THE EQUILIBRIUM SHAPE OF InAs QUANTUM DOTS GROWN ON A GaAs (001) SUBSTRATE

E. PEHLKE, N. MOLL,[a] AND M. SCHEFFLER
Fritz-Haber-Institut, Faradayweg 4-6, D-14195 Berlin-Dahlem, Germany

The equilibrium shape of strained InAs quantum dots grown epitaxially on a GaAs(001) substrate is derived as a function of volume. InAs surface energies are calculated within density-functional theory, and a continuum approach is applied for the elastic relaxation energies.

1 Introduction

The strain-induced self-assembly of small three-dimensional islands during heteroepitaxial growth represents an efficient way of producing quantum dots. A frequently studied example is InAs/GaAs(100),[1] with the lattice mismatch amounting to about 7%. InAs grows on GaAs in the Stranski-Krastanov mode: Due to the smaller surface energy of InAs as compared to GaAs first a wetting layer forms, but when more InAs is deposited coherent, dislocation-free three-dimensional islands appear. The driving force for the island-formation originates in the gain of elastic relaxation energy which overcompensates the energetical costs due to the increased surface area. However, the details of the growth mechanism, especially the reason behind the narrow size distribution found for InAs/GaAs(100) quantum dots, are still controversial.[2,3]

In this paper we will derive the equilibrium shape of the InAs quantum dots as a function of size. With restricting ourselves to the energetics we do not imply that kinetic effects are unimportant for shaping the dots. However, we expect the equilibrium 3D-island shapes to be observable under appropriate experimental conditions. When the concentration of dots is low, shape equilibration by atomic diffusion on the small islands will be faster than material exchange between the islands leading to Ostwald ripening.

To calculate the equilibrium 3D-island shapes we have computed InAs surface energies from first-principles for a variety of surface orientations. A continuum approach is adequate to calculate the elastic relaxation energy. Combining these two contributions we get the total energy, which is minimized with respect to the island shape. The more delicate energetical effects, like island-island interaction, the strain-dependence of surface energy etc., which

[a] Present address: Massachusetts Institute of Technology, Department of Physics, Cambridge, MA 02139, USA.

have to be considered when discussing island sizes,[3] can be neglected for the purpose of predicting island shapes at fixed volume.

2 InAs Surface Energies

Surface energies have been calculated using density-functional theory, with the local-density approximation being applied to the exchange-correlation functional. The surface is represented by a periodically repeated slab, about 10 atomic layers thick, with the topmost 4 layers being fully relaxed. The In and As atoms are described by *ab initio* pseudopotentials, and the electron density is calculated using special **k**-point sets with a density in reciprocal space equivalent to 64 **k**-points in the whole (100) (1×1) surface Brillouin-zone. The wave-functions are expanded into plane waves with a kinetic energy ≤10 Ry. We have employed a generalized version of the computer code `fhi93cp`[4] to calculate the energy density according to Chetty and Martin.[5] The surface energy is derived from integrals of the energy density over appropriately chosen subvolumes of the supercell. For a detailed description of similar calculations for GaAs see Ref. 6.

Table 1: The equilibrium surface reconstructions of InAs under As-rich conditions and their surface energies.

orientation	reconstruction	surface energy [meV/Å^2]
(110)	(1×1) relaxed cleavage plane	41
(100)	α(2×4)	48
(100)	c(4×4)	47
(111)	(2×2) In vacancy	42
($\bar{1}\bar{1}\bar{1}$)	(2×2) As trimer	36

As the InAs surface reconstructions are expected to be similar to those of GaAs we have carried out calculations for the same surface reconstructions as in Ref. 6. Epitaxial growth most often takes place under As-rich conditions, thus the data in Table 1 refer to surfaces in equilibrium with bulk As (A7 structure). As opposed to GaAs the As-terminated InAs(110) (1×1) surface does not become stable, instead the relaxed cleavage plane is energetically preferred. For InAs(100) the As-terminated c(4×4) reconstruction yields the lowest surface energy. However, the energy difference with respect to the α(2×4) reconstruction is so small that our calculation is compatible to the experimental observation of a (2×4) reconstruction.[7] Our (111) and ($\bar{1}\bar{1}\bar{1}$) equilibrium reconstructions are consistent with recent core-level and valence-band photoemission

studies.[8,9] The (111)(2×2) As-trimer reconstruction, which is stable for GaAs under As-rich conditions, does not become stable in case of InAs. The surfaces in Table 1 are thermodynamically stable against faceting into each other. In fact, all four facets have been observed on large, and thus presumably relaxed, InAs islands grown on a GaAs(001) substrate.[10]

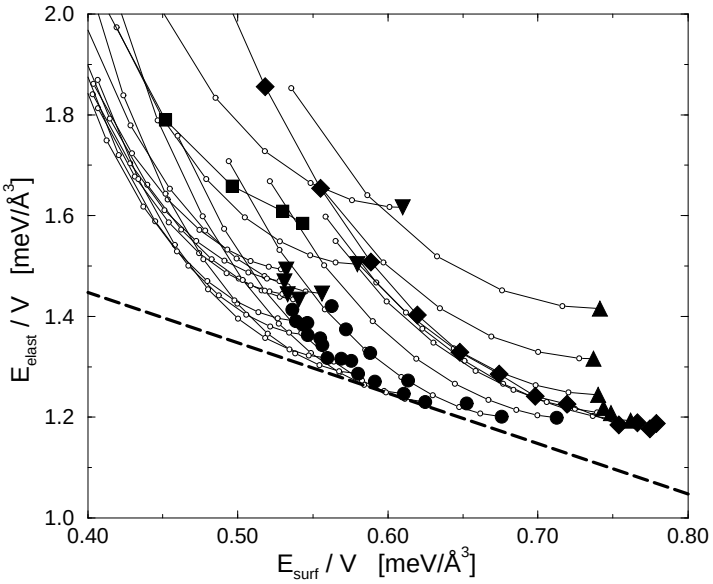

Figure 1: Elastic energy per volume E_{elast}/V vs. surface energy per volume E_{surf}/V for InAs islands with volume $V = 2.88 \times 10^5 \text{Å}^3$. Squares: square based pyramid with {101} faces and (001)-truncated {101}-pyramids. Diamonds: square based pyramids with {111} and $\{\bar{1}\bar{1}\bar{1}\}$ faces and (001)-truncated pyramids. Triangles up: "huts" with {111} and $\{\bar{1}\bar{1}\bar{1}\}$ faces. Triangles down: square based {101} pyramids with $\{\bar{1}\bar{1}\bar{1}\}$-truncated edges. Dots: islands with {101}, {111}, and $\{\bar{1}\bar{1}\bar{1}\}$ faces. Filled symbols denote numerical results, while open circles correspond to a simple analytical approximation for (001)-truncated "mesa-shaped" islands. It is assumed that the elastic energy does not change when the (almost fully relaxed) top of an island is cut off. Full lines connect islands that are created in this way, varying the height of the (001) surface plane. The dashed line is the curve of constant total energy $E_{elast} + E_{surf}$ that selects the equilibrium shape.

3 Elastic Energy and Equilibrium Shape of Quantum Dots

The elastic energy is calculated within continuum theory, using a finite-element type of approach. The strain field both in the island and in the substrate is fully accounted for. For simplicity, the same elastic constants[11] have been taken

both for the InAs island and the GaAs substrate. The results are summarized in Fig. 1 for a specific volume V of the islands. Generalization to arbitrary volume can be done by means of the scaling relations $E_{elast} \sim V$ and $E_{surf} \sim V^{2/3}$. The equilibrium island-shape is marked by the point where the lines of constant total energy $E_{elast} + E_{surf}$ (dashed line in Fig. 1) touch the manifold of island energy-curves from below. The volume-dependence of this shape can be inferred from the same figure by rescaling the E_{surf}/V axis: For larger volume V the slope of the total-energy lines becomes smaller.

The equilibrium island shape results from the competition between elastic and surface energy. According to the different scaling properties of E_{elast} and E_{surf} for large volumes the elastic energy dominates, favoring a steep pyramidal shape, while small islands become more and more flat. Within the configuration space we have examined the optimum shapes can be described as mesa-type hills bounded by {101}, {111} and $\{\bar{1}\bar{1}\bar{1}\}$ faces and an (001)-surface on the top. While they differ from the {101}-pyramids seen in experiment [12] they are in fact similar to shapes observed for InP/GaInP islands.[13]

¿From Fig. 1 it is obvious that the shape evolution is continuous with respect to the volume. This causes an additional V-dependence in $E_{elast}^{equil}(V)$ and $E_{surf}^{equil}(V)$, which does not follow "standard" scaling anymore. Though our total-energy expression does not yield any optimum island size by its construction, this effect is important for a theory of equilibrium island size.[3]

References

1. D. Leonard *et al*, *Appl. Phys. Lett.* **63**, 3203 (1993).
2. C. Priester and M. Lannoo, *Phys. Rev. Lett.* **75**, 93 (1995).
3. V. A. Shchukin *et al*, *Phys. Rev. Lett.* **75**, 2968 (1995).
4. R. Stumpf and M. Scheffler, *Comput. Phys. Commun.* **79**, 447 (1994).
5. N. Chetty and R. M. Martin, *Phys. Rev. B* **45**, 6074 (1992).
6. N. Moll *et al*, *Phys. Rev. B.* (in press).
7. H. Yamaguchi and Y. Horikoshi, *Phys. Rev. B* **51**, 9836 (1995).
8. L. Ö. Olsson *et al*, *Phys. Rev. B* **53**, 4734 (1996).
9. C. B. M. Andersson *et al*, *Surf. Sci.* **347**, 199 (1996).
10. E. Steimetz *et al*, to be published.
11. The experimental second and third order elastic moduli of GaAs were used. *Data in Science and Technology, Semiconductors, Group IV Elements and III-V Compounds*, Ed. O. Madelung (Springer, Berlin, 1991).
12. S. Ruvimov *et al*, *Phys. Rev. B* **51**, 14766 (1995).
13. K. Georgsson *et al*, *Appl. Phys. Lett.* **67**, 2981 (1995).