Thursday Afternoon, November 7, 2002

Surface Science
Room: C-112 - Session SS+EL-ThA

Growth & Etching on Semiconductor Surfaces

Moderator: A.C. Kummel, University of California, San Diego

2:00pm **SS+EL-ThA1 Epitaxial Growth Dynamics of Semiconductor Quantum Dot Structures**, ***S.R. Leone***, University of California and Lawrence Berkeley National Laboratory **INVITED**

The formation of Ge nanodots on Si(100) occurs by strain-induced mechanisms (Ge is 4% larger than Si) and obeys the Stranski-Krastanov (SK) growth mode: a wetting layer (3-5 layers) is followed by the formation of three-dimensional (3D) Ge structures. Quantitative studies of Ge island size distributions and their shape transformations, including huts, pyramids, domes, and superdomes, and shape changes due to annealing of the islands under the influence of surfactants, such as arsenic, are studied by molecular beam epitaxial growth and atomic force microscopy (AFM) post-analysis. For device applications, it is important to attain control over the size and spatial distributions of self-assembled nanostructures. The Ge growth experiments are also carried out on patterned silicon substrates (mesas formed by electron beam lithography followed by etching), for specific positioning of the dots. A 'one island on one mesa' relationship is achieved. The density of islands is higher than can normally be produced on unpatterned silicon, where island coalescence would usually occur well before this density is possible. Preferential growth on the tops of the mesas most likely occurs because the Si mesa tops are deformable, fulfilling a strain relaxation condition. In this work, pyramid-type islands as small as 25 nm are also aligned on the mesa tops, and no limit to the size reduction of the islands is apparent, being controlled mainly by the size of the etched features that can be introduced.

2:40pm **SS+EL-ThA3 Si Deposition on H-terminated Si(100) Surfaces***, ***J.-Y. Ji***, *T.T. Barus, T.-C. Shen*, Utah State University, *G. Qian*, University of Illinois at Urbana-Champaign, *X. Luo*, National Renewable Energy Laboratory, *S. Ren*, Illinois State University, *S. Zhang*, National Renewable Energy Laboratory, *Y.C. Chang*, University of Illinois at Urbana-Champaign

The presence of H has long been considered adverse to Si homoepitaxy. Copel and Tromp[1] reported that while no apparent effect on epitaxy was observed at H coverage <1 ML and H segregates at growth temperature >400 K, a drastic epitaxial temperature increase was required at H coverage >1 ML. We will present the results of our STM, and first principle molecular dynamics studies on Si monohydride and dihydride effect in Si epitaxy. We confirmed that at growth temperatures ~ 500 K, H stays on the growth front on monohydride surfaces and epitaxy can be achieved but the domain sizes are much smaller than those grown on the bare Si. Continuous rebonding apparently is responsible for the epitaxial growth on Si monohydride surfaces. The diffusion barrier for the Si adatom along the Si monohydride dimer rows is calculated to be 1.1 eV which is significantly higher than the corresponding 0.6 eV barrier on the bare Si(100)-2x1 surface. To account for the H segregation, a mechanism to exchange H atoms between a surface Si atom and the incident Si atom is proposed. The experimental and theoretical result on the dihydride effect will be discussed.

*This work is supported by NSF-DMR9875129, ARDA/ARO DAAD 19-00-1-0407 and DARPA-QuIST DAAD 19-01-1-0324.

[1]M. Copel and R. M. Tromp, Phys. Rev. Lett. 72, 1236 (1994).

3:00pm **SS+EL-ThA4 An Atom-Resolved Study of Vacancy Dynamics and Surface Roughening on Bromine-Etched Si(100) Surfaces**, ***C.F. Herrmann****, *J.J. Boland*, University of North Carolina, Chapel Hill

Halogen etching of Si(100) surfaces has long been considered to involve the selective removal of atoms from an essentially static surface. However, our high temperature scanning tunneling microscopy (STM) study reveals that halogen-covered surfaces are highly unstable. This instability stems from the inherent steric repulsions between halogen adatoms on the surface. At high temperatures, repulsive interactions are relieved by vacancy formation, diffusion and surface roughening, each of which is directly observed in real-time by STM. Together, these dynamical processes result in surface features identical to those found after high temperature etching. These results demonstrate that diffusion and roughening must be considered in any model of halogen etching. Moreover, steric repulsions and the instability they create place fundamental limits on the ability to achieve atomically smooth morphologies using halogen etching.

3:20pm **SS+EL-ThA5 Dynamics of Si(100)-(2x1) Surface Modification with Cl**, ***G. Xu***, *E. Graugnard, V. Petrova, K.S. Nakayama, J.H. Weaver*, University of Illinois at Urbana-Champaign

The etching dynamics of Cl-Si(100)-(2x1) at elevated temperature have been studied with variable temperature scanning tunneling microscopy. Clean samples were exposed to Cl_2 at room temperature to near saturation and then heated to 700 K for over 20 hours. By scanning the same area of the sample, we observed pit creation, diffusion, incorporation and annihilation. Pit annihilation has not been reported previously under conditions of steady state etching and surface saturation. We also observed regrowth islands creation, growth and decay. Surface reactions at 700 K produced single Si adatoms, which were bonded to Cl-free Si dimers. Single Si adatoms diffused through these Cl-free Si dimer sites, but adatom diffusion was restricted by the high Cl concentration. The adatoms could form regrowth dimers when they met or they could be accommodated at the ends of regrowth structures. The adatoms could also be released from the regrowth dimer rows with the assistance of bare dimers. We have also observed (3x2) and (5x2) surface structures and, for the first time, the phase transition between (3x2) and (5x2) structures and dimer vacancy lines.

3:40pm **SS+EL-ThA6 Surface Modification without Desorption: Recycling of Cl on Si(100)-(2x1)**, ***K.S. Nakayama***, *E. Graugnard, J.H. Weaver*, University of Illinois at Urbana-Champaign

We demonstrate the structural consequences of thermally activated reactions of Cl on Si(100)-(2x1). We used scanning tunneling microscopy at room temperature to obtain atomic-resolution images of the surface before and after thermal processing. We show surface modification under conditions where Cl is recycled rather than desorbed as $SiCl_2$. In this unexpected reaction, the surface roughens as dimer vacancies are produced. First, a dimer with 2 Cl atoms, 2SiCl, converts to $SiCl_2$ + Si. This allows the destabilized, bare Si atom to escape onto the terrace. At temperatures where the desorption is negligible, the $SiCl_2$ unit decays as the Cl atoms can move to other active sites of the Si surface, allowing the second Si atom to escape. The result is a dimer vacancy, Si atoms on the terrace that can form self-organized regrowth structures, and Cl that is able to participate in another pitting event. Access to this unexpected roughening pathway is controlled by the Cl concentration and temperature. This previously overlooked process represents an important component of Si(100) surface processing.

4:00pm **SS+EL-ThA7 Spontaneous Roughening -- Fundamental Limits in Si(100) Halogen Etch Processing**, ***D. Chen***, *J.J. Boland*, University of North Carolina at Chapel Hill

A dynamical Scanning Tunneling Microscopy and Density Functional Theory study of the thermodynamic stability of halogen-terminated Si(100) surfaces is presented. Defects-free halogen-covered Si(100) surfaces are shown to be intrinsically unstable and prone to spontaneous roughening. This instability is the result of steric effects and is observed for all halogens except fluorine (which is already known to yield rough surfaces). These results demonstrate that an atomically smooth Si(100) morphology cannot be realized using halogen etch processing which sets a lower bound on the atomic scale perfection that can be achieved using such processing.

4:20pm **SS+EL-ThA8 Preparation of Atomically Flat Si(100) Surface by Ion Etching**, ***J. Kim***, *J.-Y. Ji, T.-C. Shen*, Utah State University, *J.S. Kline, J.R. Tucker*, University of Illinois

Preparation of atomically flat Si(100) surface under limited thermal budget has been one of the challenging processes in the research of atom-scale electronic device fabrication. While atomically flat monohydride surfaces can be obtained by aqueous NH_4F etching on Si(111) surfaces,[1] wet-chemical process has yet to produce an atomically flat Si(100) hydride surface at a scale of more than a few nanometers. Ion irradiation effects on Si surfaces have been investigated extensively from mid-70s to early 90s as a surface cleaning process prior to Si epitaxy. Recent atomistic studies of ion sputtering on pristine Si and metal surfaces were more focused on the dynamics of ion-induced defects.[2] We are currently using STM to investigate the surface morphologies during ion etching of oxide or wet-chemically prepared Si(100) surfaces by 0.4 - 1.5 keV Ar and Xe ions. We will delineate the effects of ion energy, ion fluence and substrate temperature, and assess the possibility of achieving atomically flat Si(100) surfaces by optimizing these parameters. This work is supported by NSF-DMR9875129, ARDA/ARO DAAD 19-00-1-0407 and DARPA-QuIST DAAD 19-01-1-0324.

[1] G. S. Higashi et al., Appl. Phys. Lett. 56, 656 (1990).

***** Morton S. Traum Award Finalist

[2] P. Bedrossian, Surf. Sci. 301, 223 (1994) and references therein.

4:40pm **SS+EL-ThA9 Probing the Chemistry of Impurities with STM: The Profound Effect of Dissolved Oxygen on Silicon Etching**, ***S.P. Garcia***, *H. Bao, M.A. Hines*, Cornell University

A new technique has been developed to quantify the surface reactivity of impurities, which combines the exquisite defect sensitivity of scanning tunneling microscopy (STM) and the analytical capabilities of atomistic kinetic Monte Carlo (KMC) simulations. This technique exploits kinetic competition between the impurity and a reference etchant to produce impurity-concentration-dependent changes in nanoscale etch morphology. These changes are then quantified using STM measurements and KMC simulations. We used this technique to measure the site-specific reactivity of dissolved O_2 -- a ubiquitous impurity in aqueous solutions -- with H-terminated Si(111) surfaces. The site-specific reactivity of O_2(aq) is surprisingly anisotropic. Oxidation of the highly strained dihydride step site is four times faster than oxidation of the relatively unstrained monohydride step site. Both steps are 10^4 times more reactive than terrace sites. FTIR measurements of the Si-H stretch vibration showed that dissolved O_2 inserts O atoms into surface Si-Si backbonds without removing the H-termination. Dissolved O_2 does not attack Si-H bonds, since neither Si-H consumption nor silanol production is observed.

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