Thursday Morning, October 28, 1999

Surface Science Division
Room 607 - Session SS2+EM+NS-ThM

Nucleation and Growth

Moderator: G.M. Nathanson, University of Wisconsin, Madison

8:20am **SS2+EM+NS-ThM1 Evolving Surface Morphology: An In Situ STM Study of 2-20 nm SiGe Quantum Wells Grown on 75 mm Si (100) Wafers**, ***G.G. Jernigan,*** Naval Research Laboratory, US*; P.E. Thompson,* Naval Research Laboratory

Electrical device improvement will come from the understanding and control of interfaces at the atomic level. We have integrated an STM with a commercial Si MBE system for in situ examination of device structures grown on full 75 mm wafers. Our first system of study is the growth of SiGe quantum wells on Si (100). SiGe is being investigated for its use in optoelectronics and high speed circuits. We will present a description of the atomic surface morphology of the initial 100 nm epitaxial Si buffer layer, followed by a Si@sub 0.8@Ge@sub 0.2@ quantum well of thickness up to 20 nm, and a Si capping layer deposited after the quantum well. The Si buffer layer is grown using a 0.08 nm/s Si flux onto a 650 @super o@C substrate. Step-flow growth occurs, but the different adatom sticking probabilities at the S@sub A@ and S@sub B@ steps produces a "wavy" surface which is the result of alternating terraces growing rapidly in different directions rotated by 90@super o@. The deposition of Si@sub 0.8@Ge@sub 0.2@, by co-depositing Ge with a 0.02 nm/s flux and Si, disrupts the "wavy" growth morphology. Ge segregation during deposition produces a "2xn" surface reconstruction that consists of rows which run perpendicular (G@sub A@) and parallel (G@sub B@) to a step edge in a manner analogous to the S@sub A@ and S@sub B@ steps, respectively. The quantum well morphology consists of a mosaic of small terraces containing short rows of G@sub A@ steps and long rows of G@sub B@ steps. The Si capping layer grown on the quantum well reinstates the "wavy" morphology. However, in addition to the S@sub A@ and S@sub B@ step-flow growth, dimer vacancy lines are now observed within the S@sub A@ terraces and are due to Ge segregation from the quantum well. The effects of S@sub A@/S@sub B@ waves, Ge "2xn" terraces, and dimer vacancy lines as a function of growth rate and temperature will be discussed. This work was supported by the ONR.

8:40am **SS2+EM+NS-ThM2 Hydrogen-Mediated Surface Morphological Evolution in Si@sub 0.7@Ge@sub 0.3@/Si(001) Layers Grown by Hydride Gas-source Molecular Beam Epitaxy**, ***T. Spila,*** *P. Desjardins, H. Kim, N. Taylor, D.G. Cahill, J.E. Greene,* University of Illinois, Urbana*; S. Guillon, R.A. Masut,* Ecole Polytechnique de Montréal, Canada

The primary mechanisms for relieving misfit strain @epsilon@ during heteroepitaxy are the formation of misfit dislocations (MD) and strain-induced roughening. These mechanisms are initially competing (due to @epsilon@-dependent activation energies) and eventually interacting once relaxation is initiated. Si@sub 0.7@Ge@sub0.3@ layers were grown on Si(001) (miscuts @<=@ 0.1°) to quantitatively investigate the effects of Ge surface segregation and steady-state hydrogen coverage @theta@@sub H@ on mechanisms of surface morphological evolution during GS-MBE from Si@sub 2@H@sub 6@/Ge@sub 2@H@sub 6@. For growth temperatures T@sub s@ > 600 °C where @theta@@sub H@ = 0, layers exhibit (by AFM and XTEM) surface morphologies similar to that observed in solid-source MBE. The islanding process observed at T@sub s@ = 800 °C relieves 45% of the strain (determined from x-ray reciprocal lattice maps) without the introduction of MDs for thicknesses t up to 31 nm. The dominant facet planes evolve from {105}/{113} to {518}/{111}/{011} with increasing t until coalescence (t = 180 nm). Decreasing T@sub s@ < 600 °C to a regime where @theta@@sub H@ > 0 allows an opportunity to probe new hydrogen-mediated surface morphological pathways. The tendency toward strain-induced roughening and faceting decreases with decreasing T@sub s@ as the initial low-thickness strain-relaxation mechanism and corresponding in-plane feature size changes from strain-induced roughening to MD nucleation when T@sub s@ is decreased below 525 °C. We discuss the details of surface morphological evolution in each of the three temperature regimes (< 525, 525-600, and > 600 °C) in terms of local precursor adsorption and H-desorption kinetics.

9:00am **SS2+EM+NS-ThM3 Interdiffusion During Growth of Ge on Si(100)**, ***H. Jonsson,*** *B.P. Uberuaga, M. Leskovar, B.R. Schroeder, S. Meng, M.A. Olmstead,* University of Washington

We present both experimental evidence and a theoretical explanation of sub-monolayer Ge epitaxy on Si(100) interdiffusion to the 4th layer of the Si substrate. XPD measurements at both 500 C and 700 C show the presence of Ge atoms in the 4th layer. DFT/GGA calculations of the energetics of a Ge atom in the Si surface, together with a statistical model of Ge occupation of the lattice sites, predict occupation of sub-surface sites, with enhanced occupation in the sites under tensile strain. The calculations indicate that the formation energy of a Ge interstitial near the surface (about 1 eV higher energy than the adatom) is significantly reduced as compared with an interstitial in bulk Si, thus lowering the energy barrier for the interstitial diffusion mechanism near the surface. This work was supported by the National Science Foundation, the University of Washington Royalty Research Fund and the Japanese New Energy and Technology Development Organization.

9:20am **SS2+EM+NS-ThM4 Confined Intermixing of Ge and Si**, ***S.-J. Kahng,*** Seoul National University, Korea*; Y.H. Ha, D.W. Moon,* Korea Research Institute of Standards and Science, Korea*; Y. Kuk,* Seoul National University, Korea

In Ge-Si superlattice system, the efficiencies of a possible optoelectric and fast devices can be improved by optimizing the composition at each layer and the abruptness at the interface. It is well- known that intermixing between Ge and Si atoms mainly occurs during the growth process of Si layers on the previously grown Ge layers. Ge atoms tend to segregate toward the surface since the surface free energy of Ge is lower than that of Si. However, with hydrogen adsorbate, the surface free energy of Ge is lower than that of Si, possibly inducing segregation of Si in the Ge overlayer. In this study, Si surface segregation was studied quantitatively for the Ge overlayers grown on Si(100)-(2 x 1) with channeled medium energy ion scattering spectroscopy. The intermixing between Ge and Si presents only at the initial layer in the presence of hydrogen surfactant. Microscopic mechanism for the growth process will be discussed in the view point of kinetics as well as energetics.

9:40am **SS2+EM+NS-ThM5 Surface Segregation and Surface Reactivity in Heteroepitaxial Vapor Phase Thin Film Growth: Si@sub 1-x@Ge@sub x@ on Si(111)**, ***Y.-J. Zheng,*** *A.M. Lam, J.R. Engstrom,* Cornell University

Supersonic molecular beam scattering, x-ray photoelectron spectroscopy (XPS) and low-energy ion scattering spectrometry (LEISS) have been employed to examine the heteroepitaxial growth of Si@sub 1-x@Ge@sub x@ thin films on Si(111) surfaces. Molecular beam scattering has been employed to measure the reactivity of Si@sub 2@H@sub 6@ and GeH@sub 4@ on the Si@sub 1-x@Ge@sub x@ alloy surfaces for a variety of growth conditions (composition and substrate temperature), and these results are compared with results obtained on the clean Si(111) and Ge(111) surfaces. We find that the alloy surfaces are less reactive than both of the pure elemental Si and Ge surfaces. This is in stark contrast to our results for the (100) orientations, where alloy reactivity was always intermediate to that observed on clean Si and Ge. These results reflect the important role played by surface reconstructions on the (111) orientations. XPS and LEISS have been employed in situ to quantify the near-surface Ge concentration of the Si@sub 1-x@Ge@sub x@ epitaxial thin films. Ge segregation is significant, although somewhat less pronounced compared to what we have observed previously on the (100) orientations. Ge segregation, which occurs also in the subsurface layers, has been successfully modeled using both a statistical thermodynamic analytical model, and Monte Carlo simulations.

10:00am **SS2+EM+NS-ThM6 The Role of Arsenic Surfactant in the Growth of Germanium Thin Films on Si(100) Surfaces**, ***C.L. Berrie,*** *J. Bright, S.R. Leone,* University of Colorado, Boulder

The role of arsenic surfactant in the growth of germanium films on Si(100) substrates is investigated using laser single photon ionization time of flight mass spectrometry, reflection high energy electron diffraction, and atomic force microscopy. The energetics of arsenic interaction with Si(100) and Ge(100) are investigated by monitoring the temperature dependence of the desorbing fluxes of As@sub 4@, As@sub 2@ and As atoms from the substrate under a constant incident As@sub 4@ flux. These measurements indicate that the As@sub 2@ interactions with Si(100) and Ge(100) differ dramatically. In particular, the As@sub 2@ flux plateaus in the case of the Si(100) substrate from 800 K to 1000 K. In the case of the Ge(100) substrate, the As@sub 2@ flux monotonically increases over this temperature range and a similar plateau is not observed. Measurements of

the desorbing As@sub n@ fluxes are also made during Ge growth on a Si(100) surface. The dramatic difference in the arsenic interaction on these two surfaces is evident in these measurements as well. The morphologies of the resulting films are monitored ex-situ using atomic force microscopy to determine the effect of arsenic coverage on the size and density distributions of islands formed. As the arsenic coverage is increases, the observed island size decreases and the density of islands increases dramatically. The mechanisms for the Ge growth and the interaction in the presence of the arsenic surfactant will be considered.

10:20am **SS2+EM+NS-ThM7 The Dynamics of the Si(111) (7x7) to (1x1) Phase Transition Investigated by Low Energy Electron Microscopy**, ***J.B. Hannon,*** Carnegie Mellon University **INVITED**

We have used low-energy electron microscopy (LEEM) to investigate the dynamics of the (7x7) to (1x1) phase transition on the Si(111) surface. Because the density of the (1x1) surface is 6 percent larger than that of the (7x7) surface, the conversion from (7x7) to (1x1) requires transport of mass to the domain boundary. By measuring the time evolution of complicated configurations of triangular (7x7) domains, and comparing the results to detailed simulations, we are able to quantify the role of mass transport in the dynamics of this phase transition. We find that individual domains decay approximately linearly in time, with a decay rate determined, not by the domain size, but by the local arrangement of neighboring domains. This observation is counter to the simplest picture of phase boundary motion, in which domain walls move with a constant velocity (independent of environment) determined by the free energy difference between the two phases. We have modeled the effect of the mass transport requirement on the observed decay by solving the two-dimensional diffusion equation for the experimentally-observed configuration of 7x7 domains. We find that the (7x7) domain decay is indeed limited by the supply of the additional material to the boundary. Detailed comparison of the diffusion model with experiment suggests the surprising result that the terrace (rather than surface steps) acts as the primary source of the additional atoms required for the (7x7) to (1x1) conversion. This model reproduces the simultaneous decay of all islands in the field of view with only one adjustable parameter.

11:00am **SS2+EM+NS-ThM9 The Motion of Atomic Steps on Ultra-Flat Si(111)**, ***P. Finnie,*** *Y. Homma,* NTT Basic Research Laboratories, Japan

The flattest silicon surfaces are typically made up of terraces of a low index crystal plane connected by atomic steps. We have studied sublimation and growth on ultra-flat Si(111) substrates@footnote 1@ which have (111) terraces of up to 50 microns in width separated by monoatomic steps. The motion of atomic steps is revealed in time lapse movies made by in situ scanning electron microscopy. When samples are heated to high temperatures (about 1000°C) the surface erodes in a step-flow mode: steps retreat in an orderly fashion, one after the other. Since new terraces are nucleated when step spacings exceed a temperature dependent critical length, the spacing between steps is tunable. Measurements of step velocities as a function of terrace width compare well with theory. The interactions between steps are observed directly by forcing steps to collide@footnote 2@ either destructively, in which opposing steps annihilate, or constructively, in which steps moving in tandem coalesce into double (or triple, or larger) steps. We also studied molecular beam epitaxy in this high temperature regime. Using an electron beam to supply a flux of elemental silicon, desorption is countered and growth occurs in the step-flow mode. Remarkably flat grown surfaces can be obtained in this way since step-flow growth maintains an ultra-flat profile. Monoatomic height island nucleation can also be observed, with circular islands growing to diameters of tens of microns, seamlessly merging with neighboring terraces as the steps collide destructively. @FootnoteText@ @footnote 1@ Y. Homma, H. Hibino, T. Ogino, and N. Aizawa, Phys. Rev. B 55 (1997) R10237 @footnote 2@ P. Finnie and Y. Homma, Phys. Rev. Lett. 82 (1999) 2737.

11:20am **SS2+EM+NS-ThM10 Novel Growth Behavior of Ge on Pb Covered Si(111) Surface**, *I.S. Huang,* Academia Sinica, ROC; *T.C. Chang,* Academia Sinica, ROC, Republic of China; ***T. Tsong,*** Academia Sinica, ROC

A surfactant can modify the growth behavior of a system to our advantages. We report discovery of a novel growth behavior in a Pb-layer promoted growth of Ge on Si(111). For this system, not only can Ge atoms grow on Si(111) surface layer by layer for many layers, but the growth behavior is contrary to traditional nucleation and growth theory and most experimental results in epitaxy. This growth is not governed by the reaction-limited-aggregation (RLA) process at low temperature as has been found in traditional epitaxy. The most interesting feature we find is that a compact-to-fractal island shape transition occurs when the sample temperature is increased, or the deposition flux is decreased. In traditional epitaxy, fractal growth occurs by diffusion-limited-aggregation (DLA) at low temperatures, thus fractal growth will disappear as the sample temperature is raised to the extent that step edge diffusion can occur. Our observation demonstrates that fractal islands can also be grown by RLA, and the importance of reactions in aggregation of Ge atoms in this system. Earlier theories neglect the reaction processes which may be acceptable for metal-on-metal systems, but are not good enough for other systems. Our result points to a need to develop a more complete nucleation theory where both diffusion and reaction are properly taken into account.

11:40am **SS2+EM+NS-ThM11 The Growth of High Density, Small Ag Islands at the Si(111)7x7 Surfaces with Adatom Defects**, ***H. Hirayama,*** *H. Okamoto, K. Takayanagi,* Tokyo Institute of Technology, Japan

We studied the growth of Ag islands on the Si(111)7x7 surfaces with missing adatoms. The missing adatoms were created by 0.5keV Ar ion bombardment. Ag atoms were deposited on the surfaces at room temperature. The growths were observed by using scanning tunneling microscope (STM). At the Si(111)7x7 surface of no missing adatoms, several half unit triangular cells of the 7x7 reconstruction changed their contrast to be bright in the initial stage of the Ag deposition . Then, at the coverage above 0.03 monolayer (ML), each bright triangular cell changed to a bright spot. With the coverage, the number of bright spots increased, and occasionally two spots in neighboring cells kissed. At 0.8ML, the surface was covered by the two-dimensional (2D) percolating network of kissing spots. On the network, three-dimensional (3D) Ag islands grew. Meanwhile, at the Si(111)7x7 surfaces with missing adatoms, the bight spots appeared at the very early stage of the Ag deposition. The spots appeared as to avoid missing adatom sites. This resulted in an imperfect 2D network; the connection of kissing spots were cut into pieces here and there. On the imperfect 2D network, 3D islands grew above 0.7ML. At the stage of the 3D island growth, the missing adatom sites of the underlying 2D network was never filled up. However, we found strong dependence of the size and density of the 3D islands on the number of missing adatoms at the starting surfaces. With an increase of missing adatoms, the size of the islands decreased, while the density increased dramatically. The effects of the missing adatoms on the 3D Ag islands growth were attributed to the substantial increase of the super saturation and the decrease of the diffusion constant of Ag atoms on the imperfect 2D network.

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