Wednesday Morning, November 4, 1998

Surface Science Division
Room 314/315 - Session SS3-WeM

Surface Dynamics and Roughening

Moderator: T.E. Madey, Rutgers, The State University of New Jersey

8:20am **SS3-WeM1 The Atomic Steps of Oxygen Adsorption on Pt(110)-(1x2)**, *S. Helveg, S. Horch, I. Stensgaard, E. Laegsgaard,* ***F. Besenbacher,*** Aarhus University, Denmark

The dissociative adsorption of Oxygen on the Pt(110)-(1x2) surface is studied using a variable-temperature, fast Scanning Tunneling Microscope. The dynamic information extracted from the STM movies direct reveal how the O structures develop as function of O coverage. A variety of new and novel O structures are observed on the atomic scale. These initial structures develop at higher O coverage into [110] oriented stripes which self organize into highly regular and periodic bands. From an interplay between these STM results and theoretical GGA calculations a new and coherent picture evolves for O adsorption on Pt(110). This picture deviates significantly from previously proposed structural models but we can within the new picture explain all previous experimentally obtained results.

8:40am **SS3-WeM2 Etching of the Si(001) Surface with Molecular Oxygen**, ***J.B. Hannon,*** *M.C. Bartelt, N.C. Bartelt, G.L. Kellogg,* Sandia National Laboratories

We have investigated the growth and decay of two-dimensional pits, or vacancy islands, at the Si(001) surface using low-energy electron microscopy (LEEM). The vacancy islands nucleate near the centers of large (10 micron diameter) terraces when the surface is exposed to molecular oxygen at elevated substrate temperature. At sample temperatures above about 1000 K, oxygen does not accumulate on the surface, but desorbs in the form of SiO, leading to a net removal of Si from the surface. In our experiments, the vacancy islands grow monotonically during oxygen exposure. The growth rates of the individual vacancy islands depend on the distribution of neighboring islands and steps. We quantify this dependence using both Monte Carlo simulations and a diffusion equation analysis. The vacancy-island growth rates can be described using a simple model of the etching process: during oxygen exposure, Si atoms are removed from the surface at a temperature-dependent rate, leaving behind surface vacancies. The vacancies diffuse on the terrace until they encounter an island edge, where they are irreversibly incorporated. The sensitivity of the vacancy-island growth rates to the island configuration is a consequence of the fact that vacancy terrace diffusion is slow compared to vacancy incorporation at steps. By measuring the growth rates as a function of temperature and oxygen pressure, we are able to determine both the mechanisms by which oxygen etches the surface as well as the activation energy associated with the etch rate. We find that the temperature dependence of the etch rate, at constant oxygen pressure, obeys an Arrhenius form with an activation energy of about 2.0 eV. This work was performed at Sandia National Laboratories, a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of Energy under Contract DE-AC04-94AL85000

9:00am **SS3-WeM3 Surface Morphology Induced on Metal Surfaces by Ion Sputtering**, *S. Rusponi, G. Costantini, C. Boragno,* ***U. Valbusa,*** University of Genova, Italy

Ion sputtering can be used not only to clean or to analyse a surface, but also to construct regular patterns on amorphous, semiconductor and metals surfaces. We studied the formation of these patterns in different metal single crystals by using a variable temperature UHV STM. By accurately dosing the ion energy, flux and fluence and by selecting the appropriate crystal temperature, we can tune the final morphology, changing the shape of holes and islands, their density and thickness. Different examples will be reported: on Ag(110)@footnote 1@ and Cu(110)@footnote 2@ a ripple structure can be induced, with an orientation respect to the surface directions depending mainly on the temperature, while on Ag(001)@footnote 3@ a checkerboard morphology with a square simmetry has been found. It has been found that many properties of the sputtered surfaces follow simple scaling laws. A general model, based on the competitive role of surface diffusion and erosion can well explain the reported results. @FootnoteText@ @footnote 1@S. Rusponi et al., Phys. Rev. Lett. 78 (1997) 2795 @footnote 2@S. Rusponi et al., Phys. Rev. Lett., submitted @footnote 3@S. Rusponi et al., Surf. Sci., submitted

9:20am **SS3-WeM4 Direct Observation of Etching Mechanisms of Cu Surfaces with STM**, *C.Y. Nakakura,* ***E.I. Altman,*** Yale University

Scanning tunneling microscopy movies of the halogenation and subsequent etching of Cu(100) and Cu(11 1 0) were recorded. Adsorption of both Br@sub 2@ and Cl@sub 2@ resulted in a c(2x2) structure on the Cu(100) terraces. As the adsorbate coverage approached saturation, the substrate steps faceted to align along the close-packed directions of the adlayer. The faceted steps were the Cu atom source for subsequent halide formation and etching. For Cl@sub 2@, the reaction was initiated at facet corners and proceeded by stripping away rows of atoms parallel to the steps. In contrast, Br@sub 2@ could also react perpendicular to the step edge creating channels etched into the terraces. For both Cl@sub 2@ and Br@sub 2@, the halide formed by reaction diffused across the surface, nucleating and growing halide clusters independently of the reaction step. At room temperature, three-fold symmetric CuBr(111) islands were observed that grew by addition of CuBr units to island edges. Remarkably, the presence of these islands did not block further reaction of the underlying Cu surface. The CuBr islands formed at room temperature roughened either with time or annealing. At temperatures above 440 K, the halide formed by reaction sublimes resulting in etching. Under these conditions, a new CuBr morphology was observed: four-fold symmetric islands. These square islands nucleated at step edges and then sublimed at rates far in excess of those of similar sized three-dimensional clusters, suggesting that controlling the morphology of the CuBr can lead to reductions in etching temperatures.

9:40am **SS3-WeM5 How Predictable Is Surface Morphology?**, ***N.C. Bartelt,*** Sandia National Laboratories **INVITED**

One way people are beginning to probe the mechanisms of surface dynamics is to compare the histories of particular surface configurations obtained using microscopy with predictions from simulations. An advantage of this technique over the usual method of studying the time dependence of quantities averaged over the surface is that it allows the direct determination of how individual surface defects (such as steps) move in response to their local environment. A fundamental question about this approach is how well, and for how long, can one expect to account for the evolution of the complicated surface configurations which are typically observed in experiments. At small enough length scales, unpredictable thermal fluctuations begin to play a role. In this talk I discuss the basic theory and implications of the breakdown of deterministic equations of motion of surface morphology caused by these fluctuations. I will illustrate the issues involved. for several systems: First, given a configuration of surface steps, I will discuss how well one can predict where, and in what sequence, islands nucleate during epitaxial growth. I will compare theory with experiments of vacancy island nucleation during O etching of Si(001) which show that nucleation gets less predictable as the temperature (and critical nucleus size) gets smaller. Second, I will discuss how long one can in principle expect to account for the structure of adsorbed layers during domain coarsening and for island configurations during ripening. I will give theoretical and experimental examples of how sensitive histories of such complicated structures are to chance events and the knowledge of initial conditions.

10:20am **SS3-WeM7 Restructuring of Non-Square Vacancy and Adatom Clusters and of Indentations and Protrusions at Step Edges for Ag/Ag(100)**, ***C.R. Stoldt,*** *A.M. Cadilhe, C.J. Jenks, J.W. Evans, P.A. Thiel,* Iowa State University

During metal(100) homoepitaxy, near-square 2D islands (which are nucleated on broad terraces) first grow and merge, forming rectangular and dumbbell shaped clusters. Approaching a coverage of one monolayer, vacancy regions remain in the first layer which display a variety of unusual compact and worm-like shapes. Also, any extended step edges advance during deposition, incorporating islands in the process, and acquiring an irregular structure with many protrusions and indentations. The rearrangement of these non-equilibrium structures, if sufficiently rapid, can significantly influence multilayer growth. Thus, we present room-temperature STM studies for Ag/Ag(100) adlayers which provide a comprehensive characterization of the restructuring dynamics: (i) for adatom and vacancy clusters with rectangular and dumbbell shapes, and for worm-like vacancy regions; and (ii) for square, triangular, and other shapes of protrusions and indentations at extended step edges. In particular, we assess the dependence of restructuring rates on feature size and shape (or on local step edge orientation), and compare behavior for "mirror-image" adatom and vacancy structures. Behavior is elucidated by

comparison with theoretical studies of "perimeter diffusion" models for step edge evolution.

10:40am **SS3-WeM8 Evolution and Structure of the Stripe Phase Reconstruction of Cu/Ru(0001)**, ***H. Zajonz***, *D. Gibbs,* Brookhaven National Laboratory*; A.P. Baddorf, D.M. Zehner,* Oak Ridge National Laboratory

X-ray scattering studies of the structure of Cu layers deposited on Ru(0001) substrates during growth and versus substrate temperature have been initiated. The nearest-neighbor spacing in bulk fcc Cu is 5.8% smaller than that in hcp Ru. Earlier STM studies,@footnote 1@ established that the first layer of Cu on Ru(0001) adopts a pseudomorphic structure, but that the two-layer film exhibits a stripe-phase reconstruction which consists of an uniaxial modulation of Cu chains along [100]. Measured x-ray intensities provide evidence that the first layer does not remain pseudomorphic in the stripe phase. Our experiments also involve characterizing the appearance and evolution of the stripe-phase reconstruction peak as a function of Cu coverage and substrate temperature. At 720 K, x-ray scattering confirms a pseudomorphic Cu structure for coverages up to about one monolayer, followed by the growth of a stripe-phase reconstruction between one and two layers. In the stripe phase, the average Cu spacing along the rows is that of Cu bulk, however between rows the spacing is equivalent to that of Ru. At two monolayers coverage, there is an abrupt contraction of the average Cu spacing by an additional ~0.5% which can be attributed to an increase in the amplitude of the transverse modulations with layer completion. This phenomenon has not been observed previously for an interfacial reconstruction. ORNL is managed by Lockheed Martin Energy Research Corp. under U.S. Department of Energy contract DE-AC05-96OR22464. Work at BNL is supported by DOE, under contract DE-AC02-98CH10886. @FootnoteText@ @footnote 1@ C. Günther et al., Phys. Rev. Lett. 74, 754 (1995)

11:00am **SS3-WeM9 Dynamics of the Striped Nanostructure of the Oxidized Cu(110) Surface: A Momentum-Resolved ESDIAD Study**, ***D. Mocuta***, *J.W. Ahner, J.-G. Lee, S. Denev, J.T. Yates, Jr.,* University of Pittsburgh

The striped structure of the partially oxidized Cu(110) surface has been studied using a novel technique, Momentum-Resolved ESDIAD. Long ...O-Cu-O... strings oriented in the direction and exhibiting attractive interactions with each other form periodically arranged stripes with widths in the nanometer range.@footnote 1,2@ Two different oxygen sites were detected, leading to a two-fold symmetrical four beam O@super +@ ESDIAD pattern with tilting of the beams of 22° in the azimuth (A) and 8° in the azimuth (B) directions. The relative intensities of the four beams have been compared for a wide range of oxygen coverages leading to a model in which the A beams correspond to O@super +@ ions desorbing from the edges of the stripes and the B beams from the internal regions of the stripes. This model is confirmed by studying the effect of the coadsorption of Ar at 32 K on the oxidized structure. The dynamical motion of the one-dimensional ...O-Cu-O... oscillator chains situated at the edges of the stripes has been studied by Momentum-Resolved ESDIAD measurements over a broad range of temperatures (70 K- 650 K). In addition, this striped surface has been used as a template for the adsorption of other species, whose interactions with the ...O-Cu-O... chains are revealed. @FootnoteText@ @footnote 1@D. J. Coulman, J. Wintterlin, R. J. Behm, G. Ertl, Phys. Rev. Lett. 64 (1990) 1761. @footnote 2@K. Kern, H. Niehus, A. Schatz, P. Zeppenfeld, J. George, G. Comsa, Phys. Rev. Lett. 67 (1991) 855.

11:20am **SS3-WeM10 Evidence and a Model for a Chemically Mediated Roughness Transition**, ***E.A. Irene***, *C. Zhao, Q. Liu,* University of North Carolina, Chapel Hill

In our previous research we have followed the changes effected by oxidation on purposely roughened and initially smooth Si surfaces via atomic force microscopy (AFM) and spectroscopic ellipsometry (SE) and a new technique called spectroscopic immersion ellipsometry (SIE).@footnote 1,2@ For initially rough surfaces both thermal and plasma oxidation yield smoother surfaces and this can be understood from the changes in the local free energy at asperities, viz. the Kelvin equation. What was surprising was that initially smooth Si surfaces roughen upon oxidation. These results are not contradictory, since the roughness magnitudes do not cross, but these results strongly suggest a limiting roughness. Furthermore, a chemically mediated roughening transition explains the observations although this mechanism is not yet proven. The experimental and modeling results will be presented. @FootnoteText@ @footnote 1@Q. Liu, L. Spanos, C. Zhao and E.A. Irene, J. Vac. Sci. Technol. A, 13, 1977 (1995). @footnote 2@C. Zhao, Y.Z. Hu, T. Labayen, L. Lai and E.A. Irene, J. Vac. Sci. Technol. A, 16, 57, (1998).

11:40am **SS3-WeM11 Reconstruction of Bimetallic Systems: Ultrathin Films of Rh, Pt and Pd on W(211)**, ***K. Pelhos***, *I.M. Abdelrehim, C.H. Nien, T.E. Madey,* Rutgers University

The (211) face of tungsten is found to undergo an (nx1) reconstruction when covered with a thin film of Rh, Pt or Pd, followed by annealing. A combination of low energy electron diffraction, Auger electron spectroscopy (AES) and temperature programmed desorption (TPD) experiments indicate that the reconstruction occurs within a very narrow coverage range, between 0.5 and 1 physical monolayer, and only when the annealing temperature exceeds a threshold value of ~900 K (Rh and Pt) or ~500 K (Pd) (1 physical monolayer corresponds to 1.63 x 10@super 15@ atoms/cm@super 2@, sufficient to cover completely all of the exposed W atoms in the W(211) rows and troughs). The reconstruction is almost exclusively (3x1) for Rh and Pt, and (~12x1) for Pd. Scanning tunneling microscopy (STM) investigations of the Rh/W(211) system reveal that the atomic structure of the (3x1) reconstruction is due to very long (up to 1000 Angstroms) double rows of Rh separated by missing rows; the troughs of the W(211) surface presumably are filled with Rh atoms. Similarly, the atomic structure of the Pd/W(211) (~12x1) reconstruction is due to the W(211) surface being covered by the Pd overlayer, except for equally spaced missing rows of overlayer atoms. Furthermore, evidence for two phase coexistence and domain growth (distinct (1x1) and (3x1) regions) is found in STM of Rh/W(211) near one physical monolayer coverage.TPD experiments performed at very high temperatures (up to 2400 K) provide insight into the adsorption energetics of these metallic overlayers. Our results are discussed in the context of previous experiments: the faceting of W(111) surface into nanoscale three-sided pyramids with (211) facets, induced by metal overlayers.

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