Quantum Dots

The continued miniaturization of electronic and optoelectronic devices is leading us closer to the need for nanoscale structural assemblies to perform the function of computer, laser, data storage, sensor and satellite communication. As the size of the structural feature (building block) in a device is reduced to the nanometer scale, its electronic, optical, electrical, and magnetic properties begin to differ drastically from its bulk counterparts, displaying a strong size and shape dependence due to quantum size effects arising from the reduced dimensions and surface/interface effects arising from the increased surface-to-volume ratio. The new properties, in particular those arising from quantum size effects, lead to a wide range of potential applications in electronic, optoelectronic, and magnetic devices. Quantum well devices, in which one of the dimensions of the underlying materials is reduced to the nanometer range so that carriers (electrons and/or holes) are confined in this dimension, are already widespread. Recent effort has focused on designing and fabricating devices using quantum wires and quantum dots, respectively the one- and zero-dimensional analogs of a quantum well. Just as uniform thickness required for quantum wells, a high degree of uniformity in size, shape, and spacing are often required for quantum wires (QWs) and quantum dots (QDs) in real applications. However, fabrication of QWs and QDs with desirable uniformity remains, so far, a challenging problem. This is because our fundamental understanding of the physical and dynamical processes at the nanometer scale is still limited and it is very difficult to design and apply fabrication and characterization tools at the nanometer scale.

Much recent effort has been devoted to the controlled fabrication of nanostructures to achieve a high degree of order and uniformity. In general, two different routes have been taken: one is the so-called top-down approach by direct surface patterning or lithography with nanometer resolution, the other is the bottom-up approach by taking advantage of self-assembly and/or self-organization of surface structures occurring naturally at the nanometer scale. Surface structures with desirable size and shape can be made *artificially* by surface patterning and lithography, either patterning the adsorbates or growing on an *a priori* patterned substrate. Alternatively, nanostructures, in particular QWs and QDs, can form *naturally* during the growth of thin films by self-assembly and/or self-organization. For example, coherent three-dimensional (3D) islands (free of dislocations) form spontaneously in strained thin films [F. Liu, et al, *Surf. Sci.* 386, 169 (1997).]. These 3D islands (clusters) form, in general, with crystalline perfection and nanometer dimensions, lending themselves naturally to use as QDs. The self-assembly and self-organization approach has shown great promise for device fabrication because it offers an economical parallel process with the added advantage of being compatible with the existing Si processing technology.

Self-assembly and self-organization processes are actually very common in nature. They lead to a variety of beautiful patterns in a wide range of length scales, from the cosmic array of galaxies to the atomic structure of crystals. The length scale of a self-assembled or self-organized pattern is governed by the typical range of interaction applied by the underlying forces. For example, galaxy arrays are governed by gravitational force acting at the celestial distance, while crystal structures are governed by chemical bonds acting at the angstrom (one tenth of nanometer) range. Therefore, for self-assembly and self-organization of nanostructures, forces acting at the nanometer

range have to be identified. One force that falls into the nanometer range is the elastic interaction in solid materials. Coincidentally, devices are often made of layered structures of thin films, in which elastic interaction can be a dominant force arising from the misfit strain that is inherently present due to the lattice mismatch between the different layers of materials. The study of morphological instability and ordering in strained thin films has, therefore, attracted considerable recent interest, because strain-induced self-assembly and self-organization provides an attractive "*natural*" route for nanofabrication, which we term it strain engineering of nanostructures.

Extensive experimental and theoretical work has been attempted in the last decade to stimulate, guide, and control the self-assembly and self-organization processes to improve the uniformity of QDs by manipulating the thermodynamics and kinetics of epitaxial growth. However, our understanding is still far from complete, especially there is a lack of quantitative understanding. So far, theoretical and computational studies have been largely limited at the qualitative level. The formation of QDs, as well as strained islands in general, is driven by relaxation of strain energy at the expense of increase of surface energy. The special feature of QDs is they are usually bound by faceted surfaces of selected orientations [such as (105) facets on Ge hut islands] that are often stabilized by strain. Thus, it will be very useful to be able to compare the energies between the QD surfaces (facets) and original substrate/film surfaces and to evaluate their strain dependence to better understand the physical origin of QD formation. These surface energies may further be used as input parameters for continuum modeling to quantitatively estimate the "critical" size for QD nucleation/formation and its strain dependence.

Surface stress tensors of QD surfaces are also useful quantities in helping understand its formation and stability. The QD (strained island) formation energy contains generally two major contributions: the strain relaxation energy proportional to QD volume and the cost of surface energy proportional to QD surface area. If only these two terms were present, there would be no stable QD size as the total energy continues to decrease with increasing size. However, if surface stress of the QD surface and of the substrate/film surface is different, there will be an additional term of elastic edge relaxation energy that may introduce a stable island size against coarsening. So far, no quantitative evaluation has been made on the significance of such edge term on QD stability.

In addition to thermodynamic parameters like surface energies and surface stresses, quantitative information of kinetic parameters, such as surface diffusion energy barriers, are needed for a more complete understanding of growth of QDs. Many questions remain to be answered. For example, how does surface diffusion depend on surface strain, as QDs are generally formed as strained islands? How does surface diffusion change as adatoms diffuse from the substrate or wetting layer surface to island surface? What's surface diffusion disparity between Ge adatom and Si adatom?

The objective of this NERSC supercomputing project of QD is to perform extensive first-principles calculations of surface energies, surface stresses, and surface diffusion barriers as well as their strain dependence of substrate, wetting layer, and QD surfaces, using Ge dots on Si(001) as a model system. By combining these calculations with continuum modeling, quantitative analyses of formation and stability of Ge and SiGe QDs on Si substrate will be possible for the first time.

Two types of Ge QDs form on the Si(001) surface, one called “hut” that forms at the early stage of growth having a smaller size is bounded by {105} facets, the other called “dome” that forms at the later stage of growth having a larger size is bounded by {113} and other high-index facets. Figure 1 shows a schematic view of Ge QD (hut) on Si(001). Calculation of surface energies and stress tensors of the QD surfaces, as well as calculations of surface diffusion barriers on the QD surfaces are computational rather demanding, because they are generally high-index surfaces that require a relatively larger unit cell. Also, they involve complex reconstructions that are difficult to determine. For example, it is until very recently that the correct surface reconstruction of the hut surface (the strained Ge(105) surface) has been finally determined, twelve years after the hut discovery. Furthermore, the calculated quantities are generally strain and/or thickness dependent, so series of calculations of different strain and/or different film thickness need to be performed.

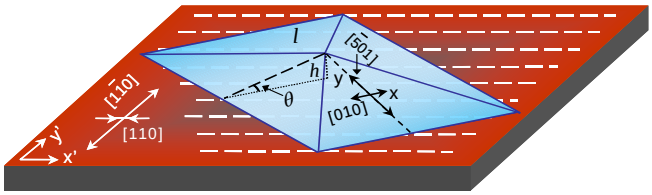

Fig. 1. Schematics of a square-based Ge hut on Si(001). Arrowed crosses indicate surface stresses.

The pseudopotential plane-wave total-energy method has been applied to carry out these large-scale calculations. The surfaces, such as Ge-covered Si(105) surface, are modeled using a supercell of slab including a sufficient thick vacuum layer. Usually, these calculations involve 100 to 200 atoms and ~100 plane-wave basis functions per atom. These make use of a MPI parallel computational code that exhibits good efficiency and scaling properties up to 32 nodes.

In the past year, two projects have been completed. In the first project, the surface energies and surface stresses of Ge/Si(001) and of Ge/Si(105) surfaces have been calculated: the former corresponds to the wetting layer surface and the latter corresponds to the Ge QD (hut island) surface. Based on these calculations, it is possible to predict, for the first time, the critical size for Ge QD nucleation/formation with an island base dimension between ~10 to 16 nm, which agrees very well with experiments. Also, a quantitative measure of the magnitude of elastic edge relaxation energy due to surface stress discontinuity at the island edge has been obtained, showing it to be too small to induce a stable island size against island coarsening, as has been proposed before as a possible mechanism for self-assembly of QDs. In the second project, the surface mobility difference between Si and Ge and its strain dependence has been calculated. An example of diffusion of Ge adatom on the QD Ge(105) surface is illustrated in Fig. 2. On a compressive surface, Ge diffusion is 10^2-10^3 times faster than Si; while on a tensile surface, they are comparable. Consequently, growth of a compressive SiGe film is rather different from that of a tensile film. Furthermore, the

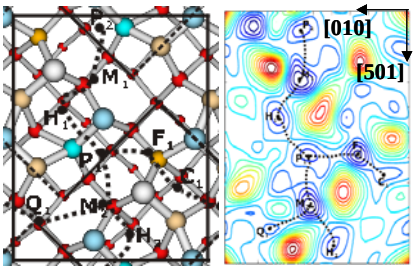

Fig. 3. (a) Top view of the unit cell of Ge(105)-(2x1) surface. Higher atoms are drawn by larger spheres. (b) Contour plot of Ge adatom potential energy surface. The letters and dashed lines indicate the main binding sites and the low-barrier diffusion paths.

diffusion disparity between Si and Ge is greatly enhanced on the strained Ge islands (QDs) compared to that on the Ge wetting layer, explaining the experimental observation of Ge enrichment in the QDs.

In the coming year, these large-scale first-principles calculations will be continued and extended to more complex systems, such as QDs of difference shapes (Ge domes), SiGe alloy QD surfaces and surfaces containing steps. These calculations require even larger supercells, and hence are much more computational demanding. This type of computation is very communication intensive and long running. It would benefit from a new computer system with much faster processor speed and also improved memory latencies and bandwidth so that it could make use of more than 32 nodes efficiently. Barring this, the calculation would need more memory per node and longer queue running times per job to allow the study of larger systems on 32 nodes.