Quantum phenomena in nanostructures

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Abstract. Quantum phenomena in nanostructures, especially epitaxial structures, are utilized for the fabrication of quantum devices. Here we discuss self organized growth of epitaxial quantum structures on silicon and their electronic structures, such as electronic density of states, and electronic transport in such systems. We discuss nanodots and nanowires, or an equivalent of zero-dimensional (0D) and one-dimensional (1D) structures in more details, and in general structures in all dimensional cross-over regimes. Further arrangements of quantum dots (QDs) in particular structures offers to form quantum bits and quantum dot molecules (QDMs). This could lead to exploration of new physics and new quantum devices.

1. Introduction
Scientific research in the area of nanostructures and nanomaterials appeared in a big way in the late 1980’s. This predominantly consisted of materials synthesis by chemical methods. However, nanoscale and even atomic scale structures were being fabricated by the physical route since the advent of Surface Science in the mid 1960’s [1]. In Surface Science, clean surfaces of single crystal materials are prepared under ultrahigh vacuum condition; atomic structures of clean surfaces are invesigated; and then desired materials can be deposited on clean surfaces with atomic control. In the past few decades, progress in thin film crystal growth technology has made it possible to control the dimensions of materials structures with atomic scale precision. This has provided a method to fabricate desired nanoscale structures. There have been many interesting discoveries of new phenomena in the field of low dimensional quantum structures and consequent developments in an effort to achieve solid state devices operating at even higher speeds and consuming lower power, and exhibiting novel capabilities.

In 2001 National Research Council, USA, published a vision document “Physics in a new era: An overview” to elucidate scientific priorities and opportunities and identified six areas of grand challenges [2]. Two among them are (i) developing quantum technologies and (ii) creating new materials. Activities in these areas have been growing around the world. Molecular beam epitaxy (MBE) [3] has been an important materials growth technique that has contributed enormously to discoveries of quantum phenomena and technologies. As mentioned earlier, nanoscale structures can be grown via chemical route. These are usually an assembly of nanoparticles. This article will not deal with this kind of growth. Here we will deal with ordered or epitaxial growth of materials on clean single crystal substrates by MBE. While quantum well structures can be directly grown by MBE, quantum wire (QW) and quantum dot (QD) structures can be fabricated via lithography from a uniformly grown layer. On the other hand QD and QW structures can also be fabricated in self-organized growth by MBE [4]. Usually smaller structures, beyond
Figure 1: (a) A LEED pattern of a Si(111)-(7×7) surface (From Ref. 6). The bright spots would be the only spots if the surface were unreconstructed. There are six additional spots between two bright spots, making the unit cell basis vectors 1/7th in the reciprocal space. This means that in the real space the basis vectors are 7 times enlarged. Hence the name 7×7 reconstruction. (b) Three 7×7 unit cells in the real space, seen in an STM image (From Ref. 7), are marked

the lithographic limit, can be fabricated by self-organized growth. This article will confine to self-organized growth of nanostructures by MBE and mainly rely on the work carried out in the author’s laboratory.

2. Substrate cleaning and clean surfaces
In MBE growth, materials are grown on single crystal substrates. Preparation and preservation of clean surfaces of the substrate require an ultrahigh vacuum (UHV) environment in order to avoid contamination. This is obtained in a UHV chamber using various pumps. The typical pressure in the UHV chamber is 10^{-11} mbar. A surface usually has different atomic arrangements compared to the bulk material. This is a consequence of missing atoms on one side of the surface. On semiconductor surfaces, the surface unit cell deviates considerably from the ideal surface. The surface is said to be reconstructed. On a Si(111) surface the basis vectors of the surface unit cell is 7 times enlarged compared to the ideal Si(111) surface. This surface is said to be (7×7) reconstructed and written as Si(111)-(7×7). This can be observed in reciprocal space in low energy electron diffraction (LEED) and reflection high energy electron diffraction (RHEED) or in direct space by scanning tunneling microscopy (STM). The atomic structure of a Si(111)-(7×7) surface is so complex that it took almost 25 years of intensive experimental and theoretical research to determine its structure. The accepted structure of the Si(111)-(7×7) surface is called the dimer adatom stacking-fault (DAS) model [5]. The LEED pattern [6] and an STM image [7] of this Si(111)-(7×7) surface are shown in figure 1. Reconstructions of Si(100) and Si(110) surfaces are different. Usually a Si(100) surface has a (2×1) reconstruction [7] and Si(110) surface has a (16×2) reconstruction [8]. In materials growth by MBE, an atomic (or molecular) beam of the material to be deposited is prepared in the UHV chamber and atom by atom deposition takes place on the clean surface of the substrate.
3. Self-organized epitaxial structures

In epitaxial growth, a crystallographically ordered material grows on a single crystalline substrate. In homoepitaxy, the grown material is the same as the substrate material, and it is a trivial case. The more interesting case is heteroepitaxy, where the grown material is different from that of the substrate. Consequently, their crystalline unit cells are different. So the grown material either has to shrink or extend laterally in order to match the substrate unit cell, as schematically shown in figure 2. Thus the unit cells of the grown material either have to extend or contract in the direction normal to the surface. That means that the grown material is strained. The competition between the surface/interface energy and the strain energy leads to three different growth modes, namely, (i) layer by layer (or Frank van der Merwe), (ii) layer plus island (or Stranski-Krastanov) and (iii) island (or Volmer-Weber) growth mode [9].

It is seen from the figure that self-organized islands grow in Stranski-Krastanov and Volmer-Weber growth [9]. These growth modes can produce self-organized QD and QW structures, without the need of lithography.

4. Ge nanostructures on Si surfaces

Let us discuss Ge growth on clean Si(100) surfaces. Ge has lower surface free energy compared to Si. So Ge begins to grow layer by layer. Both Ge and Si have diamond structure with cubic unit cell. However, Ge lattice constant is 4% larger compared to Si. That is why Ge is strained and the growth proceeds via layer plus island or Stranski-Krastanov mode. MBE growth facility
Figure 3: (a) The RHEED pattern from a Si(100)-(2×1) reconstructed surface. (b) RHEED pattern after deposition of about two atomic layers of Ge. (c) RHEED pattern following about an equivalent of ten atomic layers of Ge. (d) STM image of the sample in (c). (e) XTEM image showing a single Ge island on Si. (From Ref. 7).

usually has also a RHEED facility. RHEED pattern can be recorded during material (Ge) deposition. From the evolution of the RHEED pattern, growth mode can be identified [7].

The RHEED pattern in figure 3(a) shows the Si(100)-(2×1) reconstructed surface. After deposition of a number of Ge atoms equivalent to two atomic layers, the RHEED pattern evolves into that in figure 3(b). The streak pattern is indicative of growth of uniform layers. After deposition of an equivalent of ten atomic layers the RHEED pattern evolves into that in figure 3(c). The spot pattern indicates that three-dimensional structures or islands have grown. The sample was then transferred into the STM chamber to take an STM image, shown in figure 3(d), which clearly shows island formation. In this case three atomic layers grow as uniform layer and additional deposited Ge grows as islands. So the growth mode is Straski-Krastanov. A cross-sectional transmission electron microscopy (XTEM) image (figure 3(e)) shows a Ge island on Si.

The shapes of the Ge islands can be controlled by modifying the growth. That offers the possibility of tuning the energy levels of quantum dots. This can be done by initially growing a thin oxide on the clean surface, and then depositing Ge on this oxide. We provide such an example using a Si(111) surface [10].

When Ge is deposited on a hot (∼550°C) oxidized Si substrate the following reaction takes place

$$SiO_2 + Ge (ad) \rightarrow SiO (g) + GeO (g)$$ (1)

When Ge adatom lands on the hot SiO_2 solid (s) substrate it reacts and produces suboxides of Si (SiO) and Ge (GeO) which are gaseous. So oxide is removed from these regions and clean Si is exposed through nanoscale holes. Further deposition of Ge leads to epitaxial growth of Ge quantum dots on Si. This process is schematically shown in figure 4. Experimental results are shown in figure 5. The growth has taken place at a substrate temperature of 550°C. The neck contact area can be controlled by choosing the appropriate substrate temperature. The electronic energy levels in the QD can be tuned by controlling the neck area. As seen above, the Ge QDs have grown at random locations on the substrate surface. For certain applications...
Figure 4: Schematic of nearly spherical epitaxial Ge QD growth on Si(111): (a) a thin oxide layer (∼2 nm) on Si; (b) a nanoscale hole formation in the oxide layer; (c) a Ge QD growth through the nanoscale hole in contact with underlying Si.

Figure 5: (a) A STM image showing Ge QDs. (b) An XTEM image showing the side view of Ge QDs in contact with Si. (c) a single Ge QD. (d) Schematic of a single QD in cross-sectional view. (From Ref. 10).

and exploring new physics, an ordered arrangement of QDs is necessary. In search of fabrication of quantum computers, there are various approaches for the fabrication of quantum bit or qbit. A classical bit (0 or 1) is obtained by the off and on state of a transistor. In quantum cellular automata (QCA) approach, there is no concept of transistors, and the bits 0 and 1 can be obtained from an array of four QDs [11,12]. Thus, fabricating ordered arrangements of QDs is important. This can be done by prepatterning the substrate before the growth of QDs [13]. The prepatterning can be done by exposing the substrate to nanometer-sized spots of an electron or ion beam. Some examples of that are shown in figure 6.

It may be noted that a QD is a super atom. A collection of a few interacting quantum dots forms a quantum dot molecule (QDM) [14]. For example, the group of five QDs seen in figure 6(c) may behave like a five-atom molecule, when the distance among the QDs is such that there is considerable overlap of electronic wavefunctions. Spectroscopy of QDM provides a new direction of scientific investigation.
5. Self-organized quantum wire via shape transition

There has been theoretical work on the possibility of formation of self organized quantum wire via shape transition of islands grown in the SK or VW modes [15]. These islands grow following the symmetry of the substrate surface. For example, equilateral triangle shaped islands grow on a surface of threefold symmetry such as Si(111) [16]. Square shaped islands grow on a substrate of fourfold symmetry such as Si(100). When these islands grow with the addition of more material, they grow in the original shape up to a critical size, where a shape transition occurs. Beyond the critical size, islands grow elongated as a long wire. This is how self-organized quantum wires can be grown. An example of such shape transition displaying the growth of self-organized nanowires is shown in figure 7. This is example of CoSi$_2$ growth on a Si(100) substrate [17].
On a surface of twofold symmetry, such as Si(110), unidirectional nanowires grow from the beginning. Growth of FeSi$_2$ nanowires on Si(110) is such a case [18].

6. Electronic structures for structures in the dimensional crossover regimes
With the available present experimental techniques, it is possible to fabricate structures which are neither zero-dimensional (0D) nor one-dimensional (1D), but between 0D and 1D. Such structures are in the dimensional crossover regime. For a long time the electronic density of states (DOS) were known for 0D, 1D, 2D and 3D structures. However, it was not known how DOS evolves for structures in the dimensional crossover regimes. A recent free electron model explains how overall DOS and DOS at Fermi energy evolve for structures in all the dimensional crossover regimes (0D-1D, 1D-2D, 2D-3D, 0D-2D, 0D-3D, 1D-3D) [19]. A 0D structure, when grows laterally, eventually becomes a 2D structure. How the DOS evolves as the lateral size of the structure increases is shown in figure 8.

Experimental results of DOS at Fermi energy using scanning tunneling spectroscopy for structures in the 0D to 2D dimensional crossover regime are available in Ref. [19].
7. Electronic transport in structures in the 2D-3D crossover regimes

Electronic tunneling transport has been investigated by scanning tunneling spectroscopy in one-atomic-layer to five-atomic-layer Ag thin films on Si(111) substrates [20]. The current-voltage (I-V) curve shows negative differential resistance (NDR). The voltage where NDR sets in depends on the number of atomic layers in the film. One-atomic-layer film is 2D. As the atomic layers are added one by one to the film it approaches 3D. So this example is a case of structures in the 2D to 3D crossover regime. NDR results are seen in figure 9. The NDR onset voltage is marked by a vertical bar. Its position depends on the number of atomic layers, because the DOS varies with the number of atomic layers as shown in figure 10. Particular peak positions in DOS have a one to one correspondence with the onset voltage of NDR.
8. Conclusions
Self-organized epitaxial structures displaying quantum phenomena have been discussed with examples. Epitaxial self-organized nanoislands can grow directly as islands on a substrate or in the layer-plus-island growth mode. Nanowires can grow via shape transition of islands. Evolution of electronic density of states in structures in the dimensional crossover regimes is shown. An example of electronic transport, namely negative differential resistance, in a system evolving from two-dimension towards three-dimension by addition of atomic layers one by one, is discussed. This shows the tunability of properties in quantum structures and their device potential.

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