also given for comparison. It can be seen that the kinetics of growth in these two modes is nearly identical, although at coverage lower than 0.4 a slightly higher fraction of the impinged atoms is bound in clusters in the case of multilayer growth.

This is not true, however, for the cluster density. From the results presented in figure 3, it is seen that at coverages higher than 0.2 the cluster density resulting from the multilayer growth is substantially higher than that resulting from monolayer growth. Thus, although these two mechanisms do not differ much in their kinetics, they do differ in the resulting microstructure.

Impingement Combined with Surface Migration

Next, the case when impingement is combined with surface migration was studied. This represents the situation when a film is deposited on a substrate which is hot enough to allow surface migration, yet is too cold to allow reevaporation of deposited atoms. The microstructure resulting from this combined process on a 40 by 40 point lattice is presented in figure 4. The kinetic results presented in figure 5 display a behavior similar to that obtained for the case of impingement, namely the kinetics of growth in the multilayer mode is almost identical to that of the monolayer mode. Again, as in the case where only impingement takes place, this is not true for the cluster density. The results presented in figure 6 show that at a high coverage multilayer growth results in higher cluster density than that resulting from the monolayer mode.

Combined Impingement, Surface Migration, and Reevaporation

Finally, the process of reevaporation of atoms from the substrate surface is also introduced. This represents the situation when a film is deposited on a very hot substrate. For the sake of clarity, the results of only one such case, namely when the probability of each single atom reevaporating at any given time is 0.5, are shown here. The microstructure resulting from this process on a 40 by 40 point lattice is presented in figure 7. The growth kinetics is given in figure 8, and, as in the former two cases, it is found to be almost identical for the two modes of growth.

Unlike the former two cases, however, the cluster density resulting from these two growth mechanisms, which is given in figure 9, is also similar. This similarity in cluster density is not due, however, to similarity in microstructure, since, as can be seen in figure 7, a considerable fraction of the atoms are located in "multilayer" positions.

CONCLUSIONS

The main conclusions of this study are:

1. The kinetics of film growth in the multilayer mode is nearly identical to that of the monolayer mode.

2. When no reevaporation takes place, the cluster density resulting from multilayer growth is substantially higher, at high coverages, than that resulting from the monolayer mode of growth.

3. When reevaporation does take place, the cluster density resulting from the two modes of growth are very similar. This is not due, however, to similarity in microstructure.

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3. J. Salik, (to be published in the Journal of Applied Physics.)

4. R.C. Feber, L.D.F. Allen and D. Grimmer, J. Vac. Sci. Technol. 8, 397-402 (1971).

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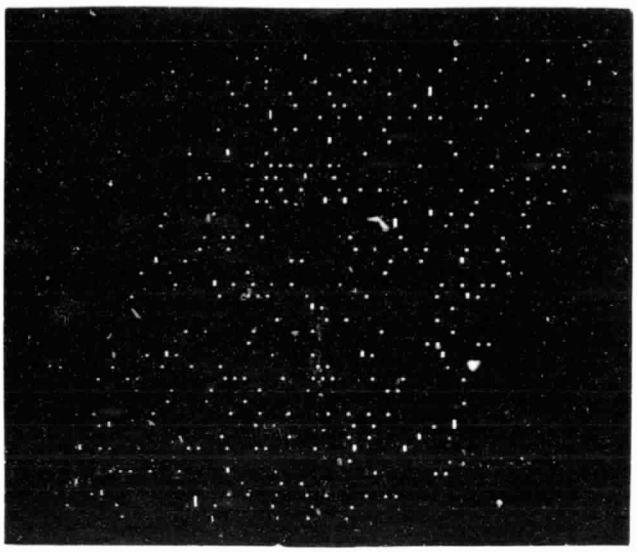

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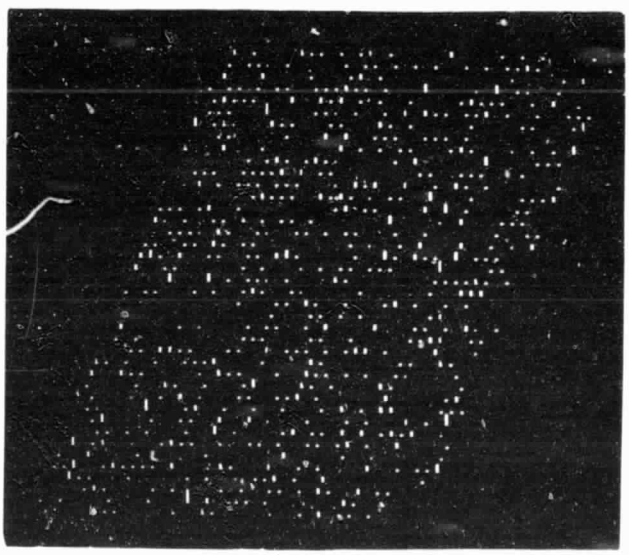

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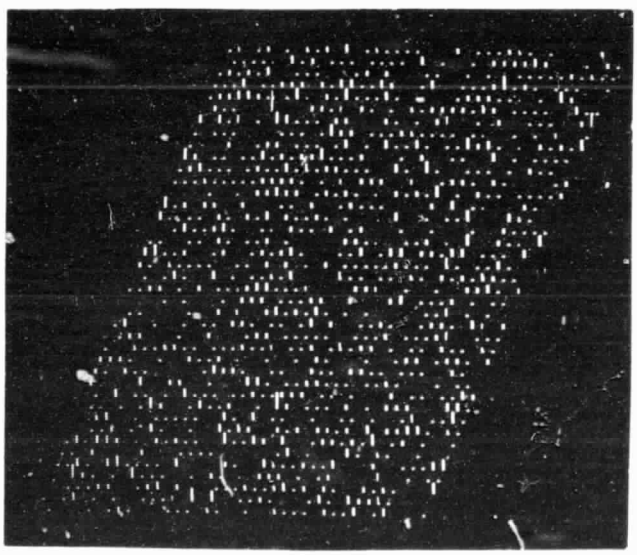

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Figure 1. - Microstructure resulting from direct impingement.

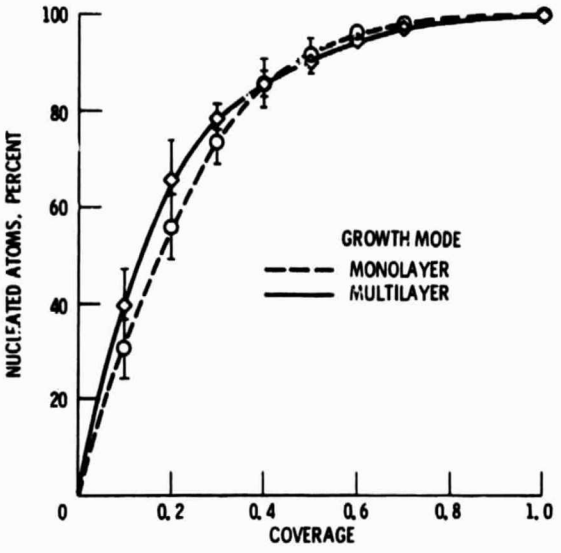


Figure 2. - Percent of atoms nucleated directly upon impingement as a function of coverage.

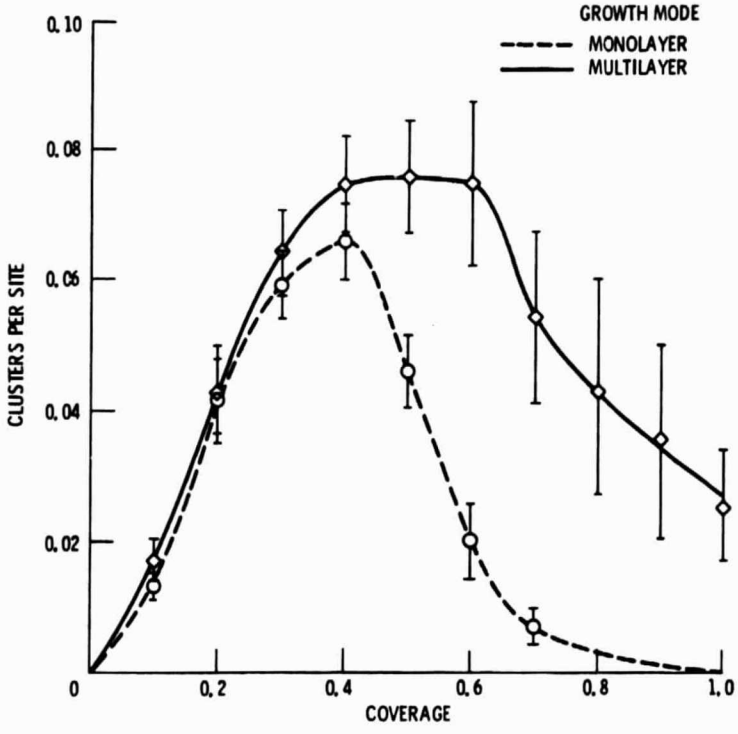
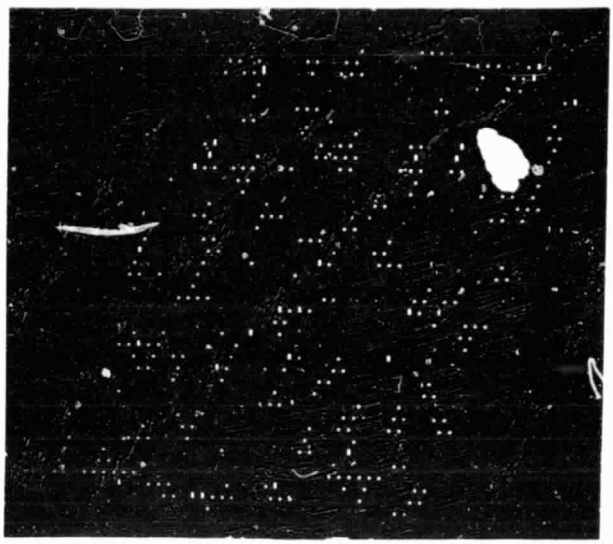
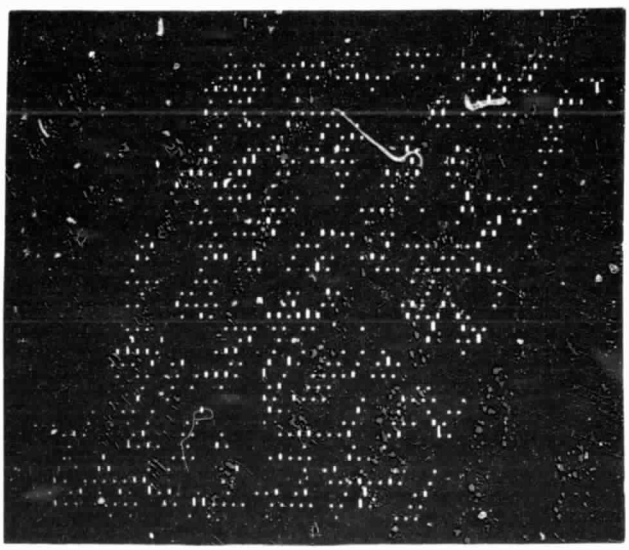


Figure 3. - Cluster density due to impingement as a function of coverage.

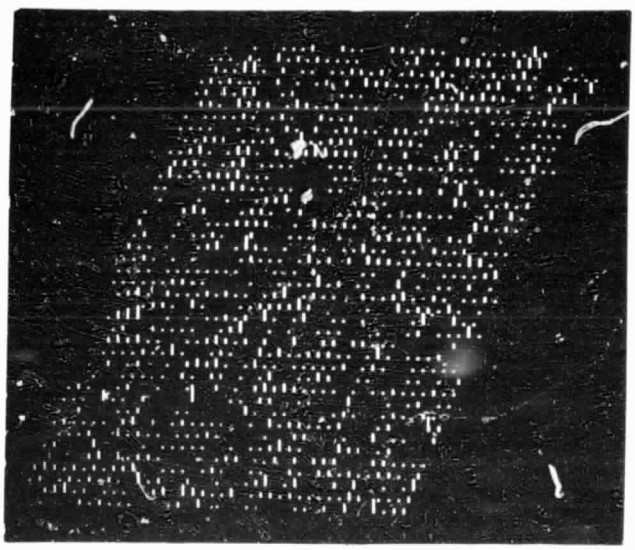
θ = 0.1



θ = 0.2



θ = 0.5



θ = 1.0

Figure 4. - Microstructure resulting from impingement combined with surface migration.

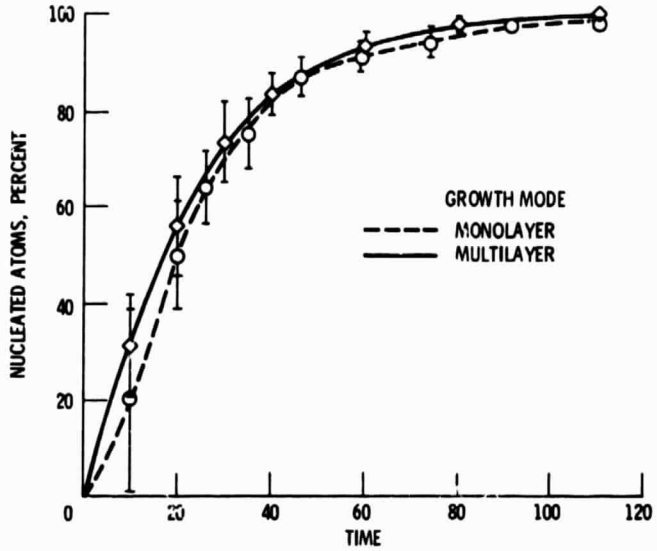


Figure 5. - Percent of nucleated atoms upon impingement combined with surface migration. Impingement rate, 2.5×10^{-3} atoms/(site x time unit); jump frequency, 1 per time unit.

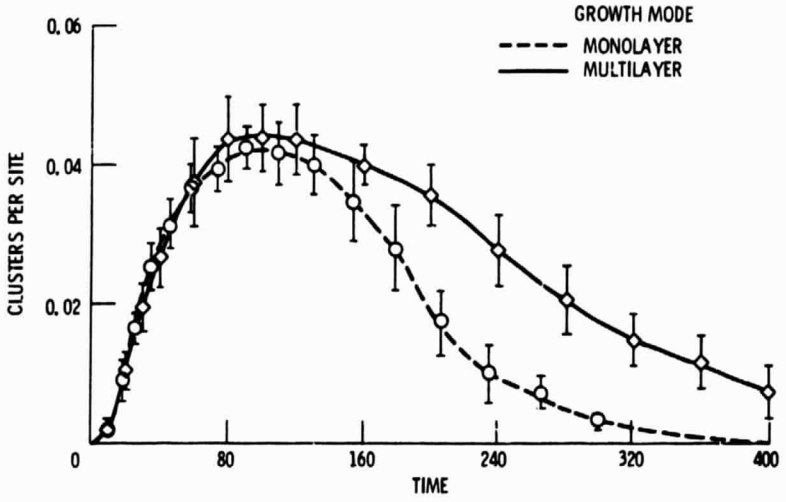
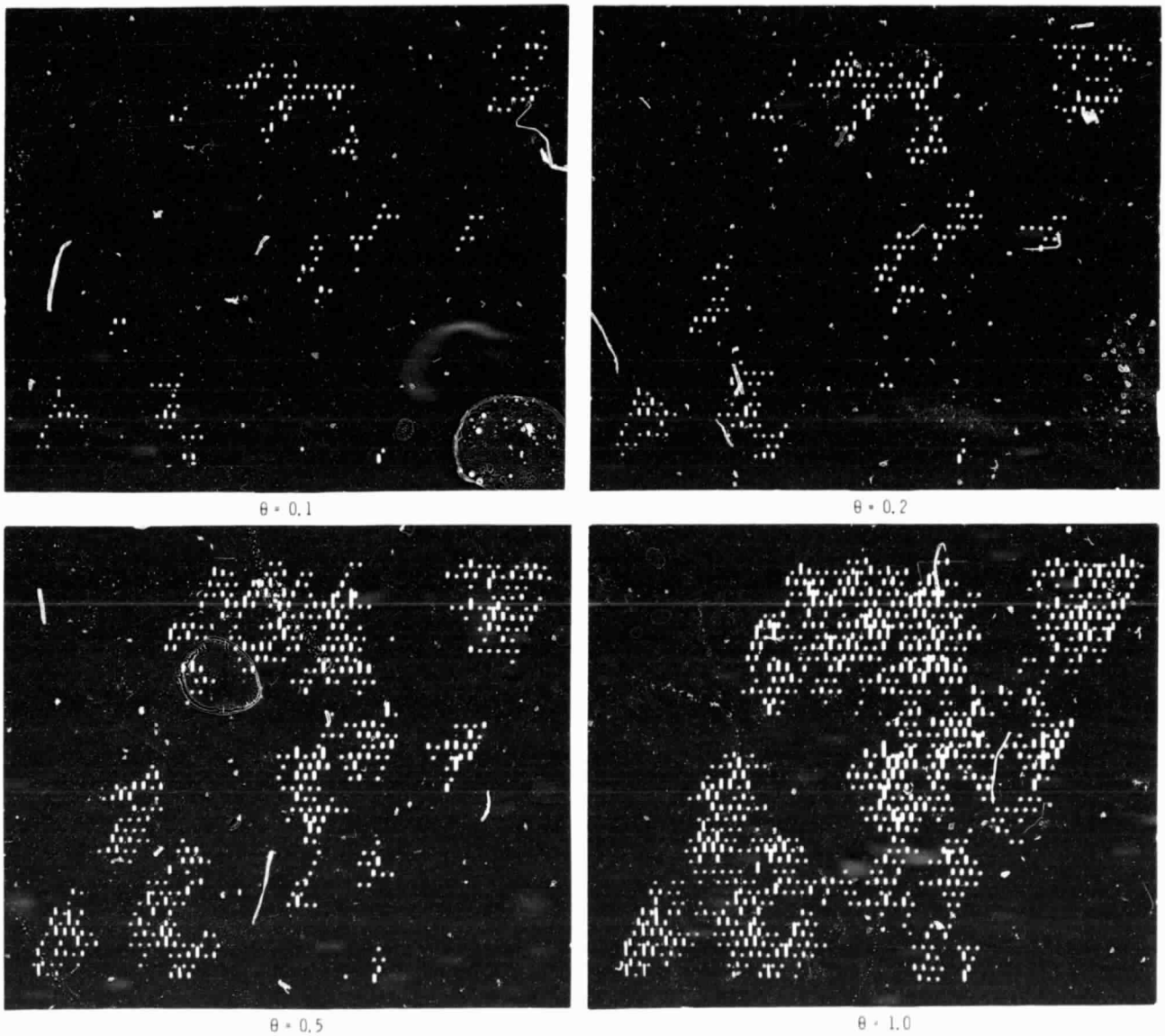


Figure 6. - Cluster density due to impingement combined with surface migration. Impingement rate, 2.5×10^{-3} atoms/(site x time unit); jump frequency, 1 per time unit.



θ = 0.1 θ = 0.2

θ = 0.5 θ = 1.0

Figure 7. - Microstructure resulting from combined impingement, surface migration, and reevaporation.

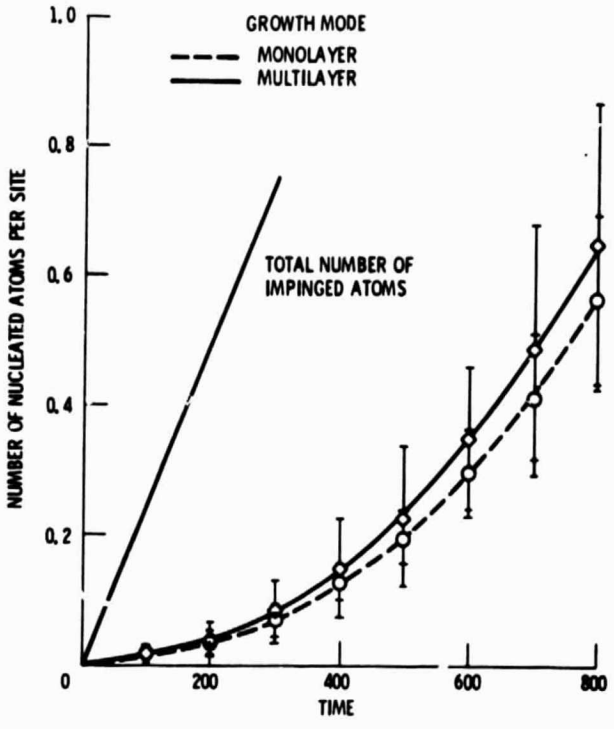


Figure 8. - Percent of nucleated atoms upon combined impingement, surface migration, and reevaporation. Impingement rate, 2.5×10^{-3} atoms/(site x time unit); jump frequency, 1 per time unit; reevaporation probability, 0.5.

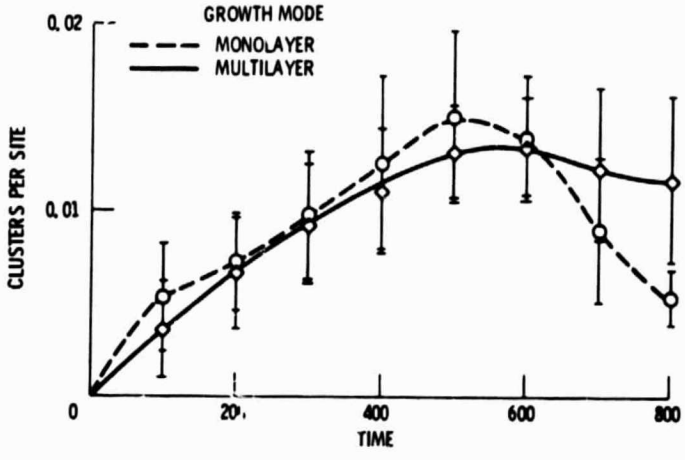


Figure 9. - Cluster density due to combined impingement, surface migration, and reevaporation. Impingement rate, 2.5×10^{-3} atoms/(site x time unit); jump frequency, 1 per time unit; reevaporation probability, 0.5.