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Form Approved  
OMB No. 0704-0188

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1. AGENCY USE ONLY (Leave blank)	2. REPORT DATE 3/19/96	3. REPORT TYPE AND DATES COVERED FINAL 01 NOV 91 through 31 OCT 95
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4. TITLE AND SUBTITLE Kinetics of Growth Phenomena at Semiconductor Surfaces Final Technical Report	5. FUNDING NUMBERS AFOSR F49620-92-J-0057  61102F 2303/FS
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9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) Dr. Michael R. Berman Air Force Office of Scientific Research/NL 110 Duncan Avenue, Suite B115 Bolling Air Force Base, D. C. 20332-0001	10. SPONSORING/MONITORING AGENCY REPORT NUMBER
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11. SUPPLEMENTARY NOTES

12a. DISTRIBUTION/AVAILABILITY STATEMENT Approved for public release; distribution unlimited.	12b. DISTRIBUTION CODE  <b>DISTRIBUTION STATEMENT</b> Approved for public release Distribution Unlimited
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13. ABSTRACT (Maximum 200 words)  
Our research is directed towards understanding the processes taking place during epitaxial growth of solid surfaces, with a view to potential control of growth. We use computer simulations and, when possible, simple models that are tested by simulations and by comparison with experiments. Issues include movement of atoms and of vacancies; nucleation processes, aggregation, and density; and the evolution of island shape. To take one example, we have studied, using theoretical modeling and simulations, early growth processes in Si(100), and predicted a complex scenario for the early growth, including a prediction that, under certain conditions, dimers will rotate, performing jumps of 90 degrees. These predictions have been confirmed by low-temperature STM experiments that we helped design, as well as by independent measurements.

19960408 077

14. SUBJECT TERMS	15. NUMBER OF PAGES 12
	16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT Unclassified	18. SECURITY CLASSIFICATION OF THIS PAGE Unclassified	19. SECURITY CLASSIFICATION OF ABSTRACT Unclassified	20. LIMITATION OF ABSTRACT UL
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20 MAR 1996

**Final Technical Report 1991-1995**  
**AFOSR F49620-92-J-0057**

**(a) List of Objectives**

The main purpose of our research is to understand the processes taking place during epitaxial growth of solid surfaces. We use computer simulations and, when possible, simple models that are tested by simulations and by comparison with experiments.

We have several broad objectives.

(A) We would like to understand the physical chemistry of shape formation during "self-organization": a central question is how atoms moving at random and without correlation with each other manage to reproducibly form islands having a well defined shape. Why are there such small errors in these shapes? And why are they so sensitive to the deposition conditions? And how can we use this knowledge to control the growth of these structures?

(B) Even the most advanced Kinetic Monte Carlo simulations are limited, now and in the predictable future, to samples of relatively small sizes and whose evolution can be followed for relatively short times. They are perfectly adequate for explaining STM observations made on a scale of ten thousand Å and a time scale of minutes. However recent experimental developments such as LEEM (low energy electron microscopy), photoelectron scanning microscopy, ellipsometric scanning microscopy, etc., provide observations of extremely intriguing phenomena taking place for hours, on a space scale of a micron. These phenomena involve massive material migration on the surface causing the formation of structures that cannot be explained by the known diffusion equations, and whose simulation by atomistic models would require computer power which will not be available in the predictable future. For this reason I became involved in research whose aim is to develop new nonlinear phenomenological equations that can describe these processes. We use atomistic simulations to establish what correct form of the equations. Once this is done, we can use them to compute the long-time behavior of large systems.

(C) An important outcome of the understanding provided by our work is learning how to control the shapes of these aggregates. There are practical reasons

for seeking such control at this time. Solid state electronics is moving relentlessly towards smaller and smaller devices, to increase storage density and computing speed, and to reduce power consumption. More remote in the future is the goal of integrating, on a single chip, optical and electronic devices. In all these tasks the shape of the active region and the perfection of its interface are essential. A well chosen shape improves electron transport and photon emission. A perfect interface means higher conductivity, enhanced luminescence, and reduced energy consumption. Moreover, if interface quality is not controlled, miniaturization will cause a severe degradation of performance. Up to now, progress in the growth of small and complex solid state structures has relied exclusively on empirical research. It is hoped that a better understanding of crystal growth will allow us to optimize device performance and perhaps imagine new growth methods and new types of structures, having new functions.

**(b) Status of the Research Effort**

All our work is directed toward understanding processes taking place during epitaxial growth. However this research field is so rich in new phenomena that we need to organize our results into classes of related processes.

1) *Elementary kinetic processes in epitaxy*

To achieve an understanding of the atomic aggregation we need the activation energies for a wide variety of elementary atomic displacements on a surface, at the edge of an island and for intra-layer transport (atomic movements down from an island onto the substrate, moves across surface steps, etc.). Calculating these energies is extremely tedious and several groups throughout the world are engaged in such research (for example: Norskov in Denmark, Sheffler in Berlin, and Voter, Feibelman and Johnsson in the United States). In papers #4 and #10 we have contributed to this effort. One of the main observations is that the elementary atomic movements are not always simple site-to-site jumps but involve intricate exchange of places with other atoms.

## 2) *Processes taking place in the early stages of growth*

If we could grow a solid layer-by-layer, we could make a perfect solid by making each layer perfectly. This is why much effort goes nowadays into understanding the growth kinetics of a single layer. One would like to know how to get rid of vacancies, what controls the number of islands (or nucleation centers), and how one can influence them to generate a perfect layer.

2a) We have studied the early growth processes on Si(100). This is a complex, reconstructed surface for which we have predicted a "scenario" for the early growth: the incident atoms will land on top of the dimer rows and move rapidly along them; they will form dimers and strings with an orientation contrary to that observed in the high-temperature measurements available at the time we made the prediction; as the temperature is increased the dimers will move to new sites between the rows and we predicted their distribution among these sites; in addition we predicted that the dimers will rotate, performing jumps of 90 degrees. In collaboration with Lagally's group we have designed specific low-temperature STM experiments to test these predictions. They have all been confirmed by the measurements (paper #6) and by independent measurements performed by Bedrossian and by Wolkov. This is especially gratifying since some of our proposals had no precedent or analog in other systems and were counter-intuitive.

2b) One of the most important quantities controlling growth is the density of the islands formed by adsorbate aggregation. It is well established that high island density leads to layer-by-layer growth, which is a desirable feature in obtaining materials of the highest quality. The main method of interpreting island-density measurements is a scaling theory proposed a while ago by Venables and extended by many people recently. In paper #8 we pointed out -- based in part on what we have learned in performing the calculations reported in #4 and #10 -- that these theories have ignored, without justification, the mobility of the dimers formed by the atoms on the surface. We have included this effect in the scaling theory, tested our conclusions by simulations, and showed that the new scaling results provide a better understanding of the experimental data. This is an essential modification of the theory.

3) *New effects in adsorbate aggregation: anomalous diffusion and surfactant action*

It has been shown, for a variety of systems, that the growth of ultra-thin films can be very substantially affected by addition of a small amount of well chosen substances (called, somewhat improperly, surfactants). A very famous example is the ability of small amounts of Sb to cause the growth of smooth Ge films on Si(100). The X-ray scattering measurements of Vlieg have shown that Ag films that are notoriously rough, become very smooth if a very small amount of Sb is deposited on the substrate prior to growth. Later, the STM measurements of Behm have shown that Sb causes an enormous increase (a factor of a thousand) in the number of nuclei formed on the surface during deposition. Moreover, the magnitude of the effect depends dramatically on Sb concentration. This phenomenon had no explanation and no precedent in the literature. We have shown (#7), by computer modeling, that the effect appears because the Sb atoms are immobile and repel the Ag atoms deposited on the surface. Because of this, the Ag atoms no longer diffuse on the surface but undergo a type of motion that we call anomalous diffusion. The distance that such an atom covers in a given time is extremely sensitive to the amount of Sb on the surface and is much shorter than that covered when Sb is absent. We have shown that the effect of such a "random network of repelling impurities" is general and will appear in any second-order process in chemical kinetics at surfaces.

4) *The collective behavior of vacancies on Si(100) surfaces*

Recently the crystal growth community has become aware that vacancy motion in the top layer is sometimes as important as the motion of single atoms. Moreover, the vacancies aggregate and have interesting collective behavior just as the atoms do. In paper #9 we have shown that vacancies on the Si(100) surface have long-range repulsive interactions due to the elastic distortion they induce in the surface layer. These, together with the weak attraction between vacancies sitting on adjacent dimer rows, cause the vacancies to form very interesting patterns on the surface. Depending on temperature and vacancy density, the vacancies form a  $2 \times n$  reconstruction, or meandering "wavy lines", or a "gas" in which their positions are practically uncorrelated. Measurements by Zandvliet in Holland have confirmed these predictions.

5) *The kinetics of shape evolution: equilibrium and non-equilibrium phenomena*

There is no doubt that adsorbed atoms move randomly on the surface and their motion is uncorrelated. It is therefore a small miracle that they manage, when the growth conditions are right, to make a large number of flawless aggregates with well defined and reproducible shapes. Since one of the main ideas of modern nanoscale physical chemistry is to grow structures whose shapes favor certain opto-electronic properties, these types of phenomena are receiving much attention. The fact that small alterations in the growth conditions lead to substantial changes in the shape of the aggregates suggests that a better understanding of the kinetics of the shape-formation process might give us an opportunity of controlling growth and designing shapes. For this reason we have invested quite a bit of effort in understanding the factors affecting shape.

5a) In our earlier work we focused on the formation of Pt islands on Pt(111), for which Comsa's group provided magnificent data. They have shown that by growing at different surface temperatures they can produce an ensemble of triangles with the tip oriented along a certain crystal axis, or triangles oriented in the opposite direction, or hexagons. At low temperatures the same system makes fractal aggregates. We have explained the kinetic mechanism for this process, in a paper (#1) that has been much quoted and has been the starting point for a number of similar studies (by Norskov in Lyngby, Denmark, and Scheffler in Berlin) and even a thesis (Jacobsen in Lyngby)

5b) Later we have used a "generic model" (i.e. a model that has all the physical features of a real crystal but it does not attempt to represent a specific material) to study the evolution of a shape to equilibrium (#5) and determine the factors influencing this process. These simulations solve exactly the kinetics of atomic motion and are therefore "computer experiments".

5c) The Au(100) surface provides another example of shape created by growth. This is a surface that should have square symmetry. However, the atoms in the top layer prefer to reconstruct and form a surface whose density is 20% higher than that of the bulk. To increase the density the atoms take a pseudo-hexagonal structure on a square second layer. The gold islands grown on this surface are long

rectangles and show a "size quantization": they tend to be six atoms wide, or 12 atoms wide. In paper #11 we have provided an explanation for this behavior. This is too technical to go into here, but we believe that this kind of symmetry-breaking process will be commonly seen in heteroepitaxy.

6) *The meaning of thermodynamics and kinetics in small systems*

Both thermodynamics and chemical kinetics have been developed for and are applicable to systems having a very large number of macroscopically identical species. Scanning electron microscopy breaks this rule rather severely. Because of this we must address the following general question: how can we apply thermodynamics and kinetics to such measurements? Recently I have made an effort to provide systematic and logical answers to this question. Our first effort concerns the meaning of equilibrium shape, of line-tension (the analog of surface tension), and of the rate equations (those we teach in freshmen chemistry) when applied to a small island. We have shown that it is possible to introduce a statistical ensemble in which both thermodynamics and kinetics can be defined precisely. However, the smallness of the sample can cause large deviations from the predictions due to large fluctuations. The latter can be avoided only by either averaging the results of the measurements over many samples or by averaging over a long observation time. The fluctuations are unavoidable but are not a complete nuisance. If measured, they can be used to determine a variety of thermodynamic quantities that are otherwise difficult to obtain

7) *Atom evaporation from an island and the kinetics of the coarsening process: a new path towards efficient simulations.*

One can prepare an ensemble of islands on a metal surface and they will have a distribution of sizes and shapes. If these islands are heated, their shapes evolve towards an equilibrium shape. In addition, atoms can leave one island, diffuse along the surface, and stick to another island. This exchange of atoms leads to a redistribution of island sizes (i.e. the number of atoms in the islands). This process is called coarsening, since it is believed (with some reason) that the small islands will disappear and the coarser ones will survive. There are many approximate theories of

this process but they are difficult to believe and even more difficult to test. One would like to simulate coarsening to provide "computer experiments" that can test these theories. The coarsening process may also be practically important. One of the new lines of research in nano-electronics aims at making and using small magnetic islands on a solid surface, since they may serve as memory elements. The subject is controversial, as some people believe that small islands cannot support ferromagnetism. To study these issues one would first have to make islands with uniform sizes and shapes. One of the proposals is that coarsening might help achieve this goal.

7a) Our understanding of coarsening is limited by the fact that the process is too complicated to allow reliable computer simulation. The departure of an atom from an island (we call this process evaporation, even though the atom does not go into vacuum but onto the substrate's surface) is a very rare event. Prior to its occurrence the atoms undergo millions of site-to-site jumps along the edge of the island. The probability of evaporation depends of these jumps: to simulate evaporation properly one must simulate accurately the motion of the atoms around the edge. To follow coarsening one must examine hundreds of islands undergoing millions of evaporation events and for each island one must follow the atoms milling around the edges. This task will overwhelm the biggest computer and wear down the most patient programmer.

In paper #12 we have found a solution for this problem. We have proposed breaking the task in two stages. First, examine one island and measure the probability that the island loses an atom at a give time  $t$ . Then use this probability in a second program in which coarsening is simulated. In this second stage we need not follow the motion of the atoms along the island's edge: the only effect of this motion, as far as coarsening is concerned, is to cause evaporation with a certain probability. Since we have already measured this probability we can now regard an island as an object with no internal structure whose only function is to eject or trap atoms with a certain probability. A system like this can be simulated without extreme difficulties.

7b) In our study of evaporation we have discovered a strange relation. It is possible to define an evaporation rate constant  $k(N,T)$ , which depends on temperature and the number of atoms in the island. The dependence on  $N$  is

$$k(N,T) = f(T) N^{0.36}$$

The strange thing is that the exponent is independent of temperature and of the rates of the elementary processes in the model. An island having rates similar to those of Xe on Pt has the same exponent as one representing a metal island on a metal surface. Furthermore, in all previous work in which evaporation processes were invoked, it has been assumed that the exponent must be 0.5; the rate of evaporation was presumed proportional to the number of atoms at the periphery. Our simulations show that this assumption is not correct and that conclusions based on it must be revised. We are now exploring both the generality and the consequences of this discovery.

8) *The equations governing pattern formation on a large time and space scale*

The Low Energy Electron Microscope (LEEM), developed recently, provides real-time movies of surface growth on a micron space-scale and on a half-hour time-scale. The intricate phenomenology observed in this way cannot be understood on the basis of atomic-level simulations. We have recently proposed a set of nonlinear equations that seem to describe surface instabilities such as facetting. The procedure for deriving the equations is general and novel, and can be used as a starting point for finding the correct equations describing growth on a large scale and at long time. These could be used to fit experiments and extract thermodynamic information regarding the parameters controlling growth.

(c) **Publications stemming from work performed under the grant**

1. A Kinetic Mechanism for Island Shape Variations Caused by Changes in the Growth Temperature, S. Liu, Z. Zhang, G. Comsa, and H. Metiu, *Phys. Rev. Lett.* **71**, 2967 (1993)
2. Self-Organized Growth of Si Adatoms on Si(100) at Submonolayer Coverages, Z. Zhang, Y.-T. Lu, and H. Metiu, in *Proceedings of the 21st International Conference on the Physics of Semiconductors*, August 10-14, 1992, Beijing, China (World Scientific, Singapore, 1994)
3. Stability and Kinetics of Step Motion on Crystal Surfaces, F. Liu and H. Metiu, *Phys. Rev. E* **49**, 2601 (1994)
4. The Mobility of Pt atoms and Small Pt Clusters on Pt(111) and Its Implications for the Early Stages of Epitaxial Growth, S. Liu, Z. Zhang, J. Norskov, and H. Metiu, *Surface Science* **321**, 161 (1994)
5. Evolution to Equilibrium of the Shape of an Island Formed by the Aggregation of Adsorbed Atoms, H. Shao, S. Liu, and H. Metiu, *Phys. Rev. B* **51**, 7827 (1995)
6. Energetics and Dynamics of Si Ad-Dimers on Si(001), Z. Zhang, F. Wu, H. J. W. Zandvliet, B. Poelsema, H. Metiu, and M. G. Lagally, *Phys. Rev. Lett.* **74**, 3644 (1995)
7. Submonolayer Growth with Repulsive Impurities: Island Density Scaling with Anomalous Diffusion, S. Liu, L. Bönig, J. Detch, and H. Metiu, *Phys. Rev. Lett.* **74**, 4495 (1995)
8. The Effect of Small-Cluster Mobility and Dissociation on the Island Density in Epitaxial Growth, S. Liu, L. Bönig, and H. Metiu, *Phys. Rev. B* **52**, 2907 (1995)
9. Missing-Dimer-Vacancies Ordering on the Si(100) Surface, P. C. Weakliem, Z. Zhang, and H. Metiu, *Surface Science* **336**, 303 (1995)
10. Interlayer Diffusion and Motion of Adatoms in the Vicinity of Steps, S. Liu and H. Metiu, *Surface Science*, to appear (1996)
11. An Effective Medium Theory Study of Au Islands on the Au(100) Surface: Reconstruction, Adatom Diffusion, and Island Formation, L. Bönig, S. Liu, and H. Metiu, *Surface Science*, to appear (1996)
12. Evaporation of Single Atoms from an Adsorbate Island or a Step to a Terrace: Evaporation Rate and the Underlying Atomic-Level Mechanism, H. Shao, P. C. Weakliem, and H. Metiu, *Phys. Rev. B*, submitted (1995)
13. Equilibrium Shapes and Shape Fluctuations for Two-Dimensional Islands on a Solid Surface, H. Shao, P. C. Weakliem, and H. Metiu, *J. Chem. Phys.*, to be submitted

**(d) Professional personnel supported by the grant**

Dr. Charusita Chakravarty, Dr. Yan-Ten Lu, Dr. Yuri Dakhnovskii, Dr. Shudun Liu,  
Mr. John Detch

**(e) Interactions**

**Presentations at seminars, meetings, conferences**

1991

Coherence and Interference in Short Pulse Excitation of Dissociating Molecules:  
Institute for Theoretical Physics, Santa Barbara, CA, September 10

Surface Migration and Epitaxy, Kendall Symposium, ACS National Meeting, Atlanta,  
March

Simulations of Epitaxial Growth, Symposium on Dynamical Processes in Condensed  
Molecular Systems, Neve-Ilan, Israel, April

Simulations of Epitaxial Growth, AFOSR Meeting, Irvine, CA, October

1992

Simulations of Epitaxial Growth, Seminars at Rice University, January 20;  
University of Houston, January 21; Texas A&M University, January 22;  
University of Texas at Austin, January 23

Si Self-Organization on a Si(100) Surface: Seminar at UCLA, April

Rate Constants of Surface Reactions: University of California, Berkeley, May 21

Adsorbate Segregation During Epitaxial Growth: Seminars at the Tokyo Institute of  
Technology, September 9; and University of Tokyo, Tokyo, Japan, September  
28

Coherence Effects in Photodissociation with Short Pulses: Seminar at the Institute of  
Molecular Science, Okazaki, Japan, September 25

Simulations of Epitaxial Growth: Gordon Conference on Chemistry of Electronic  
Materials, Ventura, March

Growth and Self-Aggregation on Surfaces: Annual American Physical Society  
Meeting, Indianapolis, IN, March

The Theory of Rate Constants: NATO Workshop on Time Dependent Quantum  
Dynamics, Snowbird, Utah, March 30 - April 3

Si Self-Organization on a Si(100) Surface: European Research Conference on  
"Fundamental Aspects of Surface Science", Davos, Switzerland, June 8-12

The Theory of Rate Constants: Workshop on the Theory of Rate Constants, Telluride,  
July 12-17

Coherence and Interference Effects in Photodissociation with Ultrashort Pulses: 11th  
Canadian Symposium on Theoretical Chemistry, Montreal, Canada, August 2-7

Coherent Phenomena in Photoexcitation of Small Molecules: Symposium at ACS  
Annual Meeting, Washington, D.C., August 23-28

Simulations of Epitaxial Growth: Symposium at ACS Annual Meeting, Washington,  
D.C., August 23-28

Adsorbate Segregation During Epitaxial Growth: Annual Meeting of the Japan  
Physics Society, Tokyo, September 27

Adsorbate Self-Organization: Symposium on "Chemistry in Electronics and  
Photonics", The Industrial Affiliates Program, Stanford University, December 7

### 1993

- Self-Organization and Patterns in Epitaxy: Seminar, Department of Materials Science, University of Wisconsin, June 3
- Coherence in Photodissociation: Seminars at the Department of Physics, Freiburg, Germany, August 18; and the Freie Universität, Berlin, Germany, September 7
- Adsorbate Self-Organization: Seminar at the Surface Science Institute, Jülich, Germany, August 26
- Epitaxial Growth Simulations: Seminars at the Fritz Haber Institute, Berlin, September 9; and the Physics Department, Technical University of Denmark, Lyngby, September 10
- Island Shapes and Surface Roughness, AFOSR Contractors meeting, Irvine CA, Oct.
- Coherence in Photodissociation with Ultrashort Pulses: International Conference of Femtochemistry, Berlin, Germany, March 1-4
- Island Shapes in Epitaxial Growth: Surface Science Symposium, ACS Annual Meeting, Denver, Colorado, March 28-April 2
- Optical Properties of Electrons in Quantum Wells: Canadian Society of Chemistry, Sherbrooke, Canada, May 30-June 3
- Patterns in Crystal Growth: Freed Symposium, University of Chicago, October 1-2
- Coherence in Molecular Photodissociation: Optical Society of American Annual Meeting, Toronto, Canada, October 3-8

### 1994

- Epitaxial Growth: Island Shapes and Step Flow, Seminar at California Institute of Technology, Pasadena, May 10
- Epitaxial Growth: Island Shapes and Step Flow: 1994 Sanibel Symposium, Florida, February 17
- Island Shapes and Step Flow: 1994 March Meeting of the American Physical Society, Pittsburgh, March 21
- Kinetic Monte Carlo Simulations: CECAM Workshop on Computer Simulation of the Growth of Semiconductor Materials, Lyon, France, May 30-June 3
- Kinetic Simulations: Workshop on Dynamics in Condensed Matter, Telluride CO, July 18-24
- Kinetic Simulations of Surface Growth, US-Japan Gordon Conference on Fundamental Concepts of Semiconductor Surfaces, Hawaii, November 13-18

### 1995

- Spectroscopy of Color Centers in Zeolites, Winter Topical Meeting SPIE-Optical Society of America, Santa Fe NM, February 8-11
- Theoretical Study of Possible Applications of Wave-Packet Interferometry, ACS National Meeting, Anaheim CA, April 2-6
- Adsorption Aggregation on Surfaces, USC Surface Science Symposium in Honor of G. Ertl, April 26-28
- Kinetic Simulations of Atom and Vacancy Aggregation on Si(100), US/Japan Workshop on Atomic-Scale Mechanisms of Epitaxial Growth, Honolulu HI, May 10-12
- The Properties of Electrons Solvated in Zeolites, XIX International Workshop on Condensed Matter Theories, Caracas, Venezuela, June 12-17

The Evolution of the Shapes of Adsorbate Islands on Semiconductor Surfaces,  
Gordon Research Conference on Dynamics at Surfaces, Andover NH, August 6-11

Six lectures at the CECAM Workshop on Computer Simulation of the Growth of Semiconductor Materials, Lyon, France, August 28 - September 8:

- (a) The Exact Calculation of the Rate Constants Involved in Adsorbate Diffusion,
- (b) The Dynamics of Island Evolution in Metal-on-Metal Epitaxy,
- (c) The Early Stages of Si Deposition on the Si(100) Surface,
- (d) The Collective Properties of Missing-Dimer Vacancies on the Si(100) Surface,
- (e) The Role of Dimer Motion and Surface Reconstruction in the Scaling Properties of Island Densities in Epitaxial Growth of Au on Au(100),
- (f) A Model for Surfactant Action in Epitaxial Growth: Anomalous Diffusion and Peculiar Scaling Equations

**No consultative or advisory functions**

**(f) No inventions or patent disclosures**