Role of elastic anisotropy in the vertical alignment of In(Ga)As quantum dot superlattices

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This letter shows the effect of the elastic anisotropic strain field in the vertical stacking for multilayer In(Ga)As/GaAs quantum dot (QD) structures grown by molecular beam epitaxy. This effect explains the QD enlargement in vertical correlated stacking and the possibility to obtain QD vertical anticorrelated stacking. The anticorrelation configuration in these structures is dependent not only on the QD size and on the interlayer spacing but also on the QD density. With appropriate choice of these structural parameters, a QD body centered cubic arrangement can be produced with a narrower size distribution which is in marked contrast to commonly observed vertically correlated QDs. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2202190]

One of the remarkable properties of strain induced nanometer-sized islands, known as quantum dots (QDs), is the presence of a number of selection processes, which control aspects such as island size and interisland lateral spacing. Recently, much attention has been focused on QD multilayer ordered both in the lateral and vertical directions.[1–5] But, while the lateral ordering of islands has not been observed to affect the island size, vertical interactions between evolving strain fields usually modify the size and shape of such islands, making very difficult to fabricate high quality superlattices based on closely vertically correlated QDs.

Vertical correlated stacking (VCS), during the epitaxial growth of QD superlattices, has been observed to occur when the separation between layers is sufficiently small such that the elastic distortion associated with QDs extends into the surrounding material, creating preferential regions for the onset of three dimensional growths in the subsequent strained layer.[4,6–11] It has often been proposed that this vertical elastic interaction can lead to lateral ordering and size homogenization of the islands.[1,2,4,10] However, under close examination quite the opposite is observed, i.e., in vertically correlated In(Ga)As stacks subsequent QDs grow larger and even splitting can occur. Theoretical treatments have been successfully applied to explain VCS based on accounting the strain induced migration of atoms of the growing layer to positions above underlying QDs.[4,6,12,13] However, the detailed mechanism is still the subject of considerable debate, depending on the relative importance of nucleation and post-nucleation effects.

Recently, vertical *anticorrelated* stacking (VAS), defined as structures in which subsequent quantum dots are shifted from the vertical of underlying QDs, has been reported for the growth of highly anisotropic structures such as PbSe/PbEuTe and CdSe/ZnSe QD superlattices.[3,5,14] To explain this behavior, some authors have proposed that it is possible to obtain near equilibrium VAS QD structures only for highly elastic anisotropic materials.[3,12,15,16] In this sense, In(Ga)As islands on GaAs (001) have been observed to show surface alignment effects,[13,17,18] which give a clear indication of the anisotropy of the intrinsic surface stress tensor and the anisotropy of the bulk elastic modulus tensor. However, the molecular beam epitaxy (MBE) growth is governed by growth kinetics and this fact explains the lack of observations of anticorrelated behavior despite numerous studies of InGaAs multilayers. Thus, only Wang *et al.*[19] have observed recently the formation of isolated areas of VAS in InGaAs multilayers.

This letter shows that working *with* the strain anisotropy of the crystal lattice to get VAS, rather than attempting to produce VCS by working *against* strain anisotropy, presents a more viable approach to achieve In(Ga)As QD superlattices with a high uniformity in size and shape and a regular in-plane spatial distribution. In(Ga)As/GaAs VASs are possible on a large scale resulting in near-perfect quantum dot superlattices with a narrower QD size distribution and that further refinement in design and MBE growth technique could produce very highly ordered anticorrelated QD pseudocrystals, exhibiting a *body centered cubic* (bcc) type arrangement. So, these structures would address a number of needs, to improved quantum dot lasers based on an increase in the gain of the active layer, due to improved close packing. However, they remain a formidable challenge to fully exploit their anticipated merits of such alignments for multidimensional quantum confinement, particularly due to the weak anisotropy of this system.

Multilayered In(Ga)As QD structures sandwiched between GaAs barriers have been grown by molecular beam epitaxy in a VG V80H system on nominally on axis (±0.05°) GaAs (001) substrates. The first sample, labeled A, consisted of five layers of 3 ML (monolayer) of InAs grown at 510 °C separated by 25 nm of GaAs barriers. After the first 15 nm of the GaAs barrier layer, the growth was interrupted and the temperature increased to ~600 °C where it was held for 2 min to achieve a total desorption of indium from the growth surface to prevent residual indium on the subsequent layer. The growth temperature was then decreased by 20 °C to grow the rest of the barrier layer under conventional GaAs growth conditions. The second sample, labeled B, consisted

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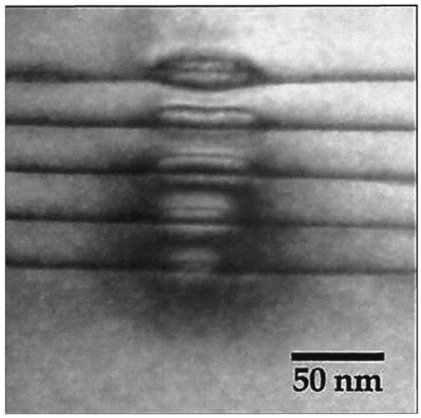

FIG. 1. g002DF CS TEM micrograph of five layers InAs/GaAs QDs structures, showing the enlargement of the dots during the stacking process.

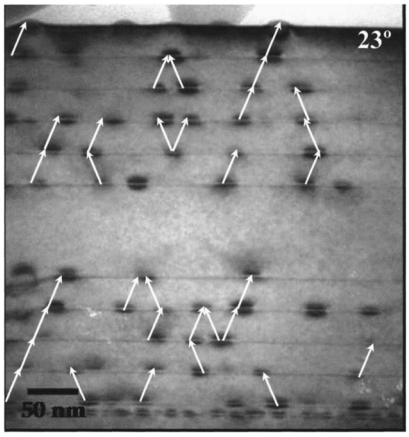

FIG. 2. g400BF CS TEM image of the designed structure of InAs/GaAs QDs, showing the shift angle of 23° of the upper dots with regard to the vertical of the buried ones.

of five layers of 2.2 ML of InAs and 35 nm of GaAs barriers and the third one, labeled C, consisted of 16 repeat 5.5 ML $In_{0.5}Ga_{0.5}As$ with a nominal GaAs barrier spacing of 25 nm, both of them grown using the growth temperature conditions described above. Plan-view (PV) and [110] cross-sectional (CS) transmission electron microscopy (TEM) images were taken using a Philips 430 microscope operating at 300 kV.

The low magnification TEM image of sample A (Fig. 1) is very similar to previous reports, e.g., Solomon *et al.*[2,8] or Xie *et al.*,[6] and shows the vertical interaction between InAs QDs, resulting in a QD size increase and in a density reduction. Specifically, the bottom layer density is $\rho=13.1\times10^4$ cm^{-1} and the top layer density is $\rho=7.1\times10^4$ cm^{-1}, with a $\Delta\rho$ of up to 50% as we move up the stack. Although there is a clear QD size increase from the bottom to the top layer (height increase $\Delta h=+68\%$ and width increase $\Delta w=+51\%$), the aspect ratio h/w for all the layers was the same and equal to 0.2. An estimation of the total volume suggests that there must be a reduction in the average In concentration of the dots, with a top layer concentration being ~25% of the bottom layer concentration. It is clear therefore that such multilayers contain very inhomogeneous arrays of QDs, with a size distribution far wider than that of the first deposited layer.

In this sample, vertical In diffusion effect that would explain the QD size increase is rejected since the temperature increase to ~600 °C, after the growth of the first 15 nm of GaAs barrier, produces the total desorption of In from the growth surface. So, these observations are explained based on the strain energy profile above the three dimensional islands. Vertical correlation of three dimensional islands has been modeled on the assumption of strain field profile for an isotropic medium.[1,4,6] However, III-V semiconductors are elastically anisotropic, with anisotropy ratios for GaAs and InAs of 1.83 and 2.08, respectively. Following this reasoning, several authors have proposed that both VCS and VAS in self-assembled QD structures can be explained, taking into account the elastic anisotropy of the strain energy profile above a predeposited QD layer.[3,5,12,14–16,20] The elastic anisotropy would change both the depth and the position of the energy minima with respect to elastically isotropic spacer layers where the energy minimum is always on the top of the buried QDs. Specifically, for GaAs(001), there are four energy minima above a quantum dot which occur at ±23° from the vertical in the ⟨110⟩ directions, assuming the calculations of Holy *et al.*[14] So, for the ~40 nm QD width observed in the bottom layer of sample A, four strain minima shifted ±10.5 nm from the vertical along the ⟨110⟩ directions would occur above these islands. However, the lateral distance among these energy minima in the surface is not large enough to give rise to the formation of four individual islands; so instead, the coalescence of the four islands occurs, originating a large island and exhibiting a hump-back shape, as is very commonly observed. It is clear therefore that, in this sample, closely spaced InAs QDs suffer from an inherent inability to match to strain minima distances, which are smaller than their relatively large lateral size.

Sample B was designed using structural parameters to produce VAS, i.e., to avoid the overlapping of the QDs due to the anisotropic strain profile of the buried islands. Figure 2 shows a g400BF TEM image of this sample in which, at a first sight, it seems that there is an arbitrary distribution of the QDs. But this appearance is due to a low QD density since a detailed analysis showed that the position of each dot exhibits a preferential shift angle of 23° with regard to the vertical of the dot immediately below (as shown by arrows) following the model of Holy *et al.* These results confirm experimentally that the distribution of dots in a multilayered In(Ga)As/GaAs QD structure is a function of the elastic anisotropy of the material. Moreover, in the g200DF TEM analysis it was observed that there exists a high island size uniformity, in marked contrast to sample A.

Figure 3(a) shows a cross section g002DF micrograph of sample C, where a high density of anticorrelated InGaAs QDs is observed. As shown by arrows, the shift angle of the position of the dots in upper layers with regard to the growth direction is ~50°, far away from the 23° proposed by Holy *et al.*[14] and found experimentally in the cross section of sample B. The reasons for the deviation from the prediction of the model of Holy *et al.* stem from the different density of QDs found in both samples. In the InAs QD structure (sample B), there is a low QD in-plane density and so most of the dots suffer the strain field of only one buried dot. In this case, the model of Holy *et al.* succeeds in predicting the position of the dots, as shown in Fig. 2. However, in sample C, the high density of dots causes the interaction of strain field profiles in the growth surface due to several buried dots, notably the overlapping strain fields of the four nearest neighbors below.

The assembly of bidimensional arrays of QDs distributed in stacked layers has been studied theoretically as func-

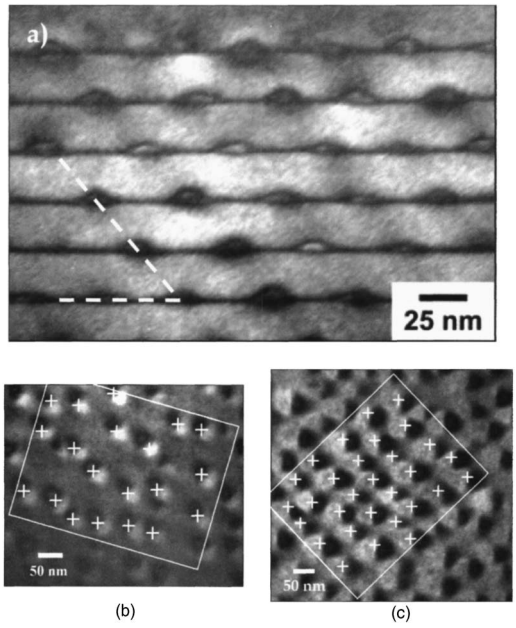

FIG. 3. TEM micrographs of InGaAs/GaAs QD anticorrelated heterostructures. (a) g002DF CS TEM images of sample C and (b) and (c) are 220 planar view TEM images of samples with one ($\rho=1.6\times10^{10}$ cm^{-2}) and two layers ($\rho=3\times10^{10}$ cm^{-2}) of InGaAs islands, respectively.

tion of the separation between QD layers, z_0, and lateral distance between QDs, D, by Shchukin *et al.*[15] For a given array of buried islands, the authors predict that the preferential anticorrelation sites are the center of the distance between QDs aligned in the [110] directions for wide z_0/D ratios. So, the lateral distance between QDs, i.e., the QD density, clearly explains the different behaviors with respect to the shift angle found in our work in InGaAs dots (sample C) with regard to InAs ones (sample B). Moreover, it should be highlighted that the proposed design gives rise to InGaAs QD structures with optimized characteristics in the {110} plane, i.e., a high density of closely packed, uniformly sized quantum dots with a regular spatial distribution.

The observed anticorrelated structure resembles a pseudocrystal, exhibiting a bcc form aligned with the zinc-blende reference system. With the aim of seeing this bcc arrangement, two samples were grown in the same conditions as sample C. Figures 3(b) and 3(c) show planar view micrograph of these samples containing one and two QD layers, respectively. As it can be observed, for the same area (represented by the white square) the density of dots is double in the sample with two layers. Note, moreover, that in the second sample the QDs follow a near-perfect square pattern by plan view. The island arrangement of the top QD layer is not just simply a reproduction of the buried islands, showing a clear effect of ordering due to anticorrelation. Although further work is needed to achieve high quality superlattices based on the anticorrelation effect, it is also clear that there are short-term benefits to be gained from the observation that anticorrelation, rather than correlation, is the most appropriate method to obtain closely spaced QD multilayers. Through appropriate design to encourage VAS, the optimal configuration to control both effects mentioned above, it is possible that large numbers of closely spaced QDs could be achieved.

In summary, the intrinsic elastic anisotropy of zinc-blende structure plays a major role in the arrangement of quantum dots on multilayer planes. Firstly, it is responsible for the commonly observed poor size homogeneity of closely vertical spaced InAs QD stacks. Nevertheless, the most important effect is, with appropriate choice of these parameters, the possibility to grow anticorrelated QD structures which exhibit much improved size and density distribution. Thus, in the case of low QD density, the VAS follows the model of Holy *et al.*, placing the upper QDs in a fixed angle of 23° regarding the VCS from the bottom QDs. However, for high density InGaAs QD multilayers, QD bcc type arrangement can be produced by exploiting the overlap between the strain fields of the four nearest neighbors below the QD. Through refinement of structural dimensions (QD size and density and interlayer spacing) and careful choice of MBE growth conditions it may be possible to produce high quality closely packed QD pseudocrystals arising from the interaction between the strain fields of an assembly of QDs in three dimensions, which would open up a new study of long range QD superlattices.

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