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A Theoretical Study of Structural Disorder and Photoluminescence Linewidth in InGaAs/GaAs Self Assembled Quantum Dots

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ABSTRACT

The past few years have seen considerable efforts in growth and device application of self-assembled quantum dots. However, the photoluminescence (PL) linewidth, which represents structural fluctuations in dot sizes, is still in the range of 30-50 meV. This large linewidth has deleterious effects on devices such as lasers based on self-assembled dots. In this paper we will examine the configuration-energy diagram of self-assembled dots. Our formalism is based on: (1) an atomistic Monte Carlo method which allows us to find the minimum energy configuration and strain tensors as well as intermediate configurations of dots; (2) an 8-band $k \cdot p$ method to calculate the electronic spectra. We present results on the strain energy per unit cell for various distributions of InAs/GaAs quantum dots and relate them to published experimental results. In particular we examine uncovered InAs/GaAs dots and show that in the uncovered state a well-defined minimum exists in the configuration energy plot. The minimum corresponds to the size that agrees well with experiments.

INTRODUCTION

The use of strained epitaxy to create quasi-0 dimensional quantum dots has been widely studied over the last decade. Self-assembled quantum dots have now been fabricated using InGaAs/GaAs, SiGe/Ge, and many other strained systems [1-2]. In addition to the study of electronic and optical properties of these dots, devices lower as quantum dot interband lasers [3] and intersubband detectors [4] have been demonstrated. However, in spite of much progress in the area of self assembly, there is still a nagging issue - dot size nonuniformity. The self assembled dots vary in sizes and shapes - a variation that reflects in the electronic spectrum, photoluminescence linewidth, and gain spectrum etc. For many potential device applications, dot size nonuniformity creates deleterious effects. To understand the reason for nonuniformity and its extent, it is important to study the configuration energy of self-assembled dots.

There have been several studies on the strain tensor and energy in self-assembled dots. A useful model has emerged the valence force field (VFF) method [5,6], which allows us to calculate the strain tensor at an atomic level. It is found that the strain tensor in dots plays a key role in determining the electronic spectrum [7]. While the strain tensor of quantum dots in the covered state (i.e. with the dot buried in the large bandgap material matrix) has been examined, to our knowledge no work has been done on why certain mean dot sizes are chosen and why there is a distribution in the dot size. In this paper we use an extension of the VFF model to examine the strain tensor and energy of "covered" and "uncovered" InAs/GaAs dots. Then we apply the 8-band $k \cdot p$ model to obtain the electronic spectrum.

THEORETICAL MODEL

It is known that when a large strain exists between an overlayer and a substrate ($\varepsilon > 3\%$), the growth of the overlayer is described by the Stranski-Krastanow mode. In this mode a thin "wetting layer" is followed by an island growth. For InAs/GaAs system, the wetting layer is ~ 1 monolayer, and the islands are approximately 60 Å high with a more or less pyramidal shape. The base of the pyramid is about twice the height [8, 9]. This description of the self-assembled dots is; however, very qualitative since there is a considerable variation in the size and shape. There is substantial theoretical work on why the growth mode is island growth for high strain epitaxy. However, there is no quantitative work on why a particular size dominates the self-assembled system. To shed light on this issue, we calculate the strain tensor and energy for various sizes/shapes of InAs dots on a GaAs substrate. We examine the "uncovered" dots where the GaAs overlayer is not present and the "covered" dots where dots are embedded in a GaAs matrix.

The strained InAs/GaAs quantum dot system is shown in figure 1 for growth along the [001] direction. The InAs island is pyramidal shaped with a small square base, lying on a 1-monolayer InAs wetting layer. The InAs QDs are embedded in a GaAs matrix.

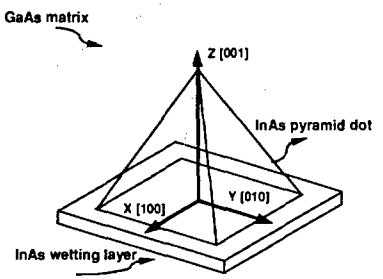

Figure 1. Schematic illustration of the Stranski-Krastanow growth mode for InAs/GaAs system.

Valence force field (VFF) model is a microscopic elastic theory, which includes the bond-stretching and bond-bending parts. The total VFF energy is taken as

$$U = \frac{1}{4}\sum_{ij}\frac{3}{4}\alpha_{ij}\left(d_{ij}^{\ 2} - d_{0,ij}^{2}\right)^{2} / d_{0,ij}^{2} + \frac{1}{2}\sum_{i}\sum_{j\neq k}\frac{3}{4}\beta_{ijk}\left(\mathrm{d}_{ij}\bullet\mathrm{d}_{ik} + d_{0,ij}d_{0,ik}/3\right)^{2} / d_{0,ij}d_{0,ik} \qquad (1)$$

where i represents all atomic sites, and j, k are the nearest neighbor sites. d_{ij} is the vector joining sites i and j, d_{ij} is the bond's length, and $d_{0,ij}$ is the corresponding equilibrium bond length. α and β are the bond-stretching and bending constants. To fully understand the formation of self assembled InAs QDs, we simulate the uncovered InAs dots by replacing the GaAs cap with a virtual material by artificially reducing α and β to very small values (factor of 10 smaller). As a result, we can observe the behavior of InAs islands before the covering GaAs is deposited.

The strain tensors are solved by minimization of the total energy within the framework of the VFF model. We use the approach taken by several authors [10-12]. In the beginning all atoms are placed on the GaAs lattice. The atoms are allowed to deviate from this starting position and periodic boundary conditions are assumed in the plane perpendicular to the growth

condition. In each process, only one atom is displaced while other atoms are held fixed. All atoms are displaced in sequence. The whole sequence is repeated until the maximum distance moved is so small that there is essentially no change in the system energy. The strain energy E_{str} can then be obtained once the strain tensors can be obtained by

$$E_{str} = \frac{a^3}{4}\left[\frac{1}{2}c_{11}\left(\varepsilon_{xx}^2 + \varepsilon_{yy}^2 + \varepsilon_{zz}^2\right) + c_{12}\left(\varepsilon_{yy}\varepsilon_{zz} + \varepsilon_{zz}\varepsilon_{xx} + \varepsilon_{xx}\varepsilon_{yy}\right) + \frac{1}{2}\left(\varepsilon_{zx}^2 + \varepsilon_{yz}^2 + \varepsilon_{xy}^2\right)\right] \quad (2)$$

where a is the lattice constant, ε_{ij} is the strain tensor component, and c_{ij} is the elastic constant. The 8-band $k \cdot p$ method [13] is used to calculate the electronic spectra.

RESULTS

The calculated results for strain energy per unit cell as a function of dot size in a covered InAs/GaAs system are shown in figure 2. The size variation is done when the base to height ratio is maintained at 2:1. No clear preference appears in the form of a well defined energy minimum. The strain energy of InAs dots for large sizes even show higher values than forming a flat thin layer (~ 0.17 eV), which is unexpected. It is clear that the InAs dot size/shape is determined by *the energies of the problem before the GaAs overlayer is deposited.*

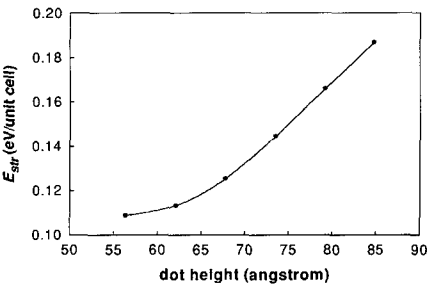

Figure 2. Strain energy per unit cell for the covered InAs dot pyramid at various dot sizes.

Figure 3 addresses the question of why certain dot sizes are preferred in high strain epitaxy. The strain energy per unit cell is illustrated as a function of the dot size. We show results for the dot alone and also the results when the wetting layer is included. Inclusion of the wetting layer is a little arbitrary since the results depend on the dot density. Nevertheless a clear minimum arises in the configuration energy plots in both cases at 62 Å dot height. This value is remarkably close to empirical observation [9]. However, due to entropy consideration we do not expect all dots to have this minimum energy size. Experimentally we know that there are variations in the dot size, causing inhomogeneous broadening of the photoluminescence line. Figure 3 also lists the values of effective bandgaps, showing how the dot size can alter the effect bandgap. We find that the effective gaps range from 0.954 to 1.2 eV as the height changes from 85 to 55 Å. The preferred size has a peak energy of ~ 1.04 eV, which agrees well with the experimental PL results [14].

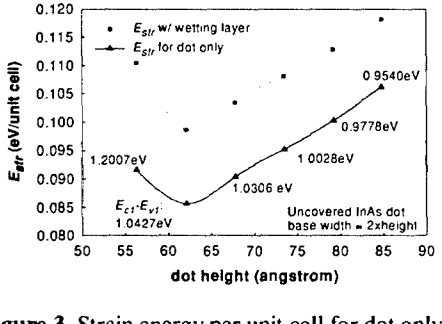

Figure 3. Strain energy per unit cell for dot only and for dot including wetting layer and effective bandgaps for the uncovered InAs dot pyramid at various dot sizes.

The electronic spectrum for different dot sizes is shown in figure 4. Here we show the ground states and the excited states of dots with heights of 56.5 Å and 62.3 Å. In each case the base is twice the height. As can be seen, the size variation will contribute to more fluctuation for higher level transitions.

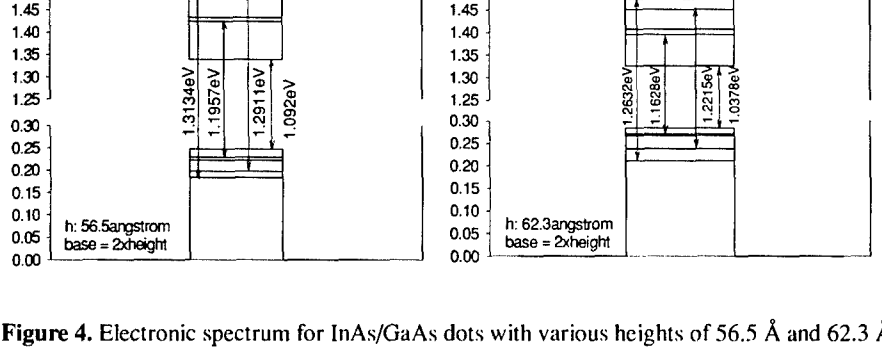

Figure 4. Electronic spectrum for InAs/GaAs dots with various heights of 56.5 Å and 62.3 Å.

Figure 5 shows how the strain energy change as a function of the shape of the dot. Here the height is fixed at the lowest configuration energy found in figure 3 while the width is adjusted. A larger base width corresponds to a wider dot pyramid and vice versa. It can be seen that when the base equals twice the island height the energy is a minimum. This result is consistent with experimental observation [9]. It is important to note that the total energy of the system would include the wetting layer strain energy and one has to then account for the dot density as well.

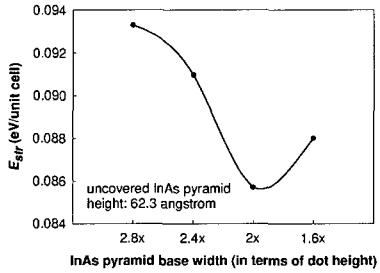

Figure 5. Strain energy of dot formation per unit cell for different dot shapes (various base).

CONCLUSION

In this paper we have examined the configuration energy profile for InAs/GaAs self-assembled dots. While it is not possible to examine the infinite possible sizes and shapes a dot can take, we find that there is a well defined size and shape as which the stain energy for the uncovered dot becomes a minimum. The shape and size calculated by us using the VFF model agree quite well with experimental findings on InAs/GaAs dots. We have also examined the configuration energy plot for covered dots, which do not show any energy minimum at the dot size observed empirically. This suggests that the dot size/shape is determined before the covering GaAs layer is deposited. Although the configuration energy is estimated assuming a pyramidal shaped dot, the calculation is done by a microscopically atomistic model. Thus this approach can be exploited to the truncated pyramidal quantum dots or even in the microlenses structure.

Another outcome of our finding is that the strain energy difference between the minimum energy configuration and other nearby dot sizes is not very large. As a result, it is expected that there will be a distribution of dots in actual growth due to entropy considerations. We have also shown how the size of dots alters the effective band-edge using the 8-band $k \cdot p$ method.

The good agreement of the minimum energy dot size and shape with published experimental results gives us confidence that the VFF model can be explored to understand how the dot uniformity can be controlled. This could be done by examining the role of pre-existing strains in the GaAs substrate. Further work will address these issues as well as the issue of dot size distributions.

ACKNOWLEDGEMENT

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