Tuesday Afternoon, November 5, 2002

Nanometer Structures
Room: C-207 - Session NS+EL-TuA

Quantum Dots
Moderator: J. Nogami, Michigan State University

2:00pm **NS+EL-TuA1 Peter Mark Memorial Award Address: Mechanisms of Semiconductor Nanostructure Formation**, ***R.S. Goldman****, University of Michigan **INVITED**

Recently, semiconductor nanostructures have shown significant promise for a wide range of electronic, optoelectronic, and magnetic applications. In this talk, I will discuss the formation mechanisms of a variety of semiconductor nanostructures, including phase separation-induced alloy nanostructures and strain-induced self-assembled quantum dots. I will show how we have used data from cross-sectional scanning tunneling microscopy, in conjunction with x-ray reciprocal space maps, to develop new models for self-ordering of InAs/GaAs quantum dot superlattices[1] and spontaneous lateral phase separation in InAlAs alloys.[2] I will also describe our recent investigations of the role of elastic anisotropy in semiconductor nanopatterning, towards the formation of three-dimensional quantum dot crystals. These mechanisms are likely to be applicable to a wide range of heteroepitaxial semiconductor nanostructures.

This work was supported in part by the National Science Foundation (CAREER Award and Nanoscale Exploratory Research Program) and the Army Research Office (MURI Program)

[1] B. Lita, R.S. Goldman, et al, Appl. Phys. Lett. 75, 2797 (1999); Surface Review and Letters 7, 539 (2000).

[2] B. Shin, A. Lin, K. Lappo, R.S. Goldman, et al Appl. Phys. Lett . 80, 3292 (2002).

2:40pm **NS+EL-TuA3 Strain Effects and Inter-Dot Coupling in Self-Assembled Quantum Dot Arrays**, ***H.T. Johnson***, *R. Bose*, University of Illinois at Urbana-Champaign, *B.B. Goldberg*, Boston University, *H.D. Robinson*, University of California at Los Angeles **INVITED**

A computational model is used to simulate optical properties of self-assembled InAlAs/AlGaAs quantum dot arrays. Array sections containing up to 30 dots of varying size, shape, and spacing are considered. Comparisons are made to experimental results for arrays characterized using near-field scanning optical microscopy (NSOM). The experimental and computational studies both measure emission/absorption spectra with energy resolution that shows the effects of individual dots in the array. In the computational approach, the optical properties are computed from the spectrum of electron and hole states found for the ensemble. The energies and wave functions in the spectrum are first computed using a strain-modified k-p Hamiltonian approach; the spectrum includes confined electron and hole states associated with individual dots in addition to some delocalized states associated with coupled dots as well as the wetting layer. By modeling the entire ensemble of dots simultaneously, it is possible to consider effects related to long range field interactions between dots, such as linear elastic fields and extended quantum mechanical states. Two key results are of interest. First, it is found that even minor contact between the optical fiber tip and the sample surface leads to indentation strain large enough to substantially shift emission wavelengths of individual dots in the array. Second, extended states in groups of neighboring quantum dots lead to sharp, well-defined resonances in the emission spectra for the arrays. Results of the simulations clearly show these effects that are also observed in the experimental data.

3:20pm **NS+EL-TuA5 Growth of Ge Quantum Dots on Si(100) Without a Wetting Layer** [1], *K. Yoo*, Oak Ridge National Laboratory, *F. Flack*, University of Wisconsin - Madison, *H.H. Weitering*, Oak Ridge National Laboratory, *M.G. Lagally*, University of Wisconsin - Madison, *Z. Zhang*, ***J.F. Wendelken***, Oak Ridge National Laboratory

When Ge atoms are deposited directly onto a Si(100) substrate, the growth follows the Stranski-Krastanov growth mode in which three-dimensional Ge islands, or quantum dots (QDs), are formed on top of three monolayer thick wetting layers. For many optical and electronic device applications, Ge QDs without the wetting layer may be highly preferred. Using a buffer-layer assisted growth approach,[2] we have achieved the formation of Ge QDs on Si(l00) without a wetting layer. These QDs are shown to possess a narrow size distribution and are also substantially smaller than the QD hut clusters that are formed with the normal SK growth mode. Using the buffer layer approach, Ge QDs have been grown in a single layer and in multiple layers with silicon spacer layers as has been done with multilayers of conventionally grown Ge hut clusters. Due to the fact that growth in the buffer layer approach is well isolated from any stress effects associated with a preceding layer of QDs, it is not expected that the QDs in separate layers will exhibit any layer to layer alignment effects as observed for SK growth. However, the smaller size of the QDs obtained with the buffer layer approach may be expected to exhibit stronger quantum size effects. Initial tests with samples prepared by this approach show a strong photoluminescence signals in the IR that exhibit striking differences from PL results[3] obtained from Ge QDs grown by conventional means with a wetting layer.

[1] Work supported in part by the U. S. DOE at Oak Ridge National Laboratory, managed by UT-Battelle, LLC under Contract DE-AC05-00OR22725, and in part by the NSF at the University of Wisconsin through the MRSEC program.

[2] J. H. Weaver and G.D. Waddill, Science 251, 1444 (1991).

[3] M.W. Dashiell, U. Denker, and O.G. Schmidt, Appl. Phys. Lett. 79, 2262 (2001).

3:40pm **NS+EL-TuA6 Spontaneous Generation of Free-Standing Ge Quantum Dots on Silicon-on-Insulator**, ***E.A. Sutter***, *P.W. Sutter, P. Zahl*, Colorado School of Mines

The growth of heteroepitaxial materials on engineered composite substrates such as silicon-on-insulator (SOI) opens a new route for controlling the structural and electronic properties of materials at the nanoscale. Local lattice strain induced by Ge quantum dots grown coherently on SOI - a composite of an ultrathin monocrystalline Si template supported by amorphous SiO_2 on a conventional Si wafer, causes significant local distortion in the Si template and can be used as a tool for nanoscale band structure engineering of the Si substrate. The Ge islands themselves form on SOI initially as huts and then transform into domes, similar to the sequence of epitaxially constrained shapes they assume on bulk Si (100). While the shape sequence of epitaxial Ge islands on bulk Si ends here, we observe further dramatic morphological changes on ultrathin SOI: a spontaneous transformation to free-standing Ge islands accompanied by a breakup of the thin Si slab. We use a combination of atomic force microscopy (AFM) and transmission electron microscopy (TEM) to document the sequence of shape transformations of Ge islands on SOI. We investigate in detail the island shape evolution and redistribution of the substrate material between the islands both before and after the breakup of the ultrathin Si slab of the SOI substrate.

4:00pm **NS+EL-TuA7 Production and Structure of Gas-phase Prepared Germanium Nanocrystals**, ***C. Bostedt***, *T. van Buuren, T.M. Willey, J. Plitzko*, Lawrence Livermore National Laboratory, *T. Moller*, Hasylab at DESY, Germany, *L.J. Terminello*, Lawrence Livermore National Laboratory

Clusters and nanocrystals represent a new class of materials, which exhibits promising novel properties. Germanium nanoparticles are particularly interesting, as the cubic as well as the tetragonal crystal phases have been reported for particle sizes below 5 nm, depending on the production method. We have developed a gas-aggregation based production method for germanium nanoclusters, with which nanocrystals in the bulk-like cubic phase from 1 to 10 nm in size can be produced. The clusters are condensed out of supersaturated Germanium-vapor, which is cooled down in a He-atmosphere and are subsequently deposited on a variety of substrates. The particle sizes and phases have been well characterized by transmission electron microscopy (TEM), x-ray diffraction (XRD) and atomic force microscopy (AFM). The crystal phase - production method relationship is discussed. Only little is know about the surface structure of nanocrystals. Information about the surface structure is difficult to obtain. No distinct particle boundaries can be imaged with TEM. Photoemission spectroscopy (PES) has been shown to be a powerful tool to investigate bulk-crystal semiconductor surfaces. PES experiments have been performed on Ge nanocrystal films and a disordered surface shell around a crystalline core is deduced for the nanoparticles. .

C. Bostedt acknowledges a fellowship from the German Academic Exchange Service DAAD in the HSP-III program, N. Franco from the Spanish Education and Culture Office. The work is supported by the US-DOE, BES Material Sciences under contract W-7405-ENG-48, LLNL.

4:20pm **NS+EL-TuA8 Surface Passivation Effects of Deposited Ge-Nanocrystal Films Probed with Synchrotron Radiation**, *C. Bostedt*, ***T. van Buuren***, *T.M. Willey*, Lawrence Livermore National Laboratory, *T. Moller*, Hasylab at DESY, Germany, *L.J. Terminello*, Lawrence Livermore National Laboratory

Clusters and nanocrystals represent a new class of materials that exhibit promising novel properties. The production of these nanostructures in the gas phase gives control over not only the size of the nanoparticles, but also over surface passivation - often not possible in other growth modes. The clusters are condensed out of supersaturated Germanium-vapor which is

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cooled down in a He-atmosphere and are subsequently deposited on a variety of substrates. Their surfaces can be subsequently passivated with different materials evaporated into the vacuum chamber. This approach allows us to probe in a controlled and dynamic fashion the effect of surface passivation on nanocluster properties. X-ray absorption spectroscopy (XAS) and photoemission (PES) were performed on thin films of Germanium (Ge) clusters. Clean Ge nanocrystal films are found to exhibit much stronger quantum confinement effects at the band edges than similar Si particle films. These findings are compared to recent electronic structure calculations. For passivated nanocluster films we find that the passivating agent strongly alters the electronic structure of the clusters. In general the absorption edge shifts to significantly higher energies compared to cluster films without surface passivation. These results will be discussed in terms of a reduction of the cluster-cluster interactions.

C. Bostedt acknowledges a fellowship from the German Academic Exchange Service DAAD in the HSP-III program. The work is supported by the US-DOE, BES Material Sciences under contract W-7405-ENG-48, LLNL.

4:40pm **NS+EL-TuA9 Growth and Properties of Si Compatible Nanostructures: Si Quantum Dots Grown on CaF_2/Si Films**, ***A. Klust***, *A.A. Bostwick, T. Ohta, Q. Yu, M.A. Olmstead*, University of Washington

Si/CaF_2 is a promising candidate for epitaxial semiconductor/insulator heterostructures for optoelectronic applications because of the low lattice mismatch (0.5%) and large band gap difference (12.4 eV for CaF_2 vs. 1.1 eV for Si). In addition, the strongly dissimilar ionic/covalent bonding character in the system CaF_2/Si allows it to serves as a model system both to study heteroepitaxy of two dissimilar materials and to study the influence of bonding character and electronic structure on scanning probe microscopy. Here, we present an investigation of ultra-thin (1-3 molecular layers) CaF_2 films and Si quantum dots grown on these films using both non-contact atomic force microscopy (ncAFM) and scanning tunneling microscopy (STM). On the one hand, the extremely large band-gap of CaF_2 makes STM measurements difficult; stable imaging is not possible for films thicker than 3-4 molecular layers. On the other hand, STM gives complementary information to that obtained with ncAFM. For instance, scanning tunneling spectroscopy is used to characterize the electronic properties of single Si QD. Furthermore, the contrast during STM imaging of the CaF_2 films depends strongly on the polarity of the bias voltage and the film thickness. Non-contact AFM is used to clarify this behavior to separate electronic and topographic contributions to the STM images. Atomically-resolved ncAFM images of the CaF/Si interface layer will be shown and compared with similar published data obtained from bulk CaF_2(111) crystals.[1] The atomic structure of the CaF/Si interface layer is practically identical to the surface structure of bulk CaF_2(111), while the electronic structure differs. We discuss the influence of the different electronic structure on atomic resolution ncAFM.

[1] A. S. Foster, C. Barth, A. L. Shluger, and M. Reichling, Phys. Rev. Lett. 86 (2001) 2373.

5:00pm **NS+EL-TuA10 Formation of Self-Assembled Copper-Oxide Nano-Dots on SrTiO3(100)**, ***I. Lyubinetsky***, Pacific Northwest National Laboratory

In addition to offering a wide range electrical, optical and magnetic properties, oxide nanostructures can be stable in a range of environments without needing to add protective layers. In this work, we have observed the formation of self-assembled oxidized-copper nanodots on the SrTiO3(100) substrate using oxygen plasma assisted molecular beam epitaxy. The composition and structure were examined by x-ray photoelectron spectroscopy (XPS), Auger electron spectroscopy, x-ray diffraction, and scanning probe microscopy in a wide range of growth parameters (temperature, oxygen pressure, and Cu flux). Under different growth conditions different shapes and/or composition have been found: truncated dots, square pyramids, and multifaceted domes with composition to be Cu2O or containing Cu metal also. Since nanostructure composition changes with temperature, observation made by interrupting growth and cooling may not be adequate. Thus, in addition to scanning probe measurements in ambient conditions, XPS spectra have been acquired at elevated temperatures in step-by-step mode at different stages of nanodots formation. Observed correlations between structural and composition changes will be discussed. Understanding of the evolution of the dot shape, size and composition would allow us to optimize formation conditions to synthesize cooper oxide nanodots with desirable parameters.[1]

[1] This work has been conducted as part of the PNNL Nanoscience and Nanotechnology Initiative supported by the Office of Biological and Environmental Research, U. S. Department of Energy.

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