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6. AUTHOR(S) B. H. Cooper				
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) Cornell University Laboratory of Atomic and Solid State Physics Clark Hall Ithaca, NY 14853-2501			8. PERFORMING ORGANIZATION REPORT NUMBER AFOSR-TR-97	
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13. ABSTRACT (Maximum 200 words) We have used Ultra High Vacuum Scanning Tunneling Microscopy (UHV-STM) to investigate the evolution of morphology on metal surfaces irradiated with energetic ion beams. Time-lapse images were used to observe the stability and decay of isolated islands and pits subsequent to the irradiation. We developed theoretical analytic models and Monte Carlo simulations to describe island decay. Key results are: 1) monolayer height islands and pits are formed in the early stages of energetic Ar irradiation of Au(111); 2) under certain experimental conditions islands decay over time; 3) analytic models of island decay based on the Gibbs-Thomson effect predict different decay rates and laws depending on the relative magnitudes of parameters that describe different macroscopic decay processes; 4) comparisons of the analytic model to simulations of island decay give insights into how these macroscopic decay parameters are determined by microscopic surface processes. These studies are part of a larger effort to probe mechanisms of thin film deposition with energetic ion beams; additional projects include i) the development of a new beamline and ultra high vacuum system for in-situ STM studies of thin films deposited with hyperthermal energy ions, and ii) scattering studies of the mechanisms of hyperthermal energy ion trapping at surfaces.				
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FINAL TECHNICAL REPORT

GRANT NO. F49620-93-1-0504 - AASERT

PARENT AWARD AFOSR-91-0137

PI: BARBARA H. COOPER
LABORATORY OF ATOMIC AND SOLID STATE PHYSICS
CORNELL UNIVERSITY
ITHACA, NY 14853

REPORT PERIOD: 9/1/93 - 8/31/96

PROJECT TITLE: ION SCATTERING AND DEPOSITION:
THE ROLE OF ENERGETIC PARTICLES IN THIN FILM GROWTH

SUMMARY OF COMPLETED WORK

PERSONNEL

The original AASERT award was entitled "Reactive Ion Scattering and Deposition: The Role of Multi-Channel Charge Transfer;" the student named to work on this project was Eric Dahl. Dahl subsequently received an AT&T Fellowship and is currently finishing his dissertation work. A change in personnel was requested and approved; graduate student James McLean was fully supported by the AASERT. The AASERT was also used for supplemental summer support for graduate students Eric Dahl, Aaron Judy, and Chad Sosolik. The new project title listed in the heading reflects this change in personnel; McLean, Dahl, Judy, and Sosolik are working on projects that are part of our overall research effort to study the use of energetic ions in thin film growth. McLean, Dahl, Judy, and Sosolik are US citizens; McLean has graduated with his PhD, and Dahl, Judy, and Sosolik are making satisfactory performance (including grades) toward the PhD.

OBJECTIVES AND OVERVIEW

Energetic ions or neutrals in the hyperthermal energy range (few eV to several hundred eV) are used in a number of thin film growth applications (e.g., sputter and plasma deposition techniques, direct ion beam and ion-assisted deposition, and pulsed laser deposition). These involve both direct deposition with an ion beam of the film species, and deposition by some other method during simultaneous ion bombardment. Experiments and simulations have shown that energetic ions can lower the substrate temperature required to achieve crystallinity, modify growth modes, change film morphology, and influence structure and crystallographic orientation in the film. In many cases, the mechanisms responsible for ion-induced modification of thin film deposition are not understood. We have initiated both scanning tunneling microscopy (STM) and scattering studies to probe these mechanisms.

In the early stages of ion beam deposition or ion beam sputtering, the morphology of the developing surface often exhibits islands and pits with characteristic size distributions. These pits and islands are nonequilibrium features, whose formation and subsequent stability depend on deposition (and/or sputtering) rates, temperature, and rates of different microscopic surface processes. We have formed

(using ion beams) and observed (with ultra high vacuum scanning tunneling microscopy - UHV-STM) nanoscale pits and islands, one to a few atomic layers in height, on Au(111). In parallel we have developed analytic models and Monte Carlo simulations of the post-formation evolution of pits and islands on terraces. These models were used to investigate the macroscopic rate-limiting processes controlling the stability and decay of nanoscale surface features, and how these are determined by microscopic surface processes. We have also designed a new ultra high vacuum system and ion beamline for studies of direct ion beam and ion beam-assisted thin film deposition. This beamline will be used with in-situ STM to directly observe the deposited films in order to determine how surface structure and morphology depend on ion energy, angle of incidence, and species. Finally, scattering measurements on model systems have been completed to determine trends in trapping mechanisms as a function of incident ion energy and angle for hyperthermal ion irradiation of metal surfaces.

TECHNICAL SUMMARY OF SIGNIFICANT WORK ACCOMPLISHED

This section summarizes results from projects in which we have investigated surface morphology evolution resulting from and subsequent to ion irradiation of metal surfaces (completed thesis work of James McLean), development of a new system for thin film deposition with hyperthermal ion beams (ongoing work of Aaron Judy), and trends in trapping mechanisms for hyperthermal ion collisions with surfaces (completed work of Eric Dahl and ongoing work of Chad Sosolik). These projects are part of a larger program to study the mechanisms of thin film growth with energetic ions. References to published work from these studies are given in the text.

Surface Morphology Evolution

Graduate student James McLean, who received full support from this grant, recently graduated with his PhD. His work had two major components [1]. The first was an experimental study using ultra high vacuum scanning tunneling microscopy (UHV-STM) to determine the effects of ion bombardment on surface morphology. The second was a theoretical study of the stability and decay of islands and pits that form on surfaces during ion irradiation and growth.

Ion-induced modification of surface morphology - experiment: We have made preliminary measurements of Au(111) surfaces irradiated with noble gas ion beams. Our motivation for using noble gas ions is two-fold: 1) they are used in ion-assisted growth, and 2) since they are nonreactive, we can assess how

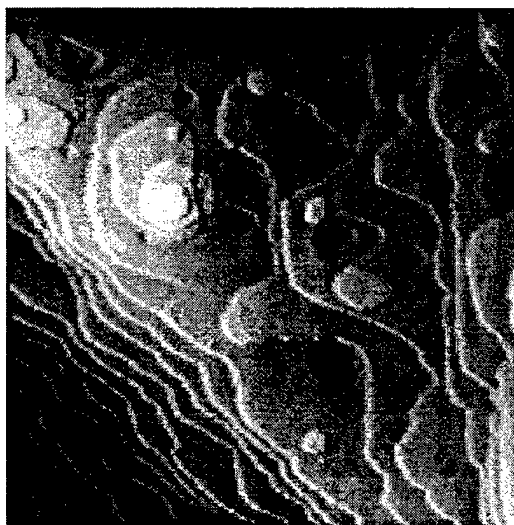


Fig. 1 Scanning tunneling microscope image of the surface of Au(111) following irradiation with 500 eV Argon ions at room temperature. The pits and islands are produced by the ion irradiation. The image size is $930 \times 970 \text{ \AA}$.

surface morphology evolves due to the “physical” aspects of the ion collisions (e.g., energy deposition, sputtering, and subsurface trapping), without the additional complications of “chemical effects”. Argon ion irradiation of Au(111) results initially in the formation of islands and pits (fig. 1). The islands and pits have characteristic size distributions which reflect the rates of different microscopic surface processes, such as production of adatoms and vacancies and their mobilities, and attachment and detachment at step edges. Experiments are ongoing to monitor the evolution of surface morphology as a function of ion beam energy, ion dose, sample temperature during irradiation, etc.

Significance: One of our goals is to better understand the role that ion beams play in modifying surface morphology during thin film deposition. These experiments are a crucial first step toward that goal; before we can understand ion-induced effects in growth, we must understand ion-induced modification of surfaces in the absence of deposition. These experiments probe the latter, giving insights into how surface modification depends on ion beam parameters. In addition, modeling of the surface evolution gives insights into the relative rates of different microscopic processes (e.g., mobilities of adatoms and vacancies, attachment and detachment rates at step edges, etc.) giving rise to the modifications.

Stability of surface nanoscale features - experiment and theory: We have also used time-lapse STM imaging to monitor the stability and decay of the islands and pits *after* they have been formed by hyperthermal ion irradiation [2,3,4]. We find that under some experimental conditions, pits merge with nearby step edges, nearby pits coalesce, and isolated islands on terraces decay over time. McLean, in collaboration with theorists Badri Krishnamachari and Jim Sethna, developed a Monte Carlo code for simulating the behavior of islands and pits [1,5,6,7]. The simulations were used for three purposes: 1) to test analytic thermodynamic models (our own and others in the literature) of the decay of nanoscale surface features, 2) to study the fundamental microscopic processes that determine stability and/or decay, and 3) to model specific geometries observed in STM experiments. Key results are presented briefly below.

In analytic thermodynamic theories of island decay, the driving force for the decay is the Gibbs-Thomson (GT) effect, which says that the vapor pressure of adatoms in equilibrium with a curved step edge (the island) is larger than that in equilibrium with a straight step edge (the terrace edge). (The same theory will apply to pits where the mobile species are vacancies.) On a surface with islands of various curvatures, the GT relation implies that a concentration gradient will be established whereby adatoms diffuse away from high curvature features toward low curvature features. Thus an isolated island surrounded by steps with lower curvature will decay. The three macroscopic processes involved in the flow of atoms from the island are interface transfer, diffusion, and incorporation of atoms at the outer boundary. The relative magnitudes of these processes determine which is the rate-limiting step in island decay; the rate and time-dependence of the decay are different for the different rate-limiting processes [6,7]. Under certain simplifying approximations, the decay will follow the simple power laws often cited in the literature; $A \propto (t_0 - t)^n$, where $n = 2/3$ corresponds to diffusion-limited decay, and $n = 1$ to interface transfer-limited decay. Here A is the island area, t is time, and t_0 is the time at which the island disappears.

We have used our Monte Carlo code to simulate decay of individual islands on terraces (the same theory will apply to pits.) Our goal was to compare the simulated decay, where we have control of the relative rates of different microscopic processes as well as step geometries, to predictions of our analytic model. At the heart of these analytic models of island decay is the Gibbs-Thomson effect which provides the driving force for the decay. Thus our first test of the analytic model was to investigate the validity of using the Gibbs-Thomson effect at small island sizes by simulating a vapor of adatoms in equilibrium with islands of varying radii [1,5,6]. At high temperatures and small island sizes (fig. 2), we find a deviation in the adatom pressure from the Gibbs-Thomson prediction. Using a theoretical analogy to the Ising model, we have shown that this deviation is due to the fact that the adatoms diffusing on the terrace do not behave as an ideal gas (e.g., dimers, trimers, etc. form on the terrace), whereas the Gibbs-Thomson formula was derived assuming the adatoms behave as an ideal, or noninteracting, gas.

We also found that reasonable agreement with simulation could be obtained with an expression that has the same functional form as the classical GT but with a phenomenological fitting parameter (we will refer to this as the “linear fit” (see fig. 2).

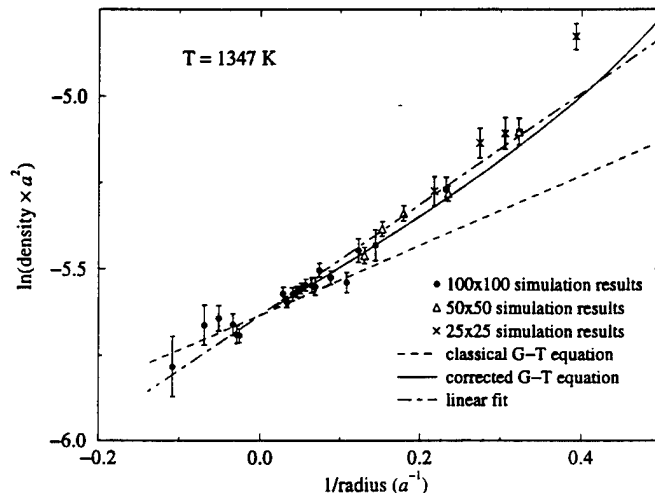


Fig. 2 Plot of the logarithm of the density of vapor outside an island vs. the reciprocal of its equilibrium radius. The dashed line represents the Gibbs-Thomson prediction assuming an ideal gas of vapor. The solid curve is the prediction using the corrected Gibbs-Thomson formula for the Ising model. The dot-dashed curve is the line of best fit assuming a functional form the same as for the Gibbs-Thomson relation but with a correction factor. Data points from three systems sizes of the simulation are shown. The data for negative values of the radius represent simulations for a pit in equilibrium with adatoms inside it.

Using this Monte Carlo code, we have simulated the decay of isolated islands on terraces (fig. 3). We have investigated the effects of using different boundary conditions (i.e., different geometries of step edges in the vicinity of the island), and different rates for microscopic processes, which we can vary

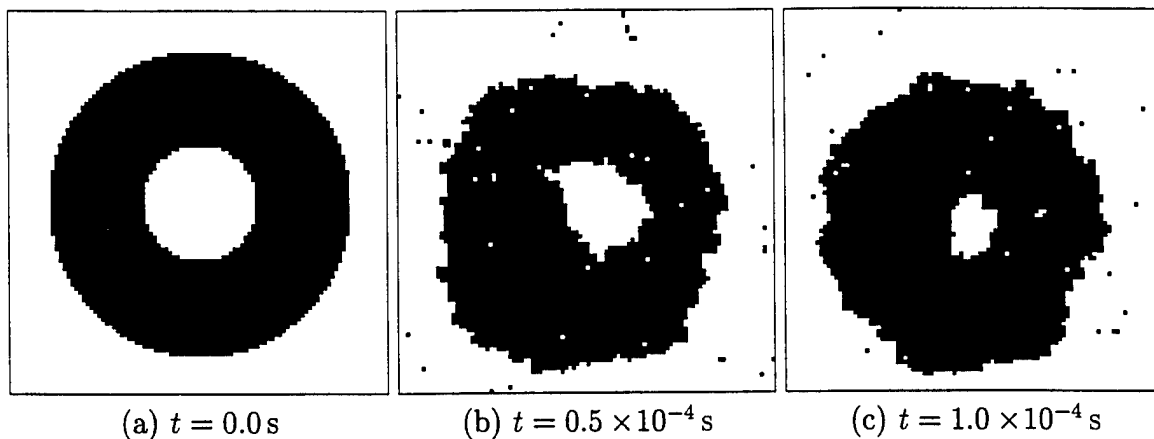


Fig. 3 Snapshots of a simulated island decay: the initial configuration of an island in a pit (a) and later stages at (b) 45% and (c) 90% of the total decay time. White regions are one atom higher than black regions. The simulation size is 100×100 lattice units with an initial circular island 15 lattice units in radius centered within a pit of radius 40 lattice units.

in the simulation. The simulations are compared to the predictions of our analytic model. Key results are summarized as follow: 1) using the simulation we can determine the macroscopic parameters in the analytic model that govern the decay; 2) using well-defined boundary conditions, the linear fit GT equation, and the macroscopic decay parameters determined from the simulation, good agreement between the analytic and simulated decay is obtained, indicating that the analytic model is correctly describing the decay down to small island sizes (fig. 4); and 3) the rate and functional form of the decay vs. time is sensitive to variations in the microscopic surface parameters and the boundary conditions.

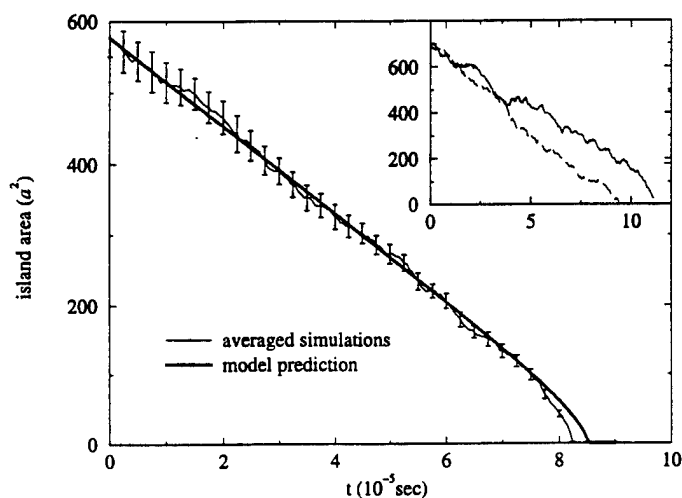


Fig. 4 Simulated island decay using barriers based on Cu(001), showing an island area versus time. The figure shows the average area of ten simulations referenced to time of island disappearance. Error bars represent standard deviations of the mean. The inset shows two representative individual decays. The heavy solid line is the analytic model using the corrected linear fit Gibbs-Thomson equation and decay parameters determined from the simulation.

Significance: These results are significant for two key reasons. First, they provide a connection between analytic and microscopic descriptions of surface processes that control stability and decay of nanoscale features. Second, a better understanding of stability and decay is important in any model describing growth or relaxation, or in understanding the stability of features manufactured on the nanoscale, such as metallic interconnects, quantum dots, etc.

New beamline for energetic beam deposition

Aaron Judy is a graduate student who received partial summer support from this grant. He is designing a new ultra high vacuum system for in-situ studies of energetic beam deposition. This project is now primarily supported by NSF and a new grant from AFOSR (Grant No. F49620-97-1-0020). A new beamline has been designed, in collaboration with Physicon Corporation, for studies of direct ion beam and ion beam-assisted deposition. The beamline is near completion with an anticipated delivery date prior to Spring 1997. The beamline will deliver ions ranging in energy from 5 eV to several hundred eV. Species include reactive ions (C^+ , N^+ , and O^+), noble gas ions, and semiconductor and metal ions. Judy has designed a system of connected ultra high vacuum chambers to interface with this beamline for in-situ surface analysis (e.g., Auger, LEED, RHEED, optical probes) and in-situ Scanning Tunneling Microscopy (STM) of the deposited films. The first experiments planned with this system are direct ion deposition of hyperthermal metal ions and ion-assisted deposition on metal substrates, using noble gas beams, to study mechanisms of ion-induced modification of growth.

Significance: In familiar growth techniques using plasmas and sputtering, hyperthermal ions are involved in the deposition process, but with uncontrolled distributions of parameters that make fundamental understanding difficult. Our goal is to use controlled beams that will make possible an

investigation of fundamental mechanisms of hyperthermal energy ion-induced modifications in growth.

Mechanisms of hyperthermal energy ion trapping at surfaces

Eric Dahl and Chad Sosolik are graduate students, both of whom received partial summer support from this grant. Eric Dahl (in collaboration with Visiting Professor David Goodstein) has used scattering techniques to measure trapping probabilities in 5 eV to 600 eV direct ion beam deposition of Na^+ on Cu(001) [8,9]. Na/Cu is a model system for which we have accurate scattering potentials. We find a strong dependence of the trapping (or deposition) probability on incident energy and angle. Using classical trajectory simulations we have developed a detailed understanding of the trapping mechanisms (involving both top layer and subsurface deposition) and energy transfer processes. These results are an important first step toward determining how the fundamental mechanisms of energetic beam deposition are influenced by beam parameters (such as energy and angle of incidence). Chad Sosolik is a new graduate student who is following up on this work.

Significance: Five eV to a few hundred eV ions are used routinely in thin film deposition and surface processing applications. These scattering measurements provide a fundamental understanding of the mechanisms of trapping (deposition) and energy transfer that play a key role in ion beam deposition and surface modification.

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