Chapter 2. X-Ray Diffuse Scattering

Sponsor

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Academic and Research Staff

Professor Robert J. Birgeneau, Dr. Joel Brock

Graduate Students

Kenneth Evans-Lutterodt, Hawoong Hong, Alan Mak, D.-Young Noh

In this research program, modern x-ray scattering techniques are used to study structures and phase transitions in thin films and on surfaces. We have two principal experimental facilities. At MIT, we have four high-resolution computer-controlled x-ray spectrometers with high intensity rotating anode x-ray generators. The angular resolution can be made as fine as 1.8 seconds of arc, enabling us to probe the development of order from distances of the order of the x-ray wavelength, ~ 1Å, up to 30,000 Å. The sample temperature may be varied between 2 K and 500 K with a relative accuracy of 2×10^{-3}K. We also maintain, in collaboration with IBM, a two-spectrometer system at the National Synchrotron Light Source at Brookhaven National Laboratory. A third beam line designed to operate at short wavelengths is in the final stages of construction at the laboratory. These systems make possible high resolution scattering experiments with a flux more than three orders of magnitude larger than that from a rotating anode x-ray generator. These experiments, in turn, have opened up the possibility for a new generation of experiments in this area.

As part of this JSEP program, we have built an x-ray compatible high-vacuum single-crystal apparatus. This apparatus enables us to use synchrotron radiation to study the structure and transitions occurring at a single surface. Our current experiments in this program are concentrated in two areas: 1) the phases and phase transitions of metal and semiconductor surfaces and surface overlayers; and 2) the structure and phase transitions of rare gas multilayers on simple substrates.

2.1 Metal Surface Studies

We have carried out detailed studies of the reconstruction of two prototype metallic surfaces: W (001) and Au (110). Both exhibit interesting new physical phenomena.

2.1.1 W (001)

The reconstruction of the clean W (001) surface at low temperatures has been widely studied since its discovery by low-energy electron diffraction (LEED). Below $\sim$ 200K a well defined $(\sqrt{2} \times \sqrt{2})$ R45° phase exists with a basic structure that is no longer seriously disputed, although certain details are still emerging (see figure 1). The nature of the high-temperature (1 x 1) phase has been investigated experimentally and described as (a) a disordered version of the low-temperature phase, (b) an ordered state, perhaps with a lower-symmetry (1 x 1) reconstruction, or (c) an incommensurate phase. Several groups have investigated the phase transition with LEED and He diffraction. Theoretical work on the phase transition has also been divided between order-order and order-disorder models.

The belief that there is a change of structure associated with this phase transition rests almost entirely on the LEED reports of a large reduction in intensity of the half-order diffraction beams above 200 K. In this

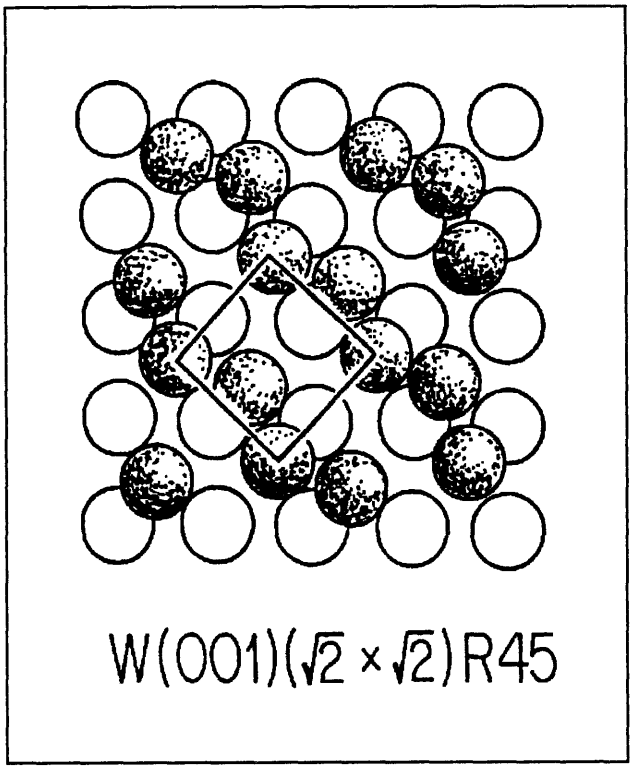


Figure 1. Sketch of the positions of tungsten atoms on the clean W(001) ($\sqrt{2} \times \sqrt{2}$)R45° surface.

research we have clarified this basic experimental fact by observing that 1) the kinematical x-ray diffraction intensity is nearly constant throughout the entire temperature region 130-360 K, and 2) above the transition temperature T_c, the peak merely changes shape to conserve its volume. Since there is no change in structure factor, the local structure is unaltered through the transition, and, therefore, the high temperature phase must be *disordered*. By measuring the peak widths as a function of T, we find that, as the ordered domains diminish in size, they retain an anisotropic 2:1 axial ratio. The peak widths scale approximately linearly with $T-T_c$, consistent with the classification of the phase transition as a 2D XY model with strong cubic anisotropy. Furthermore, we see no shift of the peak centroid from the true half-order position over 130-360 K, and, thus, no sign of incommensurability. This work, therefore, has elucidated and made quantitative all of the essential features of the W (001) surface reconstruction.

2.1.2 Au (110)

Many noble metal facets are known to favor reconstructed structures at low temperatures. Furthermore, at some temperature T_c, a reconstructed surface will typically undergo a reversible "deconstruction" to a high temperature structure which is no longer reconstructed. One such transition, which has been studied theoretically as well as with electron diffraction, is the Au (110) 1 x 2 to 1 x 1 deconstruction. While the "missing row" model of the reconstructed 1 x 2 surface is well established, the nature of the deconstruction transition itself remains controversial.

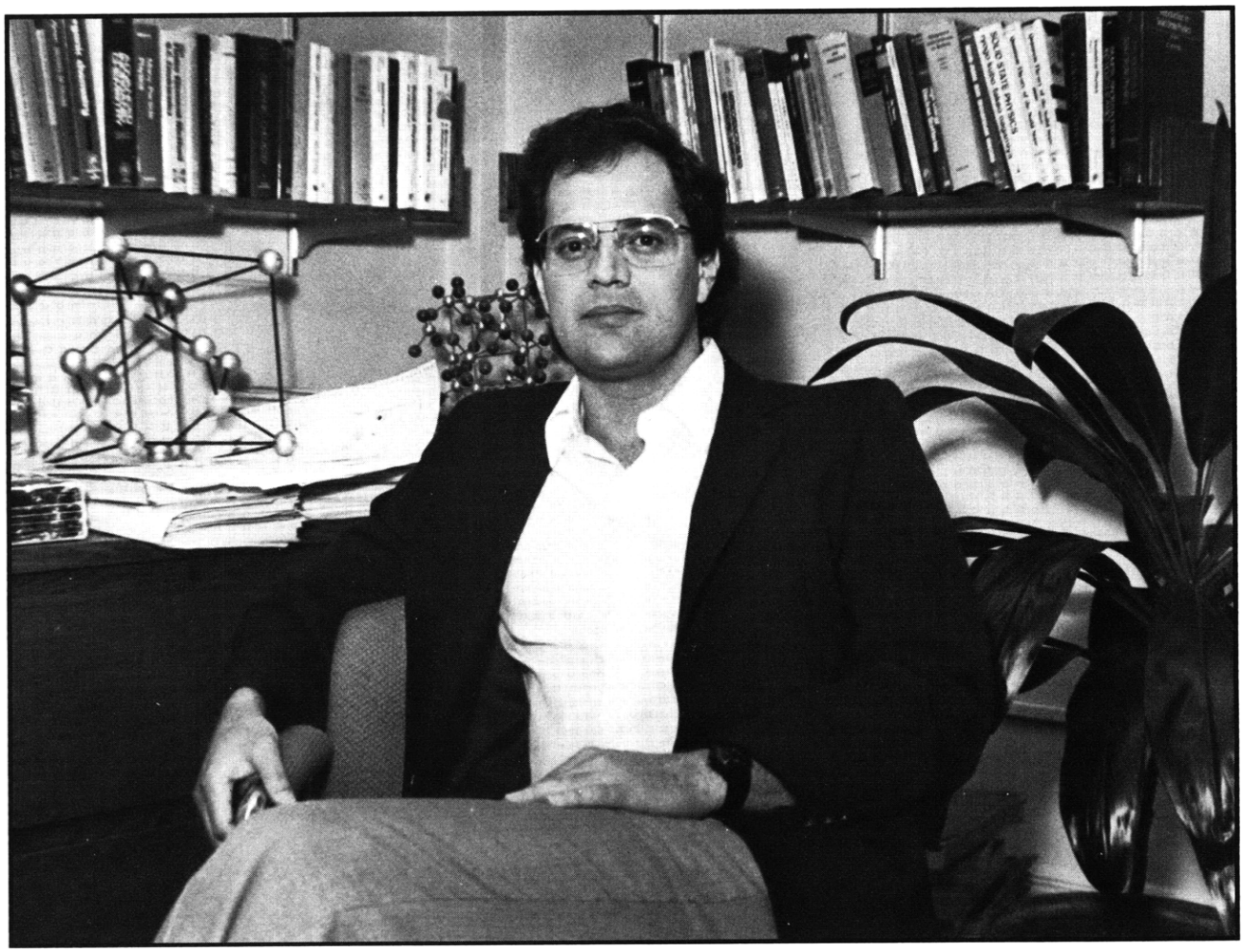
Previous work by our group has demonstrated that x-ray scattering is an effective means for studying surface phase transitions. Specifically, the lineshapes of bulk-forbidden surface peaks can provide information about the surface height-height (or step-step) correlation functions, and are thus a sensitive probe of the surface roughening transition. This transition is characterized by a proliferation of atomic steps which results in logarithmically divergent height fluctuations of the crystalline surface. We have carried out glancing angle synchrotron x-ray scattering experiments as a means of studying the Au (110) 1 x 3 to 1 x 1 deconstruction transition. Observing the temperature dependence of the superlattice and integral order bulk-forbidden (anti-Bragg) surface peaks, we find that at $T_c = 485°C$, the Au (110) 1 x 3 surface becomes incommensurate with the bulk crystalline lattice. During the commensurate-incommensurate transition, the surface both roughens and deconstructs. Because this picture is fundamentally different from that suggested by the current literature, further experimental and theoretical work on noble metal surface problems is required.

2.2 Rare Gases on Graphite

We have initiated a long-term research program to use rare gas multilayers on graphite as model systems for the study of equilibrium crystal growth and surface roughening. These experiments should complement our ongoing measurements of noble metal surfaces and overlayers. In the first generation of these experiments, we have

studied films of xenon on graphite with thicknesses of 1, 2, 3, 6, 24, and 44 layers. Our emphasis to date has been on the low temperature structures, the temperature dependence of the thicknesses, and the integrity of the layer stacking.

In our experiments, we have used x-ray scattering to monitor the thickness change accompanying the wetting of the xenon layers physisorbed onto the basal plane of single crystal graphite. At the same time, we have observed the in-plane structure of the layers, particularly of the first few layers. The motivation for this was the fact that the wetting behavior will probably depend on the structural adjustment of the first few layers. We have, in fact, succeeded in observing the in-plane structures of the xenon layers as well as out-of-plane structures in some detail. These observations reveal a variety of unexpected aspects in the structure of the physisorbed films. Previously it was reported that xenon wets a graphite surface completely at low temperatures. By contrast, we find that xenon exhibits incomplete wetting of the graphite at low temperatures. Additionally, the xenon films exhibit pronounced stacking disorder between the layers. The most unexpected result, however, is that the first xenon layer adjacent to the graphite is commensurate with the graphite surface structure at low temperatures, while the remaining layers are incommensurate with a structure close to that of bulk xenon.



Professor John D. Joannopoulos