The Evolution and Self Assembly of Quantum Dots

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Introduction

Nanostructures can be employed to yield devices with electronic properties that are different than those of the same materials in bulk. For example, many commercially available lasers and detectors have been manufactured using periodic layers of low- and high- band gap materials. Still more interesting electronic properties can be obtained by confining electrons in a nanometer-scale dot, or a zero dimensional system. The electronic density of states becomes delta-function like even though the domains consist of 10^5 to 10^6 atoms, in direct contrast to the continuous density of states found in the bulk materials. Such nanodomains, or ``quantum dots'' (QDs), can, in principle, be employed to produce novel electronic devices ranging from high-efficiency lasers and detectors to exotic single-electron transistors and cellular automata.

A promising approach to producing large arrays of QDs is to use the intrinsic self-assembly process that occurs spontaneously during heteroepitaxy. It has been observed that during the growth of a planar film deposited on a substrate, where the film and substrate have a different lattice parameter, the film surface can undergo a morphological instability wherein dislocation-free islands form. Moreover, these islands tend to self-organize into a periodic array and, under certain conditions, are immune to the coarsening process that would tend to broaden the size distribution. This self-organization process occurs over very large length scales allowing patterning and production of an enormous number of dots.

While these observations indicate that this self-organization process holds great promise, the understanding of the underlying physics and strongly nonlinear nature of the dynamics of the self-organization process makes it difficult at present to control this process to the extent required to produce QD devices. The ultimate goal of the effort is to develop an understanding of the QD formation process to the level that a first-principles design of QD nanostructures becomes a reality [1].

Progress

Ge and SiGe on Si(001) have been widely used as model systems for understanding hetero-epitaxy. A rich body of literature has showed that, in equilibrium, small islands are square pyramids, while larger islands develop a more complex multi-faceted shape after passing through a first-order shape transition [2]. Moreover for smaller sizes experiments show that the islands can from continuously from surface ripples [3].

We have developed a unified picture of the initial stages of SiGe island or QD formation on Si(001) [4]. We examined the equilibrium island shapes in the SiGe system. The island morphologies are determined by finding shapes that yield extrema in the sum of the elastic and surface energies if the system. The surface energy is taken to be anisotropic such that (105) facets are energy minima. We find that the behavior of SiGe islands on Si(001) can be consistently explained by one simple assumption --- that for strained SiGe, the surface-energy anisotropy allows all orientations near (001), with the first facet being (105). With this anisotropy, we predict that tiny ``prepyramidal'' islands form with no nucleation barrier. They are unfacetted, with low height/width ratio and indistinct edges. As islands increase in size, they undergo a transition in which (105) facets are introduced discontinuously at the steepest point on the island. This first-order shape transition is expected to drive anomalously abrupt coarsening and the rapid disappearance of any nearby prepyramids. Our experimental observations confirm the nature of the prepyramids and of the faceting transition, as well as the key assumption of the model.

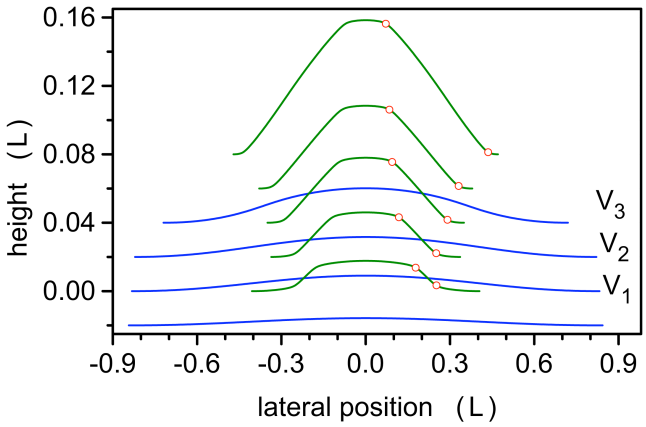

Figure 1. Equilibrium island shapes calculated for islands of various volumes. Shapes for different size V are offset vertically. When there are two (meta)stable shapes for the same V shapes are superposed and "facet" edges are indicated by the open circles. V_1 is the size of the smallest metastable "faceted" island, V_3 is the size of the largest metastable smooth island, both shapes are stable and have degenerate energy at V_2. The substrate is not shown, the island edges meet the substrate at zero contact angle

The above study examined island shapes from a strictly thermodynamic standpoint. However, the role of various *kinetic* mechanisms on the development of island size distributions remains less clear. It is clear theoretically, however, that there are potentially important consequences for island growth kinetics arising from strain dependencies in adatom binding and migration energies [5]. The adatom binding energy sets the equilibrium concentration of adatoms on the surface, which allows islands to communicate and hence coarsen. The migration energy is related to the rate at which atoms diffuse along the surface. Both of these energies can thus play an important role in setting the kinetics of island coarsening. However, there have been relatively few attempts to determine the magnitude of these effects in specific systems.

To this end we have determined the binding and migration energies in one of the most widely studied quantum dot forming systems, the SiGe system, using first principles calculations [6]. We determined the strain dependence of Ge adatom binding energies and migration energies for diffusion on both Si(001) and Ge(001) surfaces. For a growth temperature of 600 C these strain-dependencies give rise to a 16-fold increase in adatom density and a 5-fold decrease in adatom diffusivity in the region of compressive strain surrounding a Ge island with a characteristic size

of 10 nm. Within a simplified model of diffusion-limited growth, these strain dependencies are found to have the qualitative effect of accelerating the natural coarsening rate of larger islands. In contrast, the corresponding strain-dependences on the Si (001) surface are much less pronounced. This implies that kinetic effects are strongly system dependent. We are in the process of incorporating these effects into our models for the dynamics of quantum dot evolution.

Describing the dynamics of the development of arrays of quantum dots is a challenging problem since it is necessary to follow the evolution of large numbers of dots where the shape and location of the dots are unknown. Most calculations are thus limited to following the evolution of small numbers of dots. To overcome these difficulties, we have examined dot formation using a long-wave nonlinear evolution equation for the film surface. The dots evolve in response to the elastic stress generated by the mismatch between the film and substrate, and surface energy. The evolution of the system is by surface diffusion. The film is assumed to wet the substrate and thus we have included a wetting potential in the model [7].

We find that the presence of wetting gives rise to a finite wave number at the onset of the stress-driven morphological instability. Weakly nonlinear analysis near the threshold of instability shows that the bifurcation from a planar state can be supercritical. The symmetry of the evolution equation for the surface implies that these supercritical states must have hexagonal symmetry. We have determined the stability of these hexagonal states and find that for certain wetting potentials, these hexagonal states can be stable. These stable states can be in the form of hexagonal arrays of dots or pits, see Figure 2.

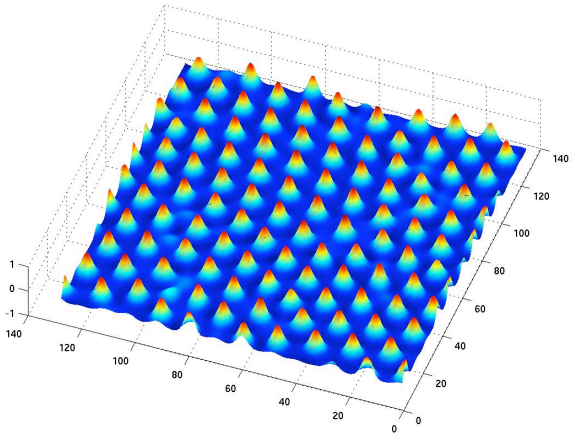

Figure 2 A stable array of dots produced by the evolution of the instability of a planar interface.

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