

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

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1.1 Introduction

Sponsor

Joint Services Electronics Program
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 2×10^{-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.

We are currently designing an undulator-based beam line which will be implemented at the Advanced Photon Source at Argonne National Laboratory. This facility will make possible a new generation of x-ray experiments with unprecedented precision. 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 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

The morphological instabilities of vicinal Si(111), caused by a DC electric current and a 7×7 reconstruction, have been studied using a variety of diffraction and imaging techniques. The current induced instabilities, which occur at sublimation temperatures or under conditions of step-flow, produce morphological features on length scales appropriate for optical studies, 5 to 50 μ . Optical microscopy, a laser diffraction technique atomic force microscopy (AFM) and high resolution x-ray

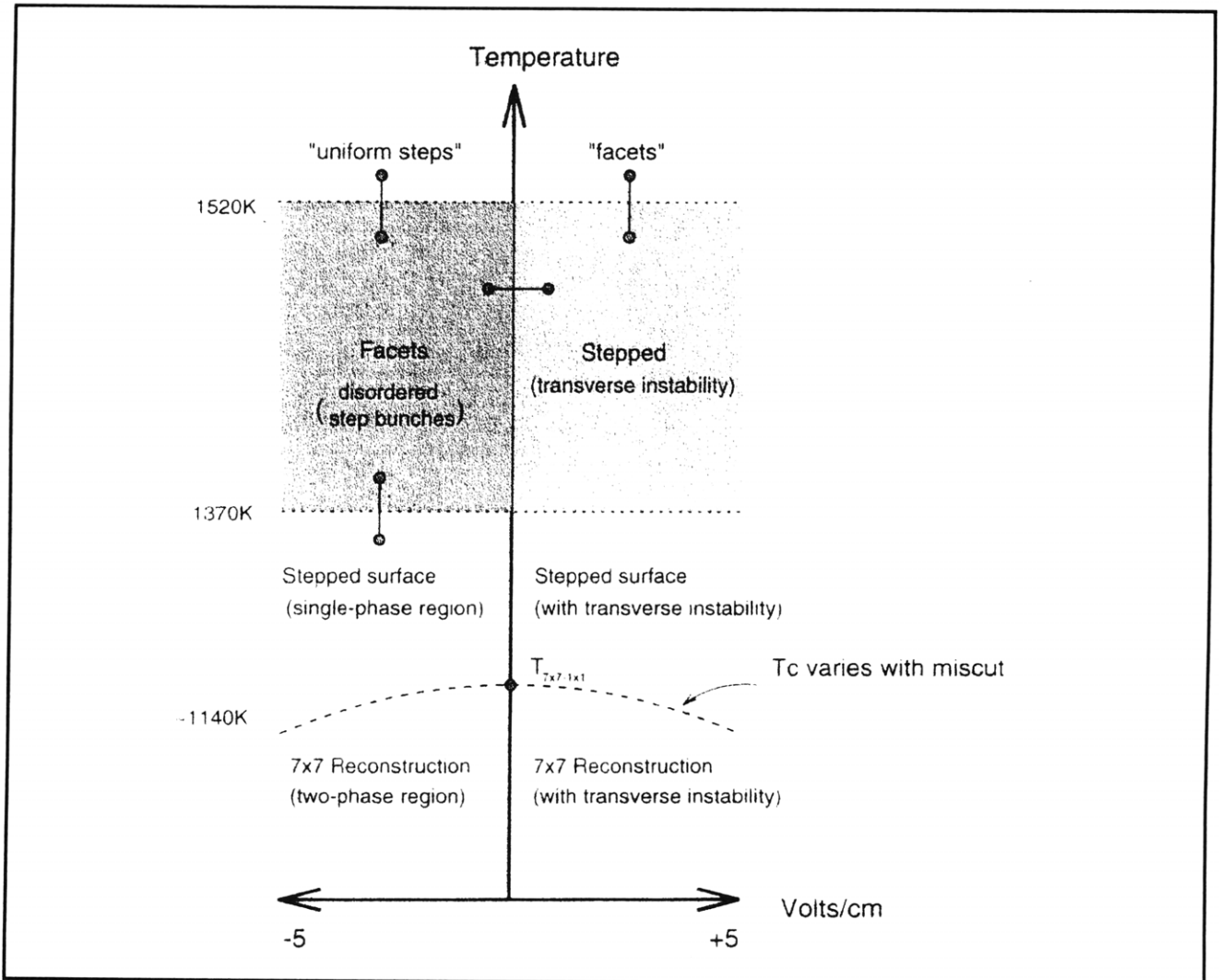


Figure 1. Stepped regions refer to single height (bilayer) steps. Bars indicate reversible transitions studied in experiment. At fixed current, the voltage is not a controllable parameter in our experiment.

diffraction have been applied to these surfaces. In addition to the step bunching instabilities reported by others, these techniques reveal a transverse instability in the step profiles for current flowing in the step-down direction at all temperatures between 700° C and 1300° C (see figure 1). For the same current direction, the step bunching instability is observed below $1025^{\circ} \pm 15^{\circ}$ C and above $1230^{\circ} \pm 15^{\circ}$ C up to at least 1300° C. On the other hand, for current flowing in the step-up direction, the step profiles are stabilized and the step bunching instability is observed between $1025^{\circ} \pm 15^{\circ}$ and $1230^{\circ} \pm 15^{\circ}$. Stoyanov's theory of the current driven step-bunching instabilities has been extended to explain the step wandering instabilities by modifying the boundary conditions of a diffusion equation at the step edges. Measurements of the rate of growth of the transverse instability indicate an activation energy of 2.7 ± 0.1 eV. Near the 7 x 7 reconstruction temperature,

$T_c = 870^{\circ}$ C, a faceting transition occurs. We have investigated the time and temperature dependence of the spinodal decomposition of step bands and the approach to the equilibrium crystal shape. The equilibrium shape profile exhibits critical behavior $z \sim x^{\alpha}$ where $\lambda = \alpha/(\alpha - 1) = 3.0 \pm 0.1$ which is consistent with a Pokrovsky-Talapov transition.

Studies of the step behavior on vicinal Si(111) have also been conducted on surfaces misoriented by 4 and 8° toward (1,1,-2). An investigation of the effect of current reversal on the dynamics of step bunching and debunching was completed at 1376 K, 1420 K, and 1470 K for the 8° sample, and at 1418 K for the 4° sample. The phase diagram (figure 1) shows the relevant temperature regions. The dashed line around 1140 K simply represents the orientational dependence of the reconstruction driven faceting transition. The applied voltage is

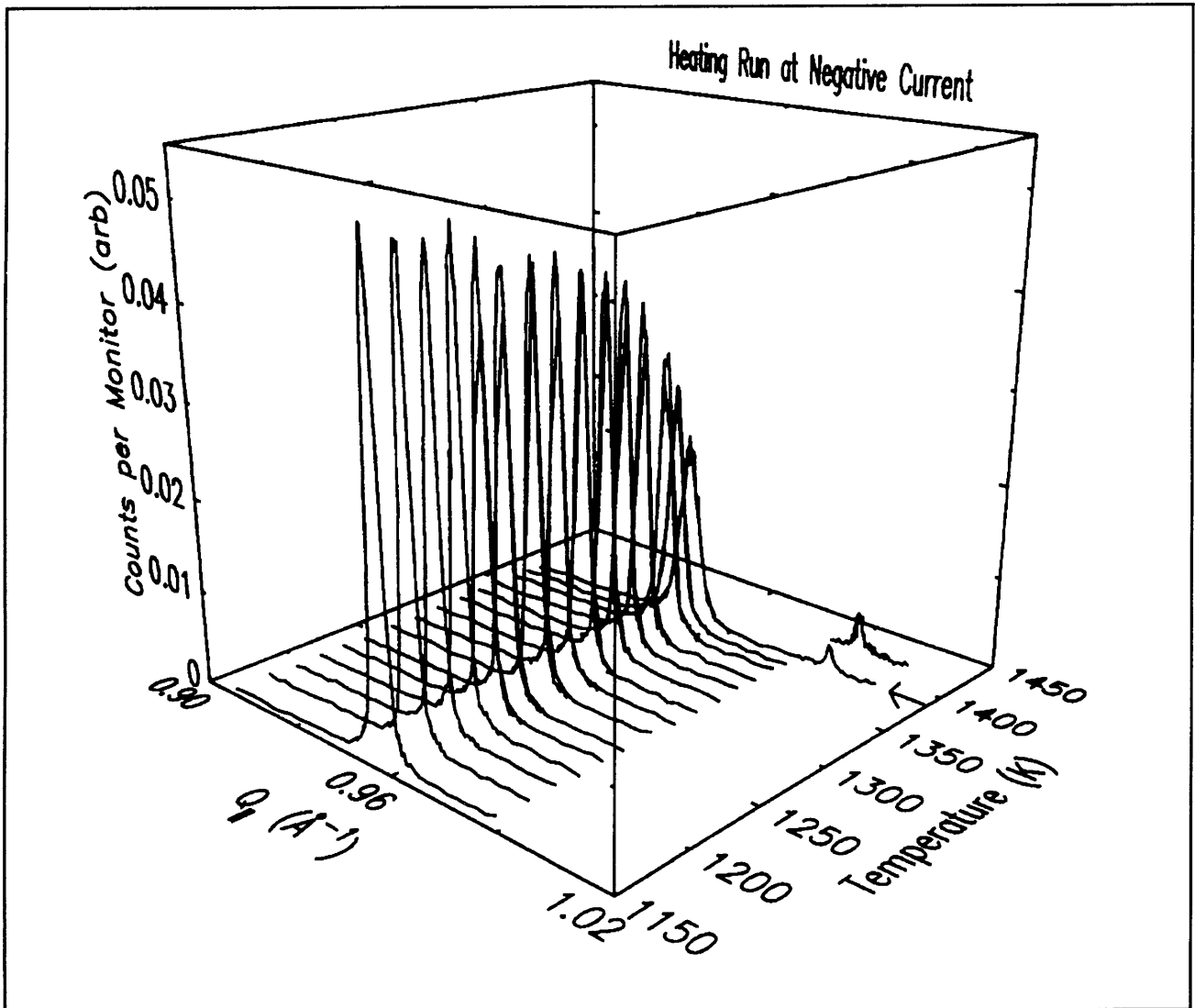


Figure 2. Temperature dependence of the step peak at negative current above the 7×7 reconstruction. Note coexistence of the (1,0) peak and the step peak at 1376 K (indicated by arrow).

not an independently controllable parameter in this phase diagram. Between 1370 K and about 1470 K, the time evolution was slow enough to permit independent measurements of the q -dependence of the scattering at the integer order facet peak (1,0) and at the step peak without significant distortion in the scattering profiles over periods ranging up to 200 minutes. The surface morphologies at these temperatures were essentially completely reversible, except after very long times at fixed current direction where either faceting or transverse step wandering could become saturated. Thus, a determination of the time dependence of the step correlation lengths, facet size, and integrated intensities was made for these temperatures over a substantial time range. The lower bound step-facet transition at ~ 1370 K was also studied on heating from the single phase stepped region at negative current

(see figure 2). In addition, a series of studies showing the time-dependence of the (1,0) peak were made on the 8° sample after quenching from above 1520 K (an unfaceted surface) to 1406 K and 1376 K at negative current showing a continual development of the morphology over periods of up to 150 minutes. In some cases facets as large as $0.7 \times 2.5 \mu^2$ were observed.

The scattering in the vicinity of the order-disorder transition at approximately 1520 K (see figure 1) was also examined for current both parallel and anti-parallel to (1,1,-2), i.e., positive and negative current, respectively. This transition temperature was found to be independent of miscut for both current directions to within the experimental error. True "step-ordering" above 1520 K at negative current was not observed at both current directions

for both the (1,0) peak and the step peak. Ex-situ optical microscopy revealed that large facets developed on samples quenched from above 1520 K at positive current and indicated a transverse instability consistent with that previously seen in other experiments on smaller miscut samples.

1.3 Monolayer Melting

We have investigated the melting transition of near-monolayer xenon at three temperatures: 121 K, 140 K and 145 K. Xenon on graphite is widely regarded as a model system for the investigation of two-dimensional melting in the presence of a weak orientational field. It may represent an example in which melting occurs continuously through the unbinding of topological defects in a way analogous to the Kosterlitz-Thouless transition in superfluids. Recent electron diffraction studies are consistent with a two-stage melting process in this system with an intermediate aligned liquid or hexatic phase between the solid and quasi-isotropic liquid phases. We have carried out isothermal high-resolution

synchrotron x-ray diffraction measurements of the transition from a finite-size limited 2D solid to a well correlated ($\sim 100 \text{ \AA}$) orientationally ordered 2D liquid. Our investigation confirms that at 140 K and 145 K there is continuous evolution of the length scale of the positional fluctuations up to approximately 1000 \AA . At 121 K, despite no discernable evolution of the correlation length, the transition is found to be no sharper in reduced units than at the higher temperatures. The exponent characterizing the decay of the order parameter corresponding to a finite-sized 2D crystal is found to be consistent with values reported for finite-sized two-dimensional magnets undergoing Kosterlitz-Thouless-like (2D-XY) continuous transitions.

1.4 Publication

Nuttall, W.J., D.Y. Noh, B.O. Wells, and R.J. Birgeneau. "Isothermal Melting of Near-monolayer Xenon on Single Crystal Graphite at 140K. *J. Phys. Condens. Matter* 7: 4337 (1995).