SYMPOSIUM KK
Kinetics-Driven Nanopatterning on Surfaces

November 29 - December 2, 2004

Chairs

Eric Chason
Division of Engineering
Brown University
Box D
Providence, RI 2912
401-863-2317

Hanchen Huang
Mechanical Engineering
Rensselaer Polytechnic Institute
(JEC 2038)
110 8th St.
Troy, NY 12180-3590
518-276-2020

George Gilmer
Lawrence Livermore National Laboratory
L-353
Livermore, CA 94550
925-423-0697

Enge Wang
CAS
Institute of Physics
Beijing, 100080 China
86-10-8264-9469

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* Invited paper

In heteroepitaxial growth of strained layers, the growing layer often remains planar up to some “critical thickness,” at which point three-dimensional (3D) islands form. This Stranski-Krastanov transition is of great practical importance. Many device structures require smooth planar layers, while conversely, 3D islands hold promise as self-assembled quantum dots. The transition from planar growth to islands poses a longstanding puzzle in the understanding of the transition is the continuous increase of surface composition due to the growth scenario is still missing. In this talk I will present a microscopic view of the growth and overgrowth of InAs quantum dots on GaAs(100) obtained by scanning tunneling microscopy. The atomic-scale experiments demonstrate that the shape and composition of quantum dots are determined by the delicate interplay between thermodynamic and kinetic effects. Moreover, the STM measurements reveal that only two well-defined island shapes, pyramids and domes exist. Both structures are very similar to the nanocrystals observed in the Ge/Si system suggesting that the pyramids and domes are universal island shapes, independent of specific material parameters. Also the overgrowth scenario that emerges from our measurements, in which the QD capping can be described as a backward transition from steeper domes to shallower pyramids, is material independent as long as the surface energy of the cap material is higher than that of the island material.

Pattern Formation on Silicon and Silicon-on-Insulator. Max G. Lagally, Department of Materials Science and Engineering, Max Planck Institute for Solid State Research, Stuttgart, Germany.


KMC Computations of Strained Epitaxy in 3 Dimensions. Peter Smereka1, Giovanni Russo2, Leonard Sanders3 and Jason Devita4; 1University of Michigan, Ann Arbor, Michigan; 2University of Catania, Catania, Italy.

that appear to be neither stable nor metastable according to a simple energetics analysis; free energy is reduced as islands translate in certain directions in response to controlled variations in oxygen partial pressure in the chamber are also observed, possibly indicating the presence of an unexpected large stoichiometric range in Cu$_2$O nano-islands compared with bulk Cu$_2$O. Significant differences are seen in the oxidation behavior of the Cu film and crystal properties of the resulting islands. Insight to the nature of these effects is obtained from kinetic simulations based on their experiments.

The early-stage oxidation behavior of the Cu (001) surface is being investigated at the Advanced Photon Source via in-situ x-ray diffraction techniques. Single crystal (001) Cu thin films grown on (001) SrTiO$_3$ substrates are oxidized to form crystallographically-aligned Cu$_2$O nano-islands on the Cu surface. By measuring the oxide growth kinetics as a function of oxygen partial pressure, the thermodynamic limit between oxide and reduction has been determined for several temperatures and film thicknesses. We find the Cu/Cu$_2$O phase boundary at much larger oxygen partial pressure than predicted by bulk phase equilibria, and also find that the temperature dependence of this phase boundary is smaller than expected. Large reversible changes in the oxide lattice parameter of up to 0.5% in response to controlled variations in oxygen partial pressure in the chamber are also observed, possibly indicating the presence of an unexpected large stoichiometric range in Cu$_2$O nano-islands compared with bulk Cu$_2$O. Significant differences are seen in the oxidation behavior of the Cu film and crystal properties of the resulting islands. Insight to the nature of these effects is obtained from kinetic simulations based on their experiments.

11:30 AM **KK1.8/JJ1.8/U1.8**

Orientation dependence behavior of self-assembled (In,Ga)$_3$As quantum structures on GaAs substrates.

Shahram Seydolamohamadi, Zhiming W. Hong, Wen and Gregory J. Salamo; Physics, University of Arkansas, Fayetteville, Arkansas.

The orientation of the substrate can play an important role in the formation of (In,Ga)$_3$As quantum structures grown on GaAs. In particular, the so-called ‘‘O-I’’ surfaces morphologies are more pronounced allow the substrate to act as a template for the growth of self-assembled quantum wires (QWRs) and quantum dots (QDs). In this work, the formation of self-assembled (In,Ga)$_3$As quantum structures are discussed with an emphasis on the crystallographic orientation of the GaAs substrates. We report on the effect of the orientation of a GaAs substrate on the evolution of strained (In,Ga)$_3$As quantum structures, along one side of the stereographic triangle from (111)A, including (100) and (111)A surfaces, where N is equal to 7, 5, 4 and 3. The samples were grown by UHV molecular beam epitaxy (MBE) and characterized by atomic force microscopy (AFM) and scanning tunneling microscopy (STM). For the same coverage of (In,Ga)$_3$As, a transition from zero dimensional (In,Ga)$_3$As QDs to one dimensional QWRs is observed as a function of the orientation of the substrate moving from (100) toward (311)A along the stereographic map. While (In,Ga)$_3$As QDs form on GaAs (100) and (711)A surfaces, we observe QWRs formation on GaAs (311)A and (411)A surfaces. In between, GaAs (511)A is covered by elongated (In,Ga)$_3$As QDs. When taken together with the observation of QDs on the GaAs (100) and (711)A surfaces and QWRs on the GaAs (311)A and (411)A surfaces, the texture observed on the GaAs (511)A surface is interpreted as observing a transition taking place from quantum dots to quantum wires on the (511)A surface. The overall picture then, that is a continuous transition from QD to QWR takes place along the mentioned stereographic line. We will present an explanation for the transition from QD to QWR behavior along the stereographic triangle based on the changing surface morphology of the substrates.

11:45 AM **KK1.9/JJ1.0/U1.0**

Energetics of Trench Formation Around Ge/Si Quantum Dots. Dhananjay Tulsiram Tambre and Vivek B. Shenoy; Solid Mechanics, Brown University, Providence, Rhode Island.

At high growth temperatures, the midfoot strain at the boundary of a Ge quantum dot on Si(001) is relieved by the formation of a trench around the base of the dot. The depth of the trench has been observed to saturate at a level that depends on the base-width of the island. Using finite element simulations, we show that the self-limiting nature of trench depth is due to a competition between the elastic relaxation energy gained by the formation of the trench and the surface energy cost for creating the trench. Our simulations predict a linear increase of the trench depth with the island radius, in quantitative agreement with recent experimental observations.

**SESSION KK2/JJ2/U2: Joint Session: Patterning and Steps on Surfaces**

Chairs: Jim Evans and Richard Vinci

Monday Afternoon, November 29, 2004

Room 210 (Hynes)

1:30 PM **KK2.1/JJ2.1/U2.1**

One-dimensional interfaces in Two-dimensional Materials Structures*. Ellen D. Williams1,2, Daniel Dougherty1, Chengguang Tao1, Oleksander Bondarchuk1,2, Theodore L. Einstein1,2, Michael S. Fuhrer3 and Philip J. Rous4; MBE University of Maryland, College Park, Maryland; 2Physics, University of Maryland, College Park, Maryland; 3Physics, University of Maryland Baltimore County, Baltimore, Maryland.

Steps, island edges and domain boundaries are one-dimensional interfaces that serve as the locus of material transport, and as interfacial barriers for electron transport. These interfaces fluctuate under thermal excitation, with length and time scales that can be observed directly using scanning probe imaging. Quantitative characterization of these fluctuations using the tools of statistical mechanics yields energetic and kinetic parameters that can be used to predict evolution of structure under external driving forces (e.g. temperature gradient, growth or sublimation, electromigration). In addition, as the size of the bounded structure decreases into the nanoscale, the stochastic aspects of the fluctuations themselves become a significant component of the material properties. Scanned probe measurement of fluctuations, correlation, autocorrelation, survival and persistence, will be presented for steps (on Ag, Pb and Cu$_2$O) and domain boundaries (Pb/Si, Ag/Si and Cu$_2$O). The meaning of system size in designing, evaluating and using these results will be explained. The impact of the one-dimensional structures on electron flow will also be presented. Direct measurements of step fluctuations in the presence of an electromigration current density of up to $10^7 	ext{A/cm}^2$ will be shown and interpreted in terms of the limits on effective charge for mass displacement at the line boundary. Measurements of the noise and resistivity in electron transport will be shown and characterized in terms of structural fluctuations in a film near the percolation threshold. * Different aspects of this work have been supported respectively by the DOE-BES, NSF-NIRT and NSF-MRSEC.

2:00 PM **KK2.2/JJ2.2/U2.2**


Abstract Adsorbed on a solid surface, a molecule can migrate and carry an electric dipole moment. A nonuniform electric field can direct the motion of the molecule. A collection of the same molecules may aggregate into a monolayer island on the solid surface. Place such molecules on a dielectric substrate surface, beneath which an array of electrodes is buried. By varying the voltages of the electrodes individually, it is possible to program molecular patterning, direct an island to move in a desired trajectory, or merge several islands into a larger one. The dexterity may lead to new technologies, such as reconfigurable molecular patterning and programmable molecular cars. This paper develops a phase field model to simulate the molecular motion and patterning under the combined actions of dipole moments, intermolecular forces, entropy, and electrodes. Slides of this talk will be available at www.deas.harvard.edu/suo Z. Suo and W. Hong, PNAS 101, 7874 (2004). Y.F. Gao and Z. Suo, J. Appl. Phy. 93, 4276 (2003). W. Lu and Z. Suo, J. Mech. Phys. Solids, 49, 1937 (2001).

2:30 PM **KK2.3/JJ2.3/U2.3**

The Quasicontinuum Monte Carlo method for simulating surface growth. Leonard M. Sande1, Jason Devita1 and Peter Smerick2; 1Physics, University of Michigan, Ann Arbor, Michigan; 2Mathematics, University of Michigan, Ann Arbor, Michigan.

We have developed an algorithm for treating growth on surfaces which treats the adatoms as a continuous fluid, and the islands and steps as
collections of discrete particles. The method gives an accurate account of shot-noise fluctuations by converting adatoms to solid one atom at a time. For the case of irreversible growth, we can treat fractal island shapes, multilayer growth, the effect of Erlich-Schwoebel barriers, and nucleation. The algorithm can, in favorable cases, be faster than Kinetic Monte Carlo. For growth near equilibrium we need to treat attachment/detachment from the same footing. This is much more delicate than the irreversible case. We will discuss the issues that arise, and how we deal with them.

3:30 PM KK2.4/J12.4/U2.4
Continuum Theory of Interacting Steps on Crystal Surfaces in (2+1) Dimensions.
Dionisos Margetis 1 and Robert V. Kohn 2.

The difference-differential equations of step motion on crystalline surfaces below the roughening transition temperature are formulated for closed interacting steps of reasonably arbitrary shape in (2+1) dimensions. Among the major kinetic processes considered are the diffusion of adatoms across terraces, and the attachment and detachment of atoms at the step edges, with inclusion of the Erlich-Schwoebel barrier. Basic ingredients of the formulation are (i) the approximate solution of the Laplace equation for the adatom concentration on each terrace within the quasistatic approach via separation of the space variables into “fast” and “slow” ones and application of asymptotics, and (ii) an analytical expression for the step chattering due to a sufficient number of interactions, which include local elastic dipole-dipole interactions as a special case. The continuum limit of the discrete step-flow equations is examined in detail, with particular emphasis on the case with nearest-neighbor interaction. This limit leads to a nonlinear differential equation (PDE) of fourth order for the height profile, which accounts for both step stiffness and step-step interaction. The PDE is compared to the one derived from the standard continuum theory.

Formation of Step/Terrace Structures is Examined in Detail, with Particular Emphasis on the Case with Nearest-Neighbor Interaction.

Formation of step/terrace structures is experimentally found to develop step arrays with large spacing of the order of 10 nm or more after annealing at temperatures where sublimation becomes important. These are caused by transient edges around the edges that initially develop during annealing and form a barrier to step motion before eventually breaking down. This produces a step distribution that can be approximated as an array of steps of the same sign, but of varying terrace width. The analysis of the evolution of such an array of steps presents new aspects from those associated with cases where the step train consists of nearly evenly spaced steps. We have done computer simulations using one dimensional Burton, Cabrera and Frank (BCF) theory with attachment-detachment and step-step repulsion for this configuration. The results suggest that under conditions where there is significant motion of the entire train of steps, the step/terrace configurations and is shown to reduce to the latter under certain local conditions on the step interactions. The role of longitudinal currents, which are parallel to the steps, is discussed.

3:45 PM KK2.5/J12.5/U2.5
Formation of Ridges on Patterned Membranes and Their Role in Evolution of Step Arrays on Membranes.
Kee-Chul Chang and Jack M. Blakely; Materials Science & Eng, Cornell University, Ithaca, New York.

Mesa structures fabricated on Si(111) surfaces have been experimentally found to develop step arrays with large spacing of the order of a micron or more after annealing at temperatures where sublimation becomes important. These are caused by transient edges around the edges that initially develop during annealing and form a barrier to step motion before eventually breaking down. This produces a step distribution that can be approximated as an array of steps of the same sign, but of varying terrace width. The analysis of the evolution of such an array of steps presents new aspects from those associated with cases where the step train consists of nearly evenly spaced steps. We have done computer simulations using one dimensional Burton, Cabrera and Frank (BCF) theory with attachment-detachment and step-step repulsion for this configuration. The results suggest that under conditions where there is significant motion of the entire train of steps, the step/terrace configurations and is shown to reduce to the latter under certain local conditions on the step interactions. The role of longitudinal currents, which are parallel to the steps, is discussed.

4:00 PM KK2.6/J12.6/U2.6
Self-ordering of Nanofacets on Vicinal Si(001) Surfaces and Its Application to Heterogeneous Nanostructures.
Satoru Tanaka, Kazunori Terada, Tomoyuki Miyamoto, Masahiro Fujii and Hideo Suzuki; Ritsumeikan Univ., Sapporo, Japan.

Surface nanostructures by taking advantages of self-ordering or self-organization have been attracted much attention in nanofabrication of semiconductor or device structures. Periodically and spontaneously ordered semiconductor surfaces reveal significantly fine and spatially uniform patterns, which are beyond the lowest limit of electron beam lithography. Formation of step/terrace structures is typically observed on technical surfaces on the step/terrace configurations. As an example we mention that Si and GaAs have been studied, both for device applications and for understanding of basic surface physics. Silicon carbide is an important compound semiconductor, which possesses possibly the highest degree of anisotropy of any semiconducting material. It is thus a promising semiconductor for the next generation of high power and high frequency electronic devices. Initially, we have investigated step/terrace configurations on vicinal Si(001) surfaces after high temperature H2 annealing and found that self-ordered nanofacets, consisting of pairs of (0001) and (11-20), were generated [1]. The origins of such periodic nanofacets were discussed in terms of equilibrium surface phase separation and surface elasticity theory followed by Marchenko et al. [2]. Note here that the periodicity of such nanofacets on 4H and 6H-SiC surfaces is superior to other materials such as Si and GaAs. We believe this is due to its polytypic nature. In this presentation we report such ordered nanofacets found under various etching conditions and discuss possible control of surface nanostructures and also an application to a heterogeneous epitaxial system such as GaN nucleation on SiC [1]. H. Nakagawa, S. Tanaka, R. Maruyama, Y. Hasegawa, Y. Toyokawa, and T. Kurihara, Japan Synchrotron Radiation Facility, Tsukuba, Japan.

4:15 PM KK2.7/J12.7/U2.7
The Controlled Self-Assembly of Nanostructures by the Activated Stranski-Krastanow Transition Method.
Cheng-hsin Chiu, Z. Huang and C. T. Poh; Department of Materials Science, National University of Singapore, Singapore, Singapore.

The growth of nano-crystalline islands on the surface of Stranski-Krastanow (SK) film-substrate systems by self-assembly is a promising technique for device applications. A critical issue in realizing the self-assembly technology is to control the sizes, the locations, and the shapes of the nano-structures. In this talk we propose that the controlled self-assembly of nano-islands can be achieved by a novel scheme, namely the activated Stranski-Krastanow transition (ASKT) method. The basic idea of the ASKT method is to make simple patterns on the film that is in a special thickness range and then anneal the system to activate the self-assembly of the patterns into the islands. The method can control the island formations, and by varying the geometry of the barriers, the method has the capability to fabricate different island structures. For example, changing the size, the aspect ratio, and the height of a rectangular pattern, the ASKT method can yield a two-by-two island array, a square ring with a single or multiple rims, and multiple wires. In addition, the feature length of the structures can be a small fraction of the pattern size, and the length can be tailored by adjusting the mismatch strain in the film without reducing the pattern size. Most importantly, the instability of the barriers against coarsening and shape transition, a common phenomenon that causes serious problems in device applications, can be avoided in the ASKT method. The theory of the ASKT method is presented in this talk together with three-dimensional numerical simulation that demonstrates the potential capability of the ASKT method.

4:30 PM KK2.8/J12.8/U2.8
Stability and Evolution of Nano-ripples on Crystalline Surfaces: Ashwin Ramasubramaniam and Vivek B. Shenoy; Division of Engineering, Brown University, Providence, Rhode Island.

The relaxation of nano-ripples on metallic and semiconductor surfaces is studied using a nonlinear continuum approach that accounts for the formation and interaction energies of surface steps as well as the Schwoebel barrier at step-edges. This method is used to model recent experimental studies of relaxation of a Cu(001) surface. The model results include process is in the attachment-detachment regime at the temperatures of interest here. Ripples are seen to decay with the formation of step-free regions or facets which indicates that the line-tension of the steps plays an important role in the decay behavior. Although the ripples have a dominant spatial frequency or wavelength, our studies find that the decay behavior is not same as that of a sinusoid of a single wavelength. The inherent nonlinearity of the evolution equations leads to significant changes in the modes in the vicinity of the dominant wavelength. Numerical calculations that account for these coupling effects are in very good agreement with the experimental observations.

4:45 PM KK2.9/J12.9/U2.9
Dislocation Driven Surface Dynamics on Solids.
Sanjay V. Khare 1,2, S. Kodambaka 1,2, W. Swiech 2,1, K. Ohmori 1,2, I. Petrov 2,1 and J. E. Greve 2,1.
1Department of Materials Science and Engineering, University of Illinois, Urbana, Illinois; 2Frederick Seitz Materials Research Laboratory, University of Illinois, Urbana, Illinois.

Using low-energy electron microscopy, we investigate the near-equilibrium dynamics of surface-terminated dislocations. We observe, in real time, the thermally-driven diffusion of dislocations that become pinned due to variations in the temperature-dependent angular velocities (w(T)) around cores of temperature-dependent angular velocities (w(T)) around cores of spiral geometry, N2 partial pressure, annealing time, and temperature.
SESSION KK3: Poster Session

Chair: Peter Anderson
Monday Evening, November 29, 2004
8:00 PM
Exhibition Hall D (Hynes)

KK3.1
Curvature Dependent Surface Relaxation on Si by Low-Energy Ion Beam Erosion. Frank Frost, Bashkim Ziberi and Bernd Rauschenbach; Leibniz-Institute for Surface Modification, Leipzig, Germany.

During the last years it has been shown that low-energy ion beams are a versatile tool for nanostructuring of various surfaces via self-assisted processes. Ballistic drift or formation caused by ion beam erosion. Additionally, ion beam erosion can be used as an alternative process for surface smoothing and the preparation of ultra-smooth surfaces. Provided that in the evolution of surface topography different relaxation mechanisms dominate roughening (e.g. caused by curvature dependent sticking) smoothing of initially rough surfaces can occur.

In this work, the surface smoothing of Si surfaces by Ar+ ion beams (ion energy E_{ion} < 2000 eV) was analyzed. Atomic force microscopy (AFM) has been used to systematically investigate the topography evolution of the surfaces with respect to different process parameters (ion energy, ion incidence angle, erosion time, sample rotation). From the AFM measurements the surface roughness was quantified by characterizing the first order (height) and second order (power spectral density - PSD) statistical quantities. Based on the time evolution of these roughness parameters the relevant surface relaxation mechanisms responsible for surface smoothing have been discussed. Especially, it is shown that (i) low-energy Ar+ ion beam erosion of Si surfaces leads to various surface topographies, (ii) smoothing dominates for normal and near-normal ion incidence, (iii) if smoothing occurs, the minimum achievable surface roughness is limited by atomic noise, (iv) for low-energy Ar+ ion beam erosion of Si surfaces ballistic drift (atomic transport parallel to surface) and ballisticdiffusion are the dominant relaxation mechanisms, (v) secondary sputter effects caused by backscattered projectile ions and sputtered Si atoms have a great impact on the evolution of topography, and (vi) smoothing by atomic ballistic drift is the most efficient smoothing process at short lateral length scales at normal and near normal ion incidence, respectively. In conclusion it will be shown that ion beam smoothing is suitable for the polishing of various Si surfaces down to 0.1 nm root mean square (rms) roughness level suggesting a great promise for large area surface processing, which is essential for many advanced optical applications (e.g. x-ray optics, components for the extreme ultraviolet lithography).

KK3.2
Formation of Aligned Microfibers via Self-Assembling Silica Nanoarrays. Valery M. Shklover; Lab of Crystallography, ETH Zentrum, Zuerich, Switzerland.

Controlled assembling of order low-dimensional nanostructures and nanomaterials is a key requirement of many modern applications, such as for numerous applications in optoelectronics and sensors.

The aim of this work, which was intended as part of more general study of the use of self-assembling in design of new nanodevices, was (a) fabrication of microscopic colloidal arrays with shear flow self-assembling method, (b) study of influence of symmetry of crystallization cell environment on the structure of self-assembled arrays, and (c) study of influence of heat treatment on the structure of self-assembled arrays. We tried also to establish the peculiarities of relaxation mechanisms, (v) secondary sputter effects caused by backscattered projectile ions and sputtered Si atoms have a great impact on the evolution of topography, and (vi) smoothing by atomic ballistic drift is the most efficient smoothing process at short lateral length scales at normal and near normal ion incidence, respectively. In conclusion it will be shown that ion beam smoothing is suitable for the polishing of various Si surfaces down to 0.1 nm root mean square (rms) roughness level suggesting a great promise for large area surface processing, which is essential for many advanced optical applications (e.g. x-ray optics, components for the extreme ultraviolet lithography).

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KK3.4
Studies of Si Surface Morphology Evolution during Ar+ Ion Bombardment. Gozde Ozaydin1, Ahmet Serkan Ozcan2, Yiyi Wang3, Justin Hutchins4, Karl F. Ludwig1, Randall L. Hendrick2, Hauke8, and Charles B. Edelstein9; 1Physics, University of Vermont, Burlington, Vermont; 2Physics, University of Vermont, Burlington, Vermont; 3 Naval Research Laboratory, Washington, District of Columbia.

A systematic study of Si surface evolution during Ar+ ion bombardment is reported. Real-time grazing incidence small-angle x-ray scattering (GISAXS) measurements were performed at the National Synchrotron Light Source of Brookhaven National Laboratory.
Laboratory. Ex-situ atomic force microscopy was also used to provide real-space information. Si (100) samples were bombarded at ion energies ranging from 300 to 1000 eV. For normal-incidence sputtering at room temperature, the development of correlated structures with two different characteristic length scales was observed. The shorter length scale features \( \text{nm} \) comained with time but had a typical spatial wavelength of 30 \( \text{nm} \) at all energies examined. The longer characteristic length, however, increased with energy for a given sputter erosion time. Studies performed on Si (111), Si (110) and amorphous samples gave similar results, suggesting that crystalline Si surfaces begin forming during ion sputter erosion. To study the temperature dependence of the surface evolution, Si (100) samples were bombarded with 500 eV ions at temperatures ranging from 25 – 500 C. At temperatures above 300 C, the dots become rougher, suggesting that, at the ion fluxes used, there may be a crossover from ion-induced surface amorphization to a crystalline surface at high temperatures. The time evolution of surface morphology during off-axis ion bombardment of Si (100) at room temperature was also studied. In some parameter regimes, two sets of perpendicular ripples were observed with distinctly different wavelength.

The constrained growth and patterned distribution of nc-Si from a-SiN\(_x\)/a-Si:H/a-SiN\(_x\) mechanism and experiments. We propose a model on constrained growth free energy and finally causes the growth to halt. From our model, we determine the critical a-Si sublayer thickness of 34nm for the constrained crystallization growth and interpret the increase of 3D island formation as the thickness of a-Si:H layers. These results demonstrate that the present model and method is promising to fabricate various patterned nc-Si arrays for device applications simply by changing the geometry of the mask.

Topography Evolution of Si(001) Substrate Fabricated by Ar\(^+\) Ion Beam Sputtering. Hyung Seok Kim, Je-Hyun Suh and Chan Gyu Kang; Materials Science and Engineering, Pohang University of Science and Technology(POSTECH), Pohang, Kyungbuk, South Korea.

Ion beam sputtering is a widespread experimental technique, used in a large number of applications, such as thin film deposition, sputter etching, substrate cleaning and surface analysis. Recently, it has been reported that ion sputtering produced nanopatterns and quantum dots on Si substrates. In certain conditions, the ion sputtered nanopatterns are very important for the development of optoelectronic and quantum devices because of the low cost and efficient mass production of nanometer scale surface structures. For improving control of the ion beam sputtering, it is required that process parameter effects on the formation characteristics of surface nanostructures should be quantitatively understood. In the present study, for the fabrication of nanopatterns, Ar\(^+\) ion beam sputtering was performed on Si(001) wafers irradiated with an Ar\(^+\) beam from a 3cm Kauffman ion gun and atomic force microscopy was performed in tapping mode. Ion sputter-etching is the successful fabrication method for nanometer scale structures such as ripples and 3-D islands on the Si(100) substrates. The 3-D island was formed in the condition of low ion beam flux and inclined incidence angle (60\(^\circ\)). As ion beam flux increased, the ratio of ripples on etched Si(100) substrate increased, in contrast to the condition of low ion beam flux and incidence angle, however, independent on ion beam energy and etching time. It was considered that the formation of 3-D islands was induced by local perturbation of surface geometry and the ion beam density and incidence angle were crucial factors for controlling the morphology of Si substrates.

KK3.7

Abstract Withdrawn

KK3.8

Fabrication of High Aspect Ratio Nanoscale Pit Using Carbon Nanotube Probe. Noriaki Arima and Akihito Matsumuro; Department of Micro/nano System Engineering, Nagoya University, Nagoya, Aichi, Japan.

Fabrication of a high aspect ratio nanoelectromechanical systems (NEMS) and nanodevices (below 50 nm) is an indispensable challenge as microelectromechanical systems shrink towards the nanoscale. However, at present, the capability to build NEMS is very limited and a great deal of improvements is required in key areas such as lithography. Here, we focus our attention on a fabrication technique that makes use of the scanning tunnel microscopy (STM) probe. Additionally, we tried to establish a manufacturing method for a nanoscale pit with a high aspect ratio by using carbon nanotube (CNT) as the probe. The CNT probes used in this study is a tungsten wire of 0.3 mm and multilayer carbon nanotube of 20-30 nm diameter. The CNT can be attached to the apex of tungsten probe by pulling-up process from CNT dispersion liquid with a bias voltage. Effective length of the CNT probe was more than 300 nm. The nanostructures produced were Au thin films on mica substrates that were prepared by magnetron sputtering. Fabrication was carried out in an ambient pressure and room temperature. The machining conditions were bias voltage of 1-5 V, tunnel current of 1-8 nA and fabricating time of 10-90 s. The optimum conditions for high aspect ratio fabrication were examined. The results of our experiment show that the threshold voltage exists for the fabrication of the pits between 1 V and 2 V. The depth and diameter of the pit increased with the increase in the bias voltage and tunnel current, respectively. Consequently, a bias voltage of 3 V and tunnel current of 4 nA were found to be the optimum conditions for a high aspect ratio nanoscale pit fabrication. In changing the fabricating time, depth of the pit increased with the increase in fabricating time, with a little change in the diameter of the pit. This demonstrates that CNT probes can be useful for fabricating structures without changing the diameter of nanoscale CNT probe.

KK3.9

Reconstruction of the Hydrogen Bonds in Protein-Drug Nanocomposite Prepared by Infrared-Pulsed Laser Deposition. Sanshiro Nagare 1, Jo Sagawa 2 and Mamoru Senna 2; 1Technical Development, Nara Machinery Co., Ltd., Tokyo, Japan; 2Faculty of Science and Technology, Keio University, Yokohama, Japan.

Nanostructured protein-drug composites for a novel drug delivery system were deposited by infrared-pulsed laser deposition (IR-PLD). Composite targets were prepared by mechanically mixing the powders or chemically mixing the solutions. Species of the background gas during deposition were varied in order to examine the extent of coalescence of ablated clusters and hence the kinetics of film growth on the substrate. For this purpose, AIM observations and in situ monitoring of the film growth by a QCM sensor were performed. Mechanical interaction between the ablated clusters and gaseous molecules results in the momentum loss of the ablated clusters, so that the deposition rate increases with an increase in the temperature of the background gas. Ablation parameters such as laser fluence and pulse frequency were varied systematically to control the degree of vibrational excitation of the target. Varying the degree of vibrational excitation results in partial reconstruction of the hydrogen bonds between the target protein and drug, which are analyzed by the chemical shift of FT-IR. Target preparation also significantly affects the reconstruction of the hydrogen bonds and hence, the nanostructure of the protein-drug composite after PLD.
Nanostructure Analysis of Protein Thin Film Prepared by Wet and Dry Process. Ichiro Taketani', Sayuri Nakayama', Sanshiro Nagare2 and Mamoru Senna1, 1Faculty of Science and Technology, Keio University, Yokohama, Japan; 2Technical Development, Nara Machinery Co., Ltd., Tokyo, Japan.

Protein (mainly silk fibroin) thin films were deposited on polyethylene and Si(100) substrates by a wet or dry process. The former was performed by dip or spin coating and the latter, by pulsed laser deposition (PLD). Microstructures of the films, i.e. film thickness, surface morphology and crystallinity, were examined by AFM, SEM, and thin film XRD. Secondary structure (β-sheet, α-helix or random coil) of the films was examined by FT-IR. In order to quantify nanostructures of the surface of films, we not only measured the average surface roughness, but also the average waveness (number of the peaks per unit length) from AFM images of the cross sections of films. The films made by the wet process is lower in the population of particles than that of the height than that by the dry process.

Emphasis is laid on comparison of nanopattern formation of the films made by both processes. We also varied the substrate and examined the interaction between the film and substrate of the interface. By the chemical shift of film and substrate by spin echo method, Si(100) was found to have higher affinity with the film compared to polyethylene, due to the interaction between the protein and the surface silanol groups.

Protein-Drug Interaction in the Nanocomposites Prepared by UV and IR Pulsed Laser Deposition. Jo Sagawa1, Sanshiro Nagare2 and Mamoru Senna1, 1Faculty of Science and Technology, Keio University, Yokohama, Japan; 2Technical Development, Nara Machinery Co., Ltd., Tokyo, Japan.

Protein (Bovine Serum Albumin)-drug nanocomposites were deposited on Si(100) by pulsed laser deposition (PLD) at two wavelengths, infrared (1064 nm) and ultraviolet (266 nm). We used uniformly dispersed mixture of protein and drug as a target. Primary structure of the protein was analyzed by matrix laser desorption ionization time of flight mass spectrometer (MALDI-TOF-MS). Effects of electronic and vibrational excitation by UV and IR laser respectively, on the nanostructure of the composite were examined. We analyzed the secondary structure of the protein and the interaction with drug using FT-IR. The infrared between protein and drug, especially, the interaction of the reconstruction of hydrogen bond, was examined. We observed surface morphology of the thin films using AFM and TEM, and studied the effects of ablation processes on the nanostructure of the surface. Protein targets without a drug were used for comparison to study the effects of conjugation with drug on the nanostructure and preservation of primary and secondary structures of the protein.


The diffusion of Ga2O3 into the surface of single crystal [001] rutile leads to the insertion of beta-gallia subunits along [210] planes of the parent rutile structure. These linear defects introduce hexagonally shaped tunnels with diameter approximately 2.5 angstroms in diameter, normal to the [001] surface. Because these tunnels may serve as highly reactive sites for the attachment of macromolecules, we are exploring the application of these linear defects for creating nanostructures. The current work investigates the kinetics of defect formation and the factors that affect defect periodicity and orientation. Gallium oxide was applied to the surfaces of [001]-oriented TiO2 single-crystal substrates via a sol-gel spin-coating process using different gallium-containing precursors. Thermal treatments were systematically varied to obtain different defect surface structures. Defect orientation and the distance between these rows of defects was characterized via tapping mode atomic force microscopy.

Photo Etching of Polymide Thin Film by TiO2 Micro Wire Prepared Using Phase Separation-Selective Leaching Method. Yoshinori Sone, Keishi Nishio and Atsuo Yasumori; Department of Materials Science and Technology, Tokyo University of Science, Noda-shi, Chiba, Japan.

The techniques for the solid surface modification have been studied extensively in order to apply to the fabrication of micro or nano devices and biochips. The present method for patterning is mainly photolithography. It employs photo-mask and photo-resist film accompanying with UV light illumination in a particular condition. Currently, electron beam and X-ray lithography are also examined for high resolution patterning, however, these equipments require further high resolution patterning. In this study, TiO2 porous micro wire was prepared by phase separation-selective leaching process, and the micro photo etching of the surface of a polyaniline thin film was examined by use of the obtained wire under UV irradiation in order to apply to the patterning.

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field is mapped using the tool of local atomic stresses. The empirical potentials of Tersoff are used for the interatomic interactions. By choosing neighboring atoms, the stress effect is manifested. Although the stress evolution as the island grows layer by layer, it is found that a trench could potentially form the elements of nanologic circuitry schemes. 

8:45 AM KK4.2

The Stress Driven Rearrangement Instabilities in Electronic Materials and in Helium Crystals. Michael Grinfeld1, Pavel Grinfeld2, Haruo Kojima3, John Little4, R. Masutomi5, Per-Olof Persson6 and Tsvetanka Zheleva7; 1US Army Research Laboratory, Adelphi, Maryland; 2US Army Research Laboratory, Aberdeen Proving Ground, Maryland; 3Massachusetts Institute of Technology, Cambridge, Massachusetts; 4Department of Physics, Rutgers University, New Brunswick, New Jersey.

It was demonstrated, on general thermodynamic grounds, that a flat boundary of non-hydrostatically stressed elastic solids is always (morphologically unstable with respect to "mass rearrangement." The rearrangement can occur via a) melting-freezing or vaporization-sublimation processes at liquid-solid or vapor-solid phase boundaries, b) surface diffusion of particles along free or interface boundaries, c) adsorption-desorption of atoms in epitaxial crystal growth, etc. At present, many researchers believe that the Stress Driven Rearrangement Instability (SDRI) is a universal phenomenon over large length scales. This universality allows investigation of nano-scale SDRI effects in, say, semiconductor nano-technology by means of manifestations of the SDRI in macro-scale experiments with He4 crystals. We discuss the role of SDRI in the problems of solid nano-films epitaxy and low temperature physics. In particular, results related to the dislocation-free Stranski-Krastanov pattern of growth of semiconductor InGaAs quantum dots (QD) on GaAs substrate grown via molecular beam epitaxy are revealed as an evidence of the SDRI mechanism of formation of these nano-structures. The structural perfection, epitaxial quality, size, shape and density of the QD and their relationship to the growth parameters are discussed in terms of the SDRI theory. Then, we discuss our recent low temperature experiments to observe the solid-liquid interface of nano-crystalline Hg under non-hydrostaticic stress. Theoretical studies of the SDRI involves highly nonlinear equations allowing analytical methods only in the initial state of unstable growth. At present, we are unable to explore analytically the most important deeply nonlinear regimes of growth. To avoid this difficulty, we developed numerical tools facilitating the process of solving and interpreting the results by means of visualization of developing morphologies.
developed during growth. These structures are known as quantum fortresses (QFs) because of their four-walled shape. QF surface morphology and strain profile were studied using Atomic Force Microscopy (AFM) and X Ray Diffraction (XRD). AFM showed that QFs are uniform in size and shape and grow preferentially along the <001> direction. It was also seen by AFM that QF inner walls consisted of elongated arrays of atoms parallel to the dimer row direction. To estimate the interplay between the QF lattice parameter (d) and the interatomic distances related to Ge concentration in the QF, XRD measurements showed a lattice parameter (d) variation from 5.36 to 5.38 Å, which brackets the Si lattice parameter (5.43 Å). Pits are initially formed to relax the strain in the QF's and substrate. The simulations were performed for different values of the capping-layer thickness and used to build a suitable model describing the nucleation and the early stages of self-ordering for additional Ge dots grown on the capping layer. We show that such Ge islands tend to form dot clusters of various shapes which depend on the capping-layer thickness. The interplay between the width of the preferred nucleation regions and the island growth dynamics is also discussed. A good agreement between the model predictions and AFM measurements of CVD-grown multilayered Si/Ge structures is obtained in terms of island positioning within the cluster.

11:00 AM KK4.7
Strain-driven nucleation of Ge islands ordered clusters on multilayered Ge/Si structures. Riccardo Marchetti1, Francesco Montalenti1, Leo Miglio1, Giovanni Capellini2, Monica De Seta2 and Florestano Evangelisti2,1 INFN and L-NESS, Dipartimento di Scienza dei Materiali, University of Milano-Bicocca, Milano, Italy; 2INFN and Dipartimento di Fisica, University of Roma Tre, Roma, Italy.

It has been experimentally observed that the growth of Ge islands on strain-modulated Si surfaces results, under suitable conditions, in the formation of nanometric clusters of ordered islands. We have investigated such a phenomenon by theoretical and semiempirical Tersoff potentials, we have computed the strain field induced in a silicon matrix by mesa-shaped Ge islands. The resulting strain profile was calculated as a function of the capping-layer thickness, and used to build a suitable model describing the nucleation and the early stages of self-ordering for additional Ge dots grown on the capping layer. We show that such Ge islands tend to form dot clusters of various shapes which depend on the capping-layer thickness. The interplay between the width of the preferred nucleation regions and the island growth dynamics is also discussed. A good agreement between the model predictions and AFM measurements of CVD-grown multilayered Si/Ge structures is obtained in terms of island positioning within the cluster.

11:15 AM KK4.8
Buffer Layer Patterned of InAs/GaAs Quantum Dot Superlattices. W. Ye, M. Reason, X. Weng and R. S. Goldman; Materials Science and Engineering, The University of Michigan, Ann Arbor, Michigan.

The vertical stacking of self-assembled quantum dot (QD) superlattices (SLs) is often explained by the preferred nucleation of islands at strain energy minima directly above buried dots. However, the mechanisms responsible for lateral ordering of QDs are the subject of continued debate. For example, anisotropic lateral alignment of QDs has been observed in a number of systems, and long "chains" of laterally aligned QDs were reported. A significant remaining question concerns the role of buffer layer patterning on the lateral growth of QDs. For example, anisotropic "mounds" have been reported during the growth of GaAs films, but their formation has not been related to the alignment of QDs. Therefore, we have examined the pattering effects of buffer layers during the growth of QD SLs. Our QD SLs consisted of 2.6 ML InAs and 5 nm GaAs grown by molecular beam epitaxy at 500°C. Prior to QD deposition, GaAs buffer layers were grown under several different conditions, involving low temperature growth and annealing at 580°C and 500°C. For high temperature grown buffers, atomic force microscopy (AFM) reveals relatively flat surfaces with wide terraces. On the other hand, AFM reveals that low temperature grown buffers contain a high density of "mounds" elongated along the [1-10] direction. When single layers of QDs are grown on either high or low temperature grown buffers, isotropic distributions of QDs are observed. Similar results are obtained for 5- and 10-period QD SLs grown on low temperature grown buffers, preferential alignment of QDs along the [1-10] direction is apparent. This anisotropic QD alignment is enhanced as the number of QD periods is increased. We propose a new mechanism for lateral QD alignment, which is based upon buffer layer patterning leading to undulated In-enriched GaAs spacer layers following the initial sets of QD SLs. Similar studies using focused-ion-beam and laser pre-patterned surfaces will also be presented. This work was supported in part by NSF, DOE, ARO, and NASA.

11:30 AM KK4.9
In-situ Monitoring of Formation and Overgrowth of InAs Quantum Dots. Michael Yakimov, Vadim Tokranov, Gabriel Agnello, Jobert van Esden and Sergo Oktyabrsky; College of Nanoscience Science and Engineering, University at Albany - SUNY, Albany, New York.

With a goal of develop a high performance active medium for optoelectronic devices, formation of InAs self-assembled quantum dots (QDs) in GaAs/AlAs multilayered superlattices (SLs) was studied using in-situ methods: RHEED, Auger electron spectroscopy, along with post-growth techniques: TEM, photoluminescence and AFM. QD formation and evolution were analyzed with the emphasis on the role of AlAs or GaAs engineering process and the observed benefits of a thin AlAs capping layer on top of QDs was found to result from the conformal coverage of pyramidal QDs, thus reducing the intermixing of InAs with the matrix. From the AFM measurements reasonably high QD densities and photoluminescence data, the QDs capped with AlAs appear to be larger than those grown in otherwise identical conditions but capped with GaAs. Auger electron spectroscopy was used to evaluate Indium redistribution during QDs capping and shape engineering process. Indium segregation coefficients were found to be almost identical for capping of a single monolayer of InAs by either AlAs or GaAs. We conclude that the QD size dependence on the chemistry of capping layer is primarily due to differences in Inladial diffusion on InAs and AlAs surfaces. These differences are caused by two factors. Firstly, as we have previously shown, diffusion coefficients of Indium adatoms on InAs surface is much smaller than on GaAs. Secondly, we believe this reduced diffusion coefficient inhibits transport of Indium between the dots capped by AlAs. Secondly, lower surface mobility of Aluminum adatoms results in conformal growth of AlAs overlay on top of QDs, while GaAs growth on QD tips is much slower than between QDs. These arguments are supported by differences in RHEED pattern evolution during the overgrowth. Beneficial properties of the developed QD medium are demonstrated by evaluation of laser diodes that demonstrated unpressured thermal stability with maximum lasing temperature of 219 °C and a characteristic temperature of 380 K. The QD structures can withstand two orders of magnitude higher defect density than quantum wells typically used in lasers. 1 2 M. Yakimov, V. Tokranov, and S. Oktyabrsky. MRS Symp. Proc., 648 (2001) P2.6 2 V. Tokranov, M. Yakimov, A. Katsnelson, M. Lamberti, and S. Oktyabrsky. Appl. Phys. Lett. 83, (2003) 833. 3 M. Lamberti, A. Katsnelson, M. Yakimov, G. Agnello, V. Tokranov, and S. Oktyabrsky. MRS Symp. Proc., 799 (2004) Z5.36.

SESSION KK5: Mislit Stress, Dots and Nanowires

1:30 PM KK5.1
Island Electromigration on Si(001) Surface. 2D Monte Carlo Simulation, Florin Nita1,2 and Alberto Pimpinelli1; 1LASMENA, Aubiere, France; 2Institute of Physical Chemistry, Bucharest, Romania.

The island motion as result of a bias in surface migration of Si dimers was studied by kinetic Monte Carlo simulation, using a model that accounts for the decay of pyramidal nanostructures on Si(001) surface.2 As for the nanopyramids decay, in the dependence of their motion direction relative to the dimer rows direction in the plain, and n and [n] are the numbers of nearest neighbours in these two directions. ED is the energy barrier due to the interaction with the substrate and can have two different values, ED1 or ED2, depending on the dimer direction relative to the dimer rows direction in the substrate. The simulations were performed for different values of the diffusion bars. The island diffuse along the substrate and the speed of its centre of mass depends on the value of the bias. For the same sign of the bias, the direction of motion changes with respect to the laboratory frame, but its centre of mass depends on the value of the bias. For the same sign of the bias, the direction of motion changes with respect to the laboratory frame, but its centre of mass depends on the value of the bias. For the same sign of the bias, the direction of motion changes with respect to the laboratory frame, but its centre of mass depends on the value of the bias. For the same sign of the bias, the direction of motion changes with respect to the laboratory frame, but its centre of mass depends on the value of the bias.
Nitride islands in the pre-coalescence growth regime.

Panos Athanasios Patsalas and Stergios Logothetidis; Physics, Aristotle University, Thessaloniki, , Greece.

In recent years the Titanium Nitride (TiN) research is focused to applications in electronics, such as diffusion barriers and ohmic contacts for GaN-based devices. [1] We use a technique called Scanning Near-field Optical Microscopy (SNOM) to characterize the surface of the islands. The research is focused on understanding the growth mechanism of TiN islands on Si(111) surfaces. The results are compared with theoretical models and simulations. The study of the growth kinetics is important for the development of new applications in electronics.

3:30 PM *KK5.5
Self-assembly of BaTiO3-CoFe2O4 Nanostructures.
Haimei Zheng1, L. Mohaddes-Ardabili2, Junlng Wang1, L. Salamanca-Riba1, D. G. Schlam2, Long-qing Chen2 and R. Namsash3;
1Department of Materials Science and Engineering, University of Maryland, College Park, College Park, Maryland; 2Department of Materials Science and Engineering, Pennsylvania State University, University Park, Pennsylvania; 3Department of Materials Science and Engineering and Department of Physics, University of California, Berkeley, California.

BaTiO3-CoFe2O4 nanostructures have been spontaneously formed during heteroepitaxial growth on a single crystal SrTiO3 (001) substrate by pulsed laser deposition. The patterns of the nanostructures change dramatically as the film thickness increases. Above certain thickness (200 nm) films form well-organized CoFe2O4 nano-platelets embedded in a BaTiO3 matrix. The lateral size of the platelets increases from 5 nm to 50 nm when the substrate temperature increases from 750 °C to 950 °C, which can be fitted to an Arrhenius behavior. Lattice mismatch strain, elastic modulus of the two phases and interface energy are considered to play an important role in the dynamic pattern formation of BaTiO3-CoFe2O4 nanostructures. This work is supported by the NSF-MSEC under contract No. DMR-08-80088.

3:45 PM *KK5.6
Self-Assembly and Shape Transitions of Epitaxial Nanowires and Strained Monolayer Islands.
Vivek B. Shenoy, Division of Engineering, Brown University, Providence, Rhode Island.

Several interesting shape transitions have been recently observed during the growth of submonolayer islands on lattice-mismatched substrates. These shapes, which allow relaxation of mismatch strain, include nanowires that show concave boundaries in equilibrium and formation of highly strained islands with widths in the single-digit nanometer range, shapes that show concave boundaries in equilibrium and formation of highly strained islands. In this work, we study the shapes of nanowires using a sharp interface model and phase-field model to study the shapes of nanowires. In addition, we study the role of the anisotropies associated with the creation of nanowire shapes. Particular emphasis will be given to self-assembled growth of regular arrays of epitaxial nanowires that have potential applications as non-lithographically fabricated interconnects.

4:15 PM *KK5.7
Endotaxial Silicide Nanowires.
Zhao H1, David J. Smith1,2, Frances M. Ross1,2 and Peter Bennett2,3;
1Physics, Arizona State University, Tempe, Arizona; 2Science and Engineering of Materials, Arizona State University, Tempe, Arizona; 3Center for Solid State Science, Arizona State University, Tempe, Arizona; T1 J. Watson Research Center, Yorktown Heights, New York.

We demonstrate the growth of self-assembled nanowires (NWs) of cobalt silicide on Si (111), (100) and (110) substrates by deposition of Co onto a heated Si substrate. Island structures are determined using Atomic Force Microscopy, High-Resolution Transmission Electron Microscopy (TEM) in cross-section and in situ UHV-TEM in plan-view. Silicide NWs grow via an endotaxial (embedded epitaxy) mechanism, with coherent B-type interfaces along inclined Si planes, which breaks the symmetry of the surface, and leads to a long, thin nanowire shape. During growth, the length and width of the islands increase linearly with time in a fixed proportion that varies strongly with growth temperature, showing that the nanowire shape is...
Nanostructure growth via the annealing is a simple technique that primarily consists in raising the temperature of a given substrate pre-deposited with a suitable metal catalyst. Previously, there has been reported nanostructure growth just from annealing where silicon oxide nanowires have been grown using gallium as the metal catalyst and the wafer itself as the source material. Recently, we have also been able to demonstrate the growth of nanowires by simple annealing using gold as the metal catalyst in the growth of silicon oxide nanowires as well as nanowires containing indium and antimony using silicon and InSb substrates, respectively. Despite the simplicity in the growth technique, the results in particular from the InSb substrate annealing have not been high quality, with evidence of tapering, variation in the chemical composition along the length of the nanowires, as well as crystal quality not quite reaching the desired single crystal. In order to resolve the shortfalls from the annealed results, a new growth technique using the annealing and pulsed laser deposition is found. InSb substrate as the nanowire source material and gold as the metal catalyst, we demonstrated the new technique. The new technique allows the precise control of the growth temperature thereby resulting in higher quality nanowires than obtained from simple annealing alone. The control over the density and the diameter of the nanowires is also shown. The results from the variation of gold deposits at the fixed growth temperature of 510 °C are shown with scanning electron microscopy (SEM) and high resolution transmission electron microscopy (HRTEM) images.

Growth of Amorphous SiO2 Nanowires on Si using a Pd/Au Thin Film as a Catalyst. Jose L. Elechiguerra, Jorge A. Martinez and Miguel Jose-Yacaman. The University of Texas at Austin, Austin, Texas; Centro de Manufactura, Tecnologico de Monterrey, Monterrey, N.L., Mexico.

Nanostructures of amorphous SiO2 were synthesized by thermal processing of a (100) Si substrate at 1100 °C in the presence of a nitrogen flow, and using a 15 nm thick Pd/Au film as a catalyst. The substrate itself was the only source of silicon for the nanowire growth. The nanostuctures produced were characterized by high resolution transmission and scanning electron microscopy and by x-ray diffraction. The nanowire growth is consistent with the vapor-liquid-solid (VLS) mechanism, with particles of Pd2Si and Au(4) being the active catalyst, and gold from the reaction between silicon and the catalytic film, and to remain at the tip of the wires. The synthesized nanowires showed a well defined flower-like morphology which could be very interesting for lasing applications.

SESSION KK6: Ion Beam Patternining of Surfaces

8:30 AM KK6.1

Coupled Atomistic and Continuum Model for Generation of Surface Morphology in Silicon by Ion Bombardment. Harly T. Johnson, Nagarajan Kalyanasundaram and Jonathan B. Freund. Mechanical & Industrial Engineering, University of Illinois at Urbana-Champaign, Urbana, Illinois; Center for Research in Applied Mechanics, University of Illinois at Urbana-Champaign, Urbana, Illinois.

Semiconductor quantum dot arrays are promising for a wide range of applications in optoelectronics, nanoelectronics, biology, and quantum computing. Successful fabrication of regular, uniform lateral arrays of quantum dots by self-assembly methods continues to be an important problem. Recent work by several experimental groups has identified a possible new method for generating arrays of semiconductor quantum dots using a simple ion-bombardment process. The basic mechanism of rippling and sputter-induced surface instability in semiconductors has been understood for some time, but little work has been done to connect the continuum level mechanisms to atomic scale surface features. In this talk, I will discuss an early stage in the development of this technique, which describes recent work to study the ion sputtering surface instability using a multiscale model that combines molecular dynamics and continuum surface evolution methods. The effects of individual ion impacts are studied using a molecular dynamics analysis of medium energy (500-eV-700-eV) argon ion bombardment of silicon. Empirical potentials are used to model the argon and silicon, sputtering and stress results are in agreement with experimental observations. Both sputtering and ion-assisted surface rearrangement effects are considered, and the statistics are then input into a continuum equation describing the surface and strain energy driven mass transport active over much longer time scales. The surface profile is then calculated using an implicit time integration, with consideration of changing elastic fields as the surface shape evolves. This analysis is valid for the regime in which the surface remains strongly disordered. Results of the multiscale analysis are compared to the arrays of quantum dots grown experimentally by the ion bombardment method.
energy ion sputtering provides a sensitive probe of microscopic surface kinetics. In this study, we report the morphological evolution of one-dimensional self-organized arrays of Au+ ion etched sputter ripples on a hydride-terminated Si(111) surface. The time evolution of ripple formation was studied in situ using UV light scattering spectroscopy in a temperature range from 560-570 °C. We have measured the temperature-flux phase space with the presence of Ehrlich-Schwoebel barriers, as revealed by the adatom capture probability. The importance of thermal fluctuations in the instability is suggested by the observed trend of increasing amplitude with flux. We also observe a substantial reduction in macrostep height and terrace width when native oxide desorption is carried out at 450 °C with V:III ratio=30, which is attributed to diffusion-mediated step attraction due to preferential attachment of Ga adatoms from the upper terrace. This reduction in step height under As-rich conditions leads to the formation of meanders, ripples, and pyramidal mounds on the surface, indicating that the adsorbed arsenic species undergo reaction and incorporate surface steps preferentially from the lower terrace. We have studied the changes undergone by the GaAs(110) surface misoriented towards < 1-12>-type steps. In contrast, growth of GaAs in the presence of H+ beam milling of Ge. Stefan Ichim and Michael J. Aziz; Div. Engrg. & Appl. Sci., Harvard University, Cambridge, Massachusetts; 2Department of Physics, Imperial College of Science, Technology and Medicine, London, United Kingdom.

11:30 AM KK6.9 Fabrication and Atomic Modeling of Ion-Etch Nanostructures on Substrates. Maria Stepanova and Steven K. Dew; Electrical and Computer Engineering, University of Alberta, Edmonton, Alberta, Canada.

We have implemented and investigated numerically a new process to fabricate self-organized metal dots, networks, and nanowires on non-metallic substrates. We have deposited a thin film of Cu on Si or SiO2 substrates and etched the film by a neutralized Ar beam in a variable shadowing ion mill system. Due to the kinetic mechanism known as the sputter instability, ion etching produces a nanostructured surface morphology. At the stage when the etched surface approaches the substrate, we have investigated the surface by SEM and observed rectangular arrays of Cu nanotubes embossed on the substrate. This system provides a useful probe of surface evolution without complicating effects of compositional inhomogeneities and anisotropic diffusion. Our examination of the Si(111) surface indicates that the rippling is driven by the competition between etching (from the ion beam) and capillary action (driving smoothing via surface diffusion). Experimental results show a strong dependence on the sputter yield and 2) presence of a Schwoebel barrier along the ion beam direction. We discuss the implications for the fundamental physics underlying the morphological instability.

10:30 AM *KK6.6 Nonlinear Stabilization Mechanisms in Amplitude Saturation During Sputter Ripping Formation on Silicon. Jepah Elishabach and Allen David Brown; Materials Science and Engineering, Johns Hopkins University, Baltimore, Maryland.

Sputter rippling refers to the formation of regular surface patterns during glancing incidence energetic ion etching of surfaces, usually as a result of a competition between etching (from the ion beam) and capillary action (driving smoothing via surface diffusion). Many different kinds of morphologies are often observed, including ripples oriented parallel or perpendicular to the projected ion beam direction and "quantum dots" arranged in hexagonal or rectangular arrays. Theoretical analyses of ripple evolution have concentrated on the initial stages of the surface instability leading to pattern formation, and the details associated with the non-linear mechanisms leading to amplitude saturation and pattern stabilization remain a subject of active interest. The Si(111) surface is a single component surface with isotropic diffusion kinetics; for these reasons, this system provides a useful probe of surface evolution without complicating mechanisms related to anisotropic diffusion, atom mobility, and surface transport. We also report our numerical investigation of this process. Our model comprises a kinetic Monte-Carlo simulation of nonequilibrium ion-etching of the surface morphology of arrays of Cu clusters on a substrate, and coarsening of the surface patterns. Our simulations correctly reproduce the observed trends and identify the major control factors for the process. The reported process of nanofabrication has a strong potential for nanoelectronics and photonics.

SESSION KK7: Novel Techniques and Applications of Nanopatterning

Chair: Eric Chason and Hanchen Huang

Wednesday Afternoon, December 1, 2004
Room 308 (Hynes)

1:30 PM KK7.1 Nanopatterning of GaAs(110) Vicinal Surfaces by Hydrogen-Assisted MBE. Miguel L. Crepio, Jose L. Sacedon, Bruce A. Joyce and Paloma Tejedor; 1Instituto de Ciencia de Materiales de Madrid, CSIC, Madrid, Madrid, Spain; 2Department of Physics, Imperial College of Science, Technology and Medicine, London, United Kingdom.

Homoeptaxial growth of GaAs on (110) vicinal surfaces under different growth conditions leads to the formation of distinct morphological instabilities associated with step propagation in the presence of Ehrlich-Schwoebel barriers at step edges. Thus, step bunching in the Ga-supply limited regime is attributed to diffusion-mediated step attraction due to preferential attachment of Ga adatoms from the upper terrace. On the other hand, growth under As-deficient conditions leads to the formation of meanders, ripples, and pyramidal mounds on the surface, indicating that the adsorbed arsenic species undergo reaction and incorporate surface steps preferentially from the lower terrace. We have studied the changes undergone by the GaAs(110) surface misoriented towards (111)A by 2 deg, in both growth kinetics and surface morphology during homoepitaxy when i) native oxide removal is carried out at low temperatures under flux of atomic hydrogen and ii) when oxide removal and epitaxial growth are carried out in the presence of atomic hydrogen. Under As-rich conditions (450 C, V:III ratio=30), we observe a substantial reduction in macrostep height and terrace width when native oxide desorption is carried out at 450 C with H+ prior to growth from As4 and Ga beams. In both cases growth proceeds in the step flow mode and leads to the formation of a rather straight step morphology, with step edges along <1-12>-type steps. In contrast, growth of GaAs in the presence of H+ after oxide removal in the same ambient takes place by simultaneous step flow and five-dimensional nucleation on the terraces, the macrostep bow is also reduced and the step edges are preferentially aligned along the <1-12>-type direction. On the other hand, hydrogen-assisted MBE growth of GaAs on the (110) surface under As-deficient conditions (450 C, V:III ratio=10) proceeds by step flow in the presence of Ehrlich-Schwoebel barriers, as revealed by the self-organized pattern of pyramidal mounds aligned along the step direction, <001>, and bounded by <1-12> edges. While this well-ordered structure is formed after deposition of 500 monolayers.
Plasma hydrogenation applied on standard (100)-oriented Czechoslovakia silicon (Cz Si) wafers at about 250°C causes a nanopatterning of the treatment surface. The patterned layers can be discovered for a deposition temperature of 100 nm which is achieved depending on the process conditions, i.e. thin nanoscale silicon layers can be created. The formation of such nanostructures at the surface regions of plasma hydrogenated wafers was investigated for various process conditions. The plasma treatment was applied either at 15.36 MHz or 110 MHz frequency. The impact of post-hydrogenation annealing in vacuum on the morphology and evolution of the wafer was studied up to 1200°C. To the properties of the plasma treated wafer surface and subsurface regions were analyzed by atomic force microscopy (AFM), high resolution scanning electron microscopy (SEM), and depth resolved microtofscopy. The formation kinetics of the nanopatterned silicon layers can be described on the base of a combined etching/redeposition mechanism. During the H-plasma treatment Si atoms are released from the wafer surface, and small Si crystallites are redeposited on the surface. During this process the type of crystalline Si (most probably SiH2) might be formed in the plasma, which are decomposed again and induce the redeposition of the small Si crystallites onto the wafer surface. It was observed that the formation of redeposited crystallites with (111)-orientation is energetically more favourable than the one with (100)-orientation. During annealing at temperatures of 600°C or higher the crystallites in nanopatterned surface layer are dissolved again, and Si atoms formerly located in the redeposited crystallites are rebright onto the wafer surface. This reconstruction is not optimally executed, i.e. the initially (100)-oriented surface is not completely reconstructed again, and extended damage areas with (111)-orientation remain on the wafer surface. Therefore, after high temperature annealing, the reconstructed surface and subsurface layers of the treated wafers exhibit strong tensile stress up to rather deep wafer regions, as was observed by depth resolved micro-Raman analysis. The impact of the hydrogen related defects and the nanopatterning at the wafer surface on various semiconductor device structures will be briefly discussed.

Carbon nanofibers can be produced by decomposition of hydrocarbon compounds on surfaces of transition metals such as Fe, Ni, and Co at moderately high temperatures. Carbon nanofibers consist of graphene layers wrapped into a conical ("herring-bone") or a cup-like ("bamboo") shape that are stacked to form cylindrical nanostructures. The shape and size of these graphene cones (i.e. cone angle) determines the type of material that is produced. For example, multiwall nanotubes are nanofibers with a zero cone angle. The cone angle depends on the shape of the surface of the catalyst nanoparticle at the time they are formed, orientation of the catalytically active surface, kinetics of the deposition process etc. In this work we present electron microscopy studies of catalyst nanoparticle (Ni, Fe, and Co) and vertically aligned carbon nanofibers produced by plasma enhanced chemical vapor deposition on Si(111) and Si(001) surfaces. The results show the correlation between nanoparticle orientation and the Si substrate that in its turn is reflected in the atomic structure of carbon nanofibers. The relationship between catalyst particle equilibrium shape (i.e. particle size) and the cone angle, h, and bcp, respectively, is studied with regard to structure and synthesis dynamics of the resulting nanofibers. A geometrical model based on assumption of growth rate anisotropy is utilized to explain the shape of the catalyst particle-carbon nanofiber interface.

We present an innovative method for the production of vertically aligned, ultrasharp silicon nanostructures with tip diameters as small as 10 nm. Silicon nanocanoes were produced using dc plasma-enhanced chemical vapor deposition (dc-PECVD) using thin film copper or gold as a catalyst and the silicon substrate as a sole source of silicon. High resolution SEM, TEM, EDX, and AES were utilized to determine the microstructure and composition of the nanocanoes. We have explored variations in the structure and growth mode of these nanocanoes with respect to growth environment. This structure will be described in detail and a growth mechanism proposed. The similarities and differences of this new growth process as compared to standard SiH4-based vapor-liquid-solid growth will be discussed. Lastly, we will describe their potential use in applications such as field emission cathodes and gene delivery arrays.

We observe the spontaneous formation of parallel rod structures upon exposing a clean NAI111011 surface to oxygen at elevated temperatures (850-1250 K). By following the self-assembly of individual nano-rods in real time with low-energy electron microscopy (LEEM), we are able to investigate the kinetic processes by which the rods lengthen along their axes and thicken in the direction normal to the surface of the substrate. The oxide rods, whose lengths can be in excess of several microns while their heights and widths are quantized respectively in units of 2 Å and 16 Å, lengthen across virgin areas of the substrate at a constant rate if the oxygen pressure and growth temperature are fixed. The exponential dependence of this axial growth rate on temperature yields an activation energy of 1.1 eV. We do not observe changes in the axial growth rate as growing rods pass within very close proximity to each other, which suggests that they do not compete for growth flux in order to elongate. Apparently, the growth is determined by the rate at which growth species attach to the ends of the rods, rather than by the rate at which growth species diffuse to the ends of the rods from distant points on the substrate. Scanning tunneling microscope (STM) images and movies captured with LEEM during synthesis (desorption) show that the nano-rods can be aligned (thin) perpendicular to the surface in a layer-by-layer fashion. We commonly observe that a rod's rate of elongation decreases sharply when it crosses a step to a lower atomic terrace and increases when it crosses a step to a higher terrace, showing that the number of adatoms incorporated at the step edge increases at step crossings. Diffraction analysis and dark-field imaging with LEEM indicate that these structures are crystalline, likely to be alumina (Al2O3) or spinel (NiAl2O4), and have an epitaxial relationship to the substrate. We wish to discuss the origins of the highly anisotropic structures. This work was supported by the Office

(ML) of GaAs in the presence of H2, we had previously observed the formation of another pattern by conventional MBE growth only after deposition of GaAs. As adatom incorporation kinetics at low temperatures, we have invariably observed a characteristic faceting of the surface with very straight steps along the <110> direction when GaAs growth is carried out at 620°C. The ratio of the step density and step length of GaAs being used during oxide desorption and/or epitaxial growth of...
3:45 PM KKE7.7
Quasi-Periodic Nanopatterning of Heteroepitaxial Layers by Misfit Dislocations Demonstrated for the PbTe on PbSe (001) Case. Gunther Stroppa and Wolfgang Knabe. Institut fuer Halbleiterphysik, University of Linz, Linz, Austria.

Nanopatterning of surfaces using dislocation arrays has recently evolved as a powerful tool for fabrication of ordered nanostructures. Apart from the method of twisted wafer bonding, dislocation formation can also be induced by strained-layer heteroepitaxial growth. Usually, however, in this case rather irregular dislocation networks are formed due to the limited dislocation kinetics. In the present work, it is shown that it is possible to produce highly periodic dislocation arrays in strain-relaxed layers if several conditions are fulfilled. These are (i) the presence of a homogenous dislocation nucleation mechanism, (ii) a high lateral dislocation mobility within the interface, and (iii) the presence of a strong repulsive dislocation-dislocation interaction. These conditions are usually met only in heteroepitaxial systems with large lattice-mismatch and with misfit dislocations that have a Burgers vector confined to the interface plane. As an experimental example, we have studied the case of PbTe on PbSe heteroepitaxial layers grown by molecular beam epitaxy on PbSe (001) substrates. From in situ scanning tunneling studies of the epilayer surface structure as a function of layer thickness it is shown that all three prerequisites are met in this system, and that therefore, nearly perfect square arrays of misfit dislocations are formed. The action of strong repulsive dislocation interactions as driving force for the underlying ordering process is also revealed by the fact that the observed course of strain relaxation can only be explained by theoretical models in which mutual dislocation interactions are taken into account. Due to the effect of the dislocation stress fields it is expected that thermal dislocation annihilation occurs, which can be used as templates for deposition of ordered self-assembled nanocylinders.

4:00 PM KKE7.8
Growth of Nanopillar Arrays using Glancing Angle Deposition. Sai V. Kesapragada, Ken D. Girardin and Daniel Gall; Department of Materials Science and Engineering, Rensselaer Polytechnic Institute, Troy, New York.

Periodic arrays of Si, Ta, Cr, and CrN nanopillars, 0.1-2 μm tall and 50-250 nm wide, were grown by ultra-high vacuum glancing angle sputter deposition, in pure Ar or N2 discharges at 1-3 mTorr. The height and width of the pillars decreases with increasing the azimuthal angle between the incident flux and the surface normal, which was varied from 66°-88°. The nanopillar spacing, 100-500 nm, is controlled by initial surface patterning. Si (100) substrates are patterned by dispersing monolayers of 50nm-diameter polystyrene or 160-nm-diameter silica colloidal spheres to form a periodic close-packed array. E-beam lithography was utilized for patterning down to 50 nm feature sizes. Nanopillars grow perpendicular to the substrate surface on epitaxial layers of high-crystalline quality regardless of the polar axis during deposition. In contrast, a stationary substrate yields columns that are tilted towards the deposition flux.

4:15 PM KKE7.9
Ripple formations in sputtering and depositions by energetic cluster beam irradiations. Noritsuki Toyoda and Isao Yamada; Laboratory of Advanced Science and Technology for Industry, University of Hyogo, Kamigori, Hyogo, Japan.

Formation of ripples in sputtering and deposition process by energetic gas cluster ion beam irradiations are studied using large cluster ions with several thousands of atoms in one cluster ion. The gas cluster ions realize high-density energy depositions near a target surface, which originates non-linear collisions and completely different collision processes from atomic or molecular ion bombardments. Due to this dense energy deposition on surface, near-surface atoms are easily ejected. The most interesting characteristic of gas cluster ion irrations in that gas cluster ions create a lot of laterally sputtered particles, which are called eLateral sputtering. These laterally sputtered particles showed significant influences on ripple formations at oblique incidence of gas cluster ion beams. A plenty of ripples were formed around incident angle of 60° but the surface was quite smooth at near normal or glancing angle irradiations. The ripple formations by gas cluster ions were only observed for sputtering process but also appeared during the gas cluster ion beam assisted depositions. In this study, the ripple formations during sputtering and ion beam assisted deposition processes are studied and formation mechanism will be discussed.

4:30 PM KKE7.10
Large-Scale Epitaxial Growth of Hexagonal Arranged ZnO Nanowires Using Au Nanohole Membranes as Templates. Hong Jin Fan1, Woo Lee1, Kornelius Nielsch1, Margit Zacharias1, Armin Dadorz2 and Alois Krost2; 1Max Planck Institute of Microstructure Physics, Halle, Germany; 2Institute of Experimental Physics, Otto-von-Guericke-University, Magdeburg, Germany.

Fabrication of single-crystalline one-dimensional semiconductor nanostructures with controlled orientation and spatial position is of great importance for their technological applications. We report here the successful large-scale growth of hexagonal arranged ZnO nanowire arrays on sapphire and GaN/Si substrates by combining substrate nanopatterning and the vapor-liquid-solid (VLS) growth process. In a first step, gold membranes were used as catalysts for nanowire growth and template for specifying the position of individual nanowires. As the size (30 to 250 nm) and separation of 500 nm of the Au nanodots are tunable by choosing suitable deposition masks for the Au deposition, the resulting ZnO nanowires are adjustable in their diameter and inter-wire distance. Such nanowire arrays could have applications as photonic crystal, electron field emitters or nanosensor arrays.

4:45 PM KKE7.11
Leakage Currents through Thin Silicon Oxide Grown on Atomically Flat Silicon Surfaces. Valerian Ignatescu and Jack M. Blakely; Materials Science and Engineering, Cornell University, Ithaca, New York.

We report some results that indicate that silicon oxide grown on step-free surfaces has better dielectric properties than that on nominally step-free surfaces. Advantages of microprocessors now feature gate oxides that are 1.2-1.3 nm in thickness. For such thin silicon oxides the leakage current is high enough to substantially increase the power consumption of the transistors and therefore the heat generated by them. The high power dissipation is already a serious problem for current high frequency microprocessors. The most obvious path is to switch to a high-k dielectric material but other insulators, such as hafnium and zirconium oxides, even though they have good dielectric properties, are not compatible with the polysilicon material used for gate electrodes. The use of atomically smooth Si surfaces can potentially reduce leakage currents through silicon oxide layers. An increased roughness of the silicon/silicon oxide interface leads to a reduced local effective oxide thickness and, in consequence, to a higher leakage current [1]. Atomically flat surfaces were obtained by heating patterned silicon samples in ultra-high vacuum following the procedure described elsewhere [2]. After that, we grew a thin oxide layer with several different thicknesses (between 3 and 6 nm) using dry thermal oxidation. The AFM analysis of the surface after the oxidation shows that the topography of the surface is preserved during the oxidation process. The leakage currents were measured for step-free regions. We find that capacitors built on step-free regions are able to withstand a significantly higher voltage before breakdown compared to those on normal stepped regions. Also the absolute value of the slope of the Fowler-Nordheim plot, which is proportional to the effective thickness of the oxide, is higher for the capacitors situated on step-free surfaces. As expected, the difference between the electrical properties of the oxide grown on stepped and atomically flat surfaces is more pronounced for thinner oxide layers. References [1] A.C. Oliver and J.M. Blakely, Mater. Res. Symp. Proc. 743, V 4.6.1 (2003) [2] D. Lee and J. Blakely, Surface Science 445, 32 (2000)
synthesis of only a narrow size distribution, while functional assembly usually implies controlling the location and/or the orientation of the nanomaterials. We emphasize that this directed assembly requirement cannot be met at this time by even the highest resolution electron beam lithography, for both technical and economic reasons. Consequently, successful syntheses often will require spatially confining and orienting growth of nanoscale structures. This presentation will be focused on chemistry approach toward the efficient synthesis and replication of one-dimensional materials. The various growth methodologies developed at our research group for synthesizing such key building block in nanotechnology will be discussed.

8:30 AM KK8.1
The effects of low-energy ion-irradiation on surface pattern formation in sputter-deposited epitaxial TiN(O01) layers. Taeyoon Lee, Kenji Ohmori, Suneel Kodambaka, Joe Greene and Ivan Petrov; Materials Science and the Frederick Seitz Materials Research Laboratory, University of Illinois at Urbana-Champaign, Urbana, Illinois.

We have chosen TiN(O01) as a model system for investigating the effects of low-energy ion-irradiation on surface morphological evolution during sputter-deposition of epitaxial transition-metal nitride layers. As a first step, we used focused ion beam milling technique followed by high-temperature (1400 °C) annealing in air to produce ~5 μm-wide, atomically-flat, step-free terraces on MgO(O01) substrates. Epitaxial TiN(O01) films, 3000 Å-thick, were then grown on these patterned substrates at low-homologous growth temperatures (0.25 < Tg/Tm < 0.37, Tg = 600 - 1050 °C) using ultrahigh-vacuum reactive magnetron sputter deposition in pure N2. The N2 to Ti metal ratio (JN/Je) incident at the growing film was varied from 1 to 14 while the ion energy was maintained constant at ~ 20 eV. Using atomic force microscopy, we find that all TiN(O01) layers grown at Tg < 925 °C exhibit mound growth with mounds whose widths increase exponentially with Tg irrespective of JN/Je. However, layers grown with high flux (JN/Je = 14) consist of mounds that are approximately two times larger than those obtained at JN/Je = 1, suggesting higher surface adatom diffusion under high-flux conditions. In both cases, we observe frustal island shapes indicative of relatively low edge-atom mobilities. TiN(O01) layers grown with high flux at Tg > 925 °C are nearly atomically-flat with only the top 3-4 atomic layers exposed. This is in dramatic contrast to the films deposited at the same temperature under the low-flux conditions, where over 10-20 atomic layer height mounds are observed. Based upon temperature-dependent measurements of the widths of growth mounds and island shapes, we propose that low-energy ion-irradiation preferentially enhances surface-adatom diffusion with little effect on edge-atom mobilities over this growth temperature range.

8:45 AM KK8.2
Self-Organization of Curved Ripples on Titanium Crystals Irradiated with Focused Ion Beam. Wei Zhou1, Xiaixia Qian1, Yongqi Fu2 and Bryan Keck Ann Ngai3; School of Mechanical & Production Engineering, Nanyang Technological University, Singapore, Singapore; NTU, Singapore, Singapore.

We report the formation and self-organization of nano- and micro-scale structural features on titanium crystal surfaces at room temperature with a rastered 1 nA, 30 keV Ga+ focused ion beam (FIB). We use in-situ real-time secondary ion and secondary electron imaging to follow the evolution of surface features from a low dose of 7.5X1016 ions/cm2 to a high dose of 4.8X1018 ions/cm2 and characterize the final surface morphology using AFM. In contrast to usual expectation that surface topology produced at normal ion beam incidence consists of hillocks or depressions, we observe periodic ripples with regular spacing. We provide evidence to demonstrate that the ripples form spontaneously rather than by direct writing of the FIB. Ripples on surface of a single crystal are oriented in the same direction, but the ripple orientation changes with the crystallographic orientation of the patterned titanium crystal surface. We explain the observation using a model in which the ripple structure is the result of the balance between the erosion rate and the anisotropic diffusion of adatoms and vacancies on surface activated by the ion beam sputtering. When the ion beam is tilted by a small angle up to 30 degree, the ripple orientation is still determined by the crystallographic direction. However, when the FIB incident angle is increased to 45 or 60 degree, curved ripples occur at high ion doses. No existing model can explain the morphological evolution from the well developed straight ripples to the curved ones. Therefore, we attempted to present a new model, which attributes the formation of curved ripples to competition between the formation of ripples due to anisotropic sputtering and the formation of incident-angle-dependent ripples of the Bradley-Harper type.

9:00 AM KK8.3

Polycrystalline TiN and related transition-metal (TM) nitride thin films are typically deposited by reactive magnetron sputter deposition and employed as diffusion barriers in microelectronics as well as hard, wear-, and corrosion-resistant coatings in mechanical and optical applications. Since cubic TM nitrides are high melting point materials, control of preferred orientation is essential. We use a combination of HR-XRD, TEM, HR-XTEM, AFM, and STM analyses to characterize micro- and nanostructures. Kinetic Monte Carlo modeling and analytic theory, accounting for anisotropic adatom diffusion, predict that Ehrlich barriers at descending step edges together with kinetic roughening and atomic shadowing processes, are used to develop an atomic scale understanding of microstructural and surface morphological evolution. Advances in ion transport and surface energies required for the models are obtained from in-situ high-temperature STM and LEEM analyses. The overall results yield design rules which allow the synthesis of controlled microstructures with greatly enhanced physical properties. Taking these concepts a step further, and using spontaneous natural patterning processes found in nature, we show that self-organized nanostructures consisting of commensurate nanolamelline, nanocolumns, nanospheres, and nanocipses can be synthesized to further extend the range of achievable properties. All of these structures are a result of kinetic limitations and require low growth temperatures combined with low-energy (less than the lattice atom displacement potential), very high flux, ion irradiation during deposition. This is easily achievable using tunable ion energies and magnetically-unbalanced magnetrons. Property enhancements include increases in oxidation resistance, diffusion barrier lifetime, and hardness by factors from 50% to 3 orders of magnitude. The range of colors available for decorative coatings is greatly expanded and new applications such as biologically active optical probes and selective sieves for biomolecules become possible. Finally, we extend these ideas to low-dimensional nanostructures such as perfectly periodic 1D wires and 2D domes, arcs, pyramids, and teardrops.

10:00 AM KK8.4
Physical self-assemble and nano-patterning. Tob-Ming Lu; Physics Department, RPI, Troy, New York.

The morphology of thin film growth front formed by a physical vapor deposition technique is controlled by many factors including: surface diffusion, sticking coefficient, and shadowing. Instabilities of growth can occur if the shadowing is more dominant compared to other surface effects and can lead to many diverse physically self-assembled 3D quasi-periodic nanostructures. In this talk I will discuss the fundamental mechanisms and growth mechanisms of isolated islands due to the shadowing effect in an oblique angle deposition configuration, and the time evolution of diverse 3D nanostructures including rods, beams, and spiral springs, and their microstructures that are grown on these isolated islands. I will present novel deposition methods that can control the width and geometry of these 3D nanostructures on a templated substrate with periodic seeds through a variety of substrate rotation and tilt schemes. *Work supported by NSF.

10:30 AM KK8.5
Chemistry-Mediated 2D-3D Transition of In Thin Films. H. L. Wei1, Hanchen Huang, Chung Ho Woo2, X. X. Zhang3 and L. G. Zhou2; 1Electronic and Information Engineering, Hong Kong Polytechnic University, Hong Kong, China; 2Mechanical Engineering, Rensselaer Polytechnic Institute, Troy, New York; 3Physics, Hong Kong University of Science and Technology, Hong Kong, China.

This talk reports a mechanism of chemistry-mediated 2D-3D transition during In thin film deposition, and the corresponding evolution of metallic nanolamellae. Using magnetron sputtering technique, we deposit In on Au substrate. Despite the fact that In wets on Au, In islands prevail over the uniform film soon after the deposition starts. The 2D-3D transition is found to be a result of the formation of AuIn on the Au substrate. The alloy formation leads to non-wetting of In, thereby the high mobility of In atoms and In clusters, and eventually well-separately In islands. The structures of In and Au are characterized by scanning electron microscopy, transmission electron microscopy, and electron diffraction.

10:45 AM KK8.6
Large Scale Engineered Nanostructured Surfaces by Reactive Ion Etching with Kinetically Self-Assembled Non-continuous Metal Films as Etching Mask. Wei Wei, Mark Bachman and Guan-Pyng Li; UC Irvine, Irvine, California.

The fabrication of nanostructured surfaces in different materials is of
Suppression of Reaction between Si Substrate and Obliquely Deposited Fe Atoms. Shinji Jomori, Motofumi Suzuki, Kohei Kimura, Kazuaki Kishimoto, Wataru Maekita, Kaoru Nakajima and Kenji Kinura, Department of Engineering Physics and Mechanics, Kyoto University, Kyoto, Kyoto, Japan.

Recent thin films with highly controlled columns such as helix and zigzag have been developed. Furthermore, integration of these morphologies has been achieved. These 'hypercolumnar' structures are engineered by oblique deposition (OD), in which the deposition angle and/or the azimuthal direction are varied dynamically during the deposition, in contrast to ordinary oblique columnar structures which are generated on the stationary substrate. Not only electronic and optical but also chemical, mechanical, and biological applications of thin films with hypercolumnar structures are expected. In particular, high porosity and the large surface area of hypercolumnar structures seem suitable for the chemical applications.

In this work, we report on the relationship between the hypercolumnar morphology and photocatalytic properties of obliquely deposited TiO₂ thin films. The hypercolumnar titanium oxide films were deposited on a glass substrate at room temperature in an electron-beam (EB) evaporation system. In order to tailor the morphology of the hypercolumnar titanium oxide films, we explored by controlling the etching process, i.e. isotropic etching or unisotropic etching, through varying etching conditions, such as RF power, chamber pressure, and gas ratio. Detailed study on the characterization of non-continuous Ag films was also carried out to verify the effects of the initial film thickness and the annealing rates on the subsequent Ag islands morphology, size, and interstitial voids size. Preliminary results regarding the dependence of the nanostructure on the annealing temperature towards an increased contact angle of hydrophobic surfaces is also shown.

11:00 AM KK8.7

The growth of iron silicides on a Si substrate has long been studied in the last two decades both from scientific and technological points of view, since the iron silicides have diverse physical properties depending on their crystal structures and compositions.

Unfortunately, crystallographic and morphological structures have not been successfully controlled. For example, it is known that Fe and Si are highly reactive with each other and form islands when Fe is deposited from the normal direction at high temperature. Some of these islands have faceted and specific height. By using oblique deposition technique, where the self-shadowing effect is dominant, the higher species among the islands will be grown selectively. In addition, it will be interesting for the case of Fe and Si system that not only the self-shadowing but also the surface reaction and diffusion play important roles in the formation of nanopatterns. In this work, we have investigated the effect of the deposition angle on the reactive growth of Fe-Si on the Si substrate. Iron was deposited intermitently up to about 10 ML (on a Si(001) substrate kept at 470 °C under the pressure less than 1.0×10⁻⁸ Torr. The deposition angle was set at 85° measured from surface normal. Between each deposition, the samples were analyzed by the high resolution Rutherford backscattering spectroscopy (HRBSS). In this paper, we measured random and <111> channeling spectra in order to estimate the depth profile of the composition and the number of (unshadowed) Si atoms displaced from their own lattice positions. Remarkably, the number of displaced Si atoms was almost constant and independent of the amount of Fe deposition, although it increases with twice of the number of deposited Fe atoms when Fe is deposited from surface normal. In addition, we confirmed the existence of islands (±10 nm height), which were small in number, by atomic force microscopy (AFM). This means that the reaction between Si and Fe is significantly suppressed and that the Fe rich nano-islands grew selectively. Therefore, by controlling the deposition geometry for the reactive systems such as Fe-Si, novel self-organization growth will be realized. This work was supported by The 21st Century COE Program "Center of Excellence for Research and Education on Complex Functional Mechanical Systems" and by The Mazda Foundation’s Research Grant.

11:30 AM KK8.9
In situ TEM Study of Three-Dimensional Patterning of Voids in Electron Irradiated CoF₂. Tianhua Ding, Shu Zhu and Lumin Wang, 1,2 Department of Nuclear Engineering and Radiological Sciences, Univ. of Michigan, Ann Arbor, Michigan; 2 Department of Materials Science and Engineering, Univ. of Michigan, Ann Arbor, Michigan.

It has been known that defect superlattices may form in electron irradiated CoF₂. However, the nature of the defects on the superlattice point (i.e., Ca colloids or voids) has not been certain and the defect patterning mechanism has not been fully understood. In this study, both natural and synthetic CoF₂ were irradiated under 200 keV electron beam with in situ TEM observation of the dynamic process of defect patterning. Through-focus bright field images from three zone axes (110), (110) and (111) demonstrated that the superlattice structure is simple cubic. Energy-filtered transmission electron microscopy (EFTEM), high-angle annular dark-field (HAADF) STEM imaging, as well as thickness mapping, were used to characterize the void nature of the superlattice formed under electron irradiation. The superlattice reached steady state after an electron dose of 6×10¹⁶ e⁻/cm² with void radius about 5-6 nm. Videos recorded during the in-situ observation reveal the patterning process of the void superlattice. Coalescence was prevalent at the initial stages. Migration and preferential growth were dominant at the final stages of the superlattice formation. At a dose higher than 1×10²⁷ e⁻/cm² the superlattice structure was destroyed. These critical values of void superlattice formation and deformation seem to be independent of dose rate. The kinetic rate equations can describe the initial growth of the voids, and the anisotropic diffusivity of the vacancies explains the superlattice formation.

11:45 AM KK8.10
Surface Nanopatterning via Dendritic Oxide Growth. Guangjun Zhou, Liang Wang, Pete M. Baldo and Jeffery A. Eastman; 1 Materials Science Division, Argonne National Laboratory, Argonne, Illinois; 2 Materials Science and Engineering Department, University of Pittsburgh, Pittsburgh.
Reaction-diffusion systems have attracted intensive attention to create a diversity of patterns due to the existence of the long-range effect of diffusion and purely local reaction interactions in these systems. In this work we present an oxidation-reaction-diffusion strategy for forming oxide nanopatterns involving preferential oxidation of metal alloys containing noble elements. Elements such as Au, Pt, Ag, etc., do not form stable oxides under normal conditions, while reactive elements, such as Cu, Ni, Fe, etc., form stable oxides. Specifically, the oxidation of Cu (100) alloys was visualized by an in situ environmental ultra high vacuum (UHV) transmission electron microscope (TEM). The oxidation of the CuxAu1-x alloy proceeds by an island growth mode, i.e., oxidation occurs at the island perimeters, followed by a transition to a dendritic growth mode, where the sharp corners of the island are the preferential directions for the dendrite development. The critical island size for the transition from a square shape to the dendritic growth and the branch size of the dendritic islands are determined. The growth of Cu oxide islands is accompanied by the rejection of Au atoms from the oxide, leading to a Au-rich zone in the alloy at the metal-oxide interface. Our in situ TEM observations indicate this gold-rich zone has a non-uniform distribution and the zone near the flat edges of the island is more Au-rich than the sharp corners, which causes preferential dendritic growth at the island corners. These experimental observations are consistent with a kinetics analysis based on the 2D Fick's diffusion equations.

SESSION KK9: Laser Patterning of Surfaces
Chair: Kalman Vasques
Thursday Afternoon, December 2, 2004
Room 306 (Hynes)

1:30 PM KK9.1
Directed Assembly of Thin Film Nanostructures Under Laser Induced Rapid Spatio-Temporal Surface Modulations. Chi Zhang1, Wei Zhang1, Adam Bauer2 and Kamal Kalyanaraman1,2;
1Physics, Washington University in St. Louis, St. Louis, Missouri; 2Center for Materials Innovation, Washington University, St. Louis, Missouri.

Thin film nucleation and growth is a self-assembly process where the initial stages naturally consist of nanometer sized clusters. Typically, on defect-free isotropic surfaces, these nanoclusters are a result of random walk surface diffusion and binding. The coupled time and length scale in nucleation, as determined by nucleation rate and saturated cluster density, can be determined by growth rate and substrate temperatures. So, appropriate surface temperature modulations should induce morphology changes. In recently published work, we presented novel studies showing that the application of a rapid spatio-temporal surface modulation in situ with film deposition directs the assembly of periodic structures. Evidence shows that the assembly is a result of anisotropic diffusion flux, reevaporation and locally modified nucleation. The experiment consists of physical vapor deposition (PVD) of CuO or TiOx with pulsed laser interference irradiation of a Si(100) surface. Deposition of CuO, Ag or TiOx results in periodic arrays of dot or line-like features whose periodicity and spacing resemble the interference patterns. The size scale of the resulting dot or line-like features can be varied from the nanometer to micron scale by deposition parameters. The rapid spatio-temporal modulation comes from the fringe spacing (200 to 2000 nm) and short laser pulse width (9 ns). In this work, we present fundamental experimental studies of the influence of these rapid surface modulations on film nucleation that leads to directed assembly of nanostructures. This experimental approach offers a simple way to assembly large-area arrays in a parallel process and is likely to lead to a cost effective manufacturing process.

2:15 PM KK9.4
Self-Organized Formation of Patterns on Tin Irradiated with Femtosecond Laser. Wei Zhou1, Hongyu Zheng2,3; 1School of Mechanical & Production Engineering, Nanyang Technological University, Singapore, Singapore; 2Singapore Institute of Manufacturing Technology, Singapore, Singapore

We used a Ti:Sapphire femtosecond laser (775 nm wavelength, 150 fs pulse duration, 1000 kHz repetition rate, intensity of above 1012 W/cm2) to irradiate a polished surface of polycrystalline tin in an ambient environment and observed a well-developed surface modulation with a period of 1.4-2.8 micron. We further showed that the ripple orientation was influenced by the crystallographic orientation of the tin surface, in contrast to usual observation that the ripples are perpendicular to the beam polarization. The classical model on laser-induced rippling predicts the dependence of the ripple period on the laser wavelength. In contrast, our experiments demonstrate that the ripple period increases logarithmically with fluence for the fixed laser wavelength used. The evolution of ripples indicates that the surface periodic patterns are not periodic due to direct writing in the ablation process and they are likely to be due to some spontaneous relaxation process on the highly non-equilibrium surface after the ions generated in the multi photon ionization process. Surface ablation species are a result of the fast ionization of metallic tin and the resulting multi-photon ionization of the substrate.

2:30 PM KK9.5
Laser Ablation of Carbon Films Used for Transfer of Electronic Devices. Yoshiyuki Samishima and Nobuyuki Andoh;
Pulsed laser-induced ablation of diamond-like carbon (DLC) films was applied to transfer thin film structures to foreign substrates. Because DLC films have a hard and flat surface, thin film layered structure such as thin film transistor circuits can be formed on the DLC films. DLC films also have high optical absorption coefficients $10^4 - 10^5$ cm$^{-1}$ from visible to ultraviolet. Moreover there is a high stress between DLC films and substrates. These properties mean that DLC is a suitable material for laser ablation and transfer of transfer thin film structures to foreign substrates. In the experiment, DLC films with a 200-nm thickness were formed on quartz glass substrates at room temperature by a sputtering method. 100-nm-thick aluminum layers were subsequently formed on the DLC films by a thermal evaporation method. The aluminum surface was coated with a paste. The samples were then pasted to plastic substrates. 35-ns-pulsed XeCl excimer laser was irradiated to DLC layers through the quartz substrates at room temperature in air atmosphere. When the laser energy density was 400 mJ/cm$^2$, the DLC layers were completely ablated and removed from the quartz substrates. No carbon remained on the substrates. Aluminum layers were also removed from the quartz substrates with no damage. The aluminum layers were successfully transferred to the plastic substrates. Dynamics of laser ablation of DLC films is discussed using transient conductance and transient reflectivity measurements. We will also demonstrate transfer of electronic device such as thin film circuits to foreign substrates using the present method.