Section 4  Surfaces and Interfaces

Chapter 1  Synchrotron X-Ray Studies of Surface Disordering

Chapter 2  Semiconductor Surface Studies

Chapter 3  Step Structures and Epitaxy on Semiconductor Surfaces
Chapter 1. Synchrotron X-ray Studies of Surface Disordering

Academic and Research Staff
Professor Robert J. Birgeneau, Dr. Barry Wells

Graduate Students
Monte J. Ramstad, Michael J. Young

Technical and Support Staff
Debra L. Harring

1.1 Introduction

Sponsor
Joint Services Electronics Program
Contract DAAL03-92-C-0001
Grant DAAH04-95-1-0038

In this research program, we use modern x-ray scattering techniques to study structures and phase transitions in thin films and on surfaces. We have two principal experimental facilities, one at MIT and the other at the National Synchrotron Light Source at Brookhaven National Laboratory. At MIT, we have four high-resolution computer-controlled x-ray spectrometers. The angular resolution can be made as fine as 1.8 seconds of arc, which enables us to probe the development of order from distances of the order of the x-ray wavelength, 1 Å, up to 30,000 Å. The sample temperature can be varied between 2 K and 500 K with a relative accuracy of 2 x 10^-3 K. At the National Synchrotron Light Source, in collaboration with IBM, we have three fully instrumented beam lines. Two of these beam lines allow us to make studies with photons varying in energy between 3 and 12 keV; the third has a fixed energy of 17 keV. These facilities make possible high-resolution scattering experiments with a flux more than three orders of magnitude larger than that of a rotating anode x-ray generator. In collaboration with Professor Simon G.J. Mochrie, we have designed, built, and commissioned a second-generation x-ray surface facility which is extremely versatile, allowing a wide range of surface scattering experiments.

Our basic scientific objective is to understand the morphologies and microscopic structures of simple semiconductor and metal surfaces at high temperatures. Possible phase changes include surface roughening, surface reconstruction, melting, amorphization, and dilution.

Semiconductor surfaces exhibit rich structural and morphological behavior in both equilibrium and non-equilibrium but steady state conditions. Equilibrium structures and phase transitions of high-symmetry facets are often closely related to important models in two-dimensional statistical mechanics. The physics of stepped (vicinal) surfaces, even in equilibrium, is subtle and poorly understood. For example, stepped surfaces should always be rough because of the freedom of the steps to wander. Step structures and spacing should be profoundly affected by any nearby high-symmetry surface reconstruction. Their association bears on issues of faceting and equilibrium crystal shapes. The steps themselves may exhibit phase transitions that involve dislocations and/or bunching in the step lattice as well as step-height changes; changes in the morphology, especially those involving vacancy aggregates, should be strongly affected by nearby steps.

Semiconductor surface physics involves fundamental issues in statistical mechanics that are important to a broad spectrum of other problems. Furthermore, elucidating the factors that control the morphology and perfection of vicinal semiconductor surfaces can be expected to lead to improved electronic device processing, and ultimately improve semiconductor device performance and lifetime.

1.2 Stability of Vicinal Si(111) Surfaces Under Sublimation and Electromigration

Two types of morphological instabilities due either to a surface reconstruction or to a current effect during resistive heating have been observed on vicinal Si(111). The first is a longitudinal instability in the step spacing, which causes the surface to separate into bands of high step density and (111) facets of low step density. Step bands are formed during the 7x7 reconstruction as the vicinal surface is removed from the equilibrium crystal shape. Step bands are also formed during DC heating for one direction of current where the destabilizing current direction depends on temperature. The
second is a transverse instability in the step profile, which causes a rippling of the surface morphology. Surface rippling is observed for one direction of DC current independent of temperature. An exception to this occurs if step pairs of opposite inclination are generated on large (111) facets due to rapid sublimation. Then, since steps with opposite inclination form ripples for opposite current directions, the rippling instability may occur for both current directions. The concurrence of step bands and surface rippling is manifested by rippled step bands and flat (111) terraces. A common property of the current dependent instabilities is that the fluctuations are damped by reversing the current direction. Using a DC current to heat a sample resistively can cause the formation of step bands or can smooth the surface depending on the direction of the current. Likewise, the transverse fluctuations in the step profiles may be damped or smoothed by a DC current.

A combination of diffraction and imaging techniques has been used to study these morphological changes. X-ray diffraction studies have been carried out in the MIT UHV chamber at Brookhaven National Laboratory using the X20 x-ray beamline. Laser diffraction studies have been carried out in our new optical chamber at MIT. Optical microscopy and atomic force microscopy studies have been carried out using CMSE central facilities.

1.2.1 Non-equilibrium Behavior
Accompanying the 1x1-7x7 Transition

On a vicinal Si(111) surface miscut toward <112> with negative current heating, the 7x7 reconstruction causes a homogeneous step density to separate into step bands. During this transformation, the x-ray diffraction peak from the homogeneous step density disappeared over a 2°C temperature range. Not only does this provide strong evidence that the transition is first order, but it also indicates that the surface temperature is very uniform. A signature of a first order transition is the existence of a meta-stable phase. A meta-stable homogeneous phase was observed by comparing cooling and heating data through the transition. At the spinodal temperature, the meta-stability vanishes and spontaneous step bunching occurs. The spinodal temperature measured during cooling was about 4°C below the transition temperature measured during heating. Another signature of a first order transition is nucleation and coarsening. Characterizing these aspects requires studies of time dependence. Using x-rays, the average sizes of the (111) terraces and step bunches were measured as a function of time and temperature for surfaces quenched from the homogeneous phase to below the transition temperature. The sizes follow a power law in time, \( t^\eta \), where \( \eta \) increases from 0.1 to 0.5 as the temperature increases toward \( T_s \). Below the spinodal temperature, the steps remain relatively well-ordered and the order increases over time. Above the spinodal temperature, the steps become disordered as the (111) terraces undergo coarsening. To avoid the decomposition time dependence and access the equilibrium crystal shape, the transition was studied by heating through transition.

The 7x7 reconstruction driven phase separation was also studied for positive current. Reversing current direction from negative to positive at 900°C causes a homogeneous step density to undergo step bunching and step rippling. The step rippling occurs first. After a certain amount of rippling occurs, step bunching begins. The time required for step bunching to appear depends on the initial degree of order of the steps. Reversing the current direction from negative to positive below \( T_s \) causes the steps to become disordered and the step bunches to wander. This is evident in the broadening of the step and (111) terrace x-ray diffraction peaks in both of the surface plane directions. We conclude that the current drive step bunching and step rippling occur both above and below \( T_s \) for positive current. However, they occur much more slowly and on larger length scales than the reconstruction-driven step bunching. The behavior through the transition of a surface initially in a homogeneous state but heated with positive current is similar to that of a surface heated with negative current. However, as time progresses, the degree of large length scale step bunching and step rippling above \( T_s \) increases. Since the current driven step bunching and step rippling occur both above and below \( T_s \), it is difficult to access the equilibrium crystal shape with positive current.

1.2.2 High Temperature Current-Dependent Morphologies

The dynamics of the instabilities between 1000°C and 1300°C were studied using x-rays by reversing the current direction while maintaining a constant temperature. Below 1250°C, switching the current to positive caused step bands to unbundle and step rippling to occur. In the early stage, the steps unbundle and a homogeneous phase is formed. After obtaining a certain amount of order, the steps begin to ripple. Switching to negative current causes the rippled steps to transform to step bunches. First, an unripped homogeneous phase is formed, then step bunching occurs. The rate of these transformations increases with temperature.
However, the time dependence has not yet been fully analyzed.

Above 1250°C, reversing current direction causes transformation between a homogeneous phase at negative current and rippled step bunches for positive current. Initially, x-ray diffraction from the rippled step bunches shows a sharp (111) facet peak. The step diffraction peak, however, is too weak to measure due to the disorder of the steps. When the current direction is reversed, steps leave the step bunches creating a low step density on the (111) facets which gradually increases to the homogeneous phase density. The width of the diffraction peak from the low density regions broadens as the step density increases. After unbunching, which occurs rapidly, a slow step ordering process begins. The shape of the step peak is Lorenzian in the <110> direction and Gaussian in the <112> direction. The width is initially about three times broader in the <112> direction than in the <110> direction. The peak width sharpens in time according to a power law where \( \eta = 0.77 \) in the transverse direction and \( \eta = 0.62 \) in the longitudinal direction. Upon switching the current direction to positive, the step bunching occurs by the rapid growth of (111) facets with simultaneous rapid increase of the step density in the step bunches. This occurs so rapidly that it is difficult to observe in real-time.

Manifestly, the phenomenological behavior is very rich. We are continuing our empirical studies of these novel phenomena. We are also trying to develop basic theoretical models involving sublimation from steps and electromigration of surface Si atoms.

### 1.3 Publications


Professor John D. Joannopoulos (Photo by John F. Cook)