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<table>
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Missing rotational symmetry vs. lateral and vertical ordering in InGaAs/GaAs(311)B quantum dot stacks</td><td>Experiment number:
HS-2652</td></tr>
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ID10B</td><td>Date of experiment:
from: 14.02.2007 to: 20.02.2007</td><td rowspan="2">Date of report:
29.01.2008

Received at ESRF:</td></tr>
<tr><td>Shifts:
18</td><td>Local contact(s):
Dr. Jiri Novak</td></tr>
<tr><td colspan="3">Names and affiliations of applicants (* indicates experimentalists):

Dr. Michael Hanke* (Martin-Luther-University Halle-Wittenberg, Germany)
Dr. Martin Schmidbauer* (Insitut for Crystal Growth, Berlin, Germany)
Dr. Peter Schäfer* (Humboldt-University Berlin, Germany)</td></tr>
</table>

Report:

We apologize for the short delay in submitting our report. This is partly due to the extensive data evaluation, and on the other hand due to sophisticated finite element calculations, which we had systematically to perform in order to receive a full picture of the strain and its relaxation in the probed InGaAs/GaAs quantum dot stacks. Eventually we have very recently published the results of this beamtime HS-3162 in the Applied Physics Letters: M. Hanke et al. APL **92**, 033111 (2008). So we have attached the corresponding paper as the essential part of our report.

Step bunch assisted two dimensional ordering of $In_{0.19}Ga_{0.81}As/GaAs$ quantum dots on vicinal GaAs(001) surfaces

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We have investigated the self-organized, step bunch assisted formation of $In_{0.19}Ga_{0.81}As/GaAs$ quantum dots in vertical superlattices consisting of one, four, eight, and ten periods. Samples were grown by molecular beam epitaxy on vicinal 2°A and 2°B GaAs(001) substrates. Those with miscut along the $[1\bar{1}0]$ (2°B) exclusively show step bunches of an aspect ratio larger than 10 but without the formation of quantum dots. This highly linear pattern is improved during subsequent periods as proved by high resolution x-ray diffraction and grazing incidence diffraction. On the other hand, a miscut along the [110] (2°A) initially causes a crosslike pattern of step bunches, which finally becomes a two-dimensional arrangement of individual quantum dots. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2838453]

Semiconductor quantum dots (QDs) and quantum wires (QWs) have attracted a long-lasting interest during the past decade because of potential applications in novel semiconductor devices.[1–3] Since then a rather promising and so widely employed attempt relies on the so-called Stranski-Krastanow (SK) process. Here, the heteroepitaxy starts with a very thin, planar layer which wets the surface. With increasing thickness (respectively, ongoing growth), the established strain energy in the deformed crystal lattice increases too. Thus, at a certain point, the energy gain due to elastic relaxation overcompensates the additional free surface energy, and the self-organized growth mechanism changes from a planar layer-by-layer to a three-dimensional (3D) mode.

While the SK growth conditions can be optimized to produce nanostructures of nearly identical shape and size, often only a random spatial distribution of the QDs is observed for a single layer of QDs. However, the characteristics of many applications (e.g., the performance of photonic crystals) might be reinforced if the QD positions can be controlled to a high degree of precision. Self-assembling in a single QD layer can either be induced by prepatterning (lithographically[4,5] or semiartificially by employing QWs as template for subsequent QD growth[6,7]), or along a fully self-organized, strain-driven path, e.g., see Refs. 8–10. An additional degree of freedom is achieved if the QDs are vertically stacked in *multiple* layers, so they usually get more uniform in size and ordering. A range of different results, from nearly perfect QD chains[11] to 3D-QD lattices[12] has been reported. In the particular case of multilayered InGaAs QDs embedded in a GaAs matrix, both the anisotropic surface diffusion and the elastic properties of the GaAs spacer layer are the most important parameters for the evolution of 3D ordering of the QDs.[13] Also, the effect of postgrowth annealing may play a crucial role.[14] However, in terms of custom-made 3D-QD lattices (with predictable lateral and vertical orderings) a more precise description of the QD growth and self-assembling has yet to be conceived.

The vertical superlattices investigated in this study were grown in a molecular beam epitaxy (MBE) chamber equipped with reflection high-energy electron diffraction. Double side polished, epitaxial ready *n*-type exact GaAs(001) and vicinal GaAs(001) substrates, tilted 2° toward the $[1\bar{1}0]$ (2°B) and [110] (2°A) directions, are loaded into the MBE chamber. As a reference point, an exact GaAs(001) sample was loaded side-by-side next to each vicinal GaAs(001) sample on a solid molybdenum block. The surface oxide layer was removed at 600 °C for 10 min, while exposed to a 6.4 μTorr As_4 beam equivalent pressure (BEP) from a solid source valved controlled As cell. After oxide desorption, substrate temperature was lowered to 580 °C and a 0.4 μm thick GaAs buffer layer was grown with a growth rate of 1.0 ML/s and an As_4 to Ga BEP ratio of 10. Subsequently, the substrate was cooled down to 540 °C, while keeping the As_4 BEP the same for the deposition of a InGaAs/GaAs multilayer structure. The multilayer structure consists of m periods (with $m=1$, 4, 8, and 10) of 20 ML of $In_{0.19}Ga_{0.81}As$ on top of 20 ML of a GaAs spacer with a 10 s growth interruption between each layer. The Ga growth rate during the growth of the $In_{0.19}Ga_{0.81}As$ multilayer structure was 0.37 ML/s. The last layer of InGaAs was left uncapped for topographic atomic force microscopy (AFM) imaging under ambient conditions. The growth was then followed by 10 s of annealing and the sample was then quickly quenched while lowering the As_4 pressure.

X-ray scattering techniques may probe entire vertical stacks with exceptional high statistics, which makes them excellent analytical tools to study 3D ordering therein. Due to comparatively small QD volumes, highly brilliant x-ray

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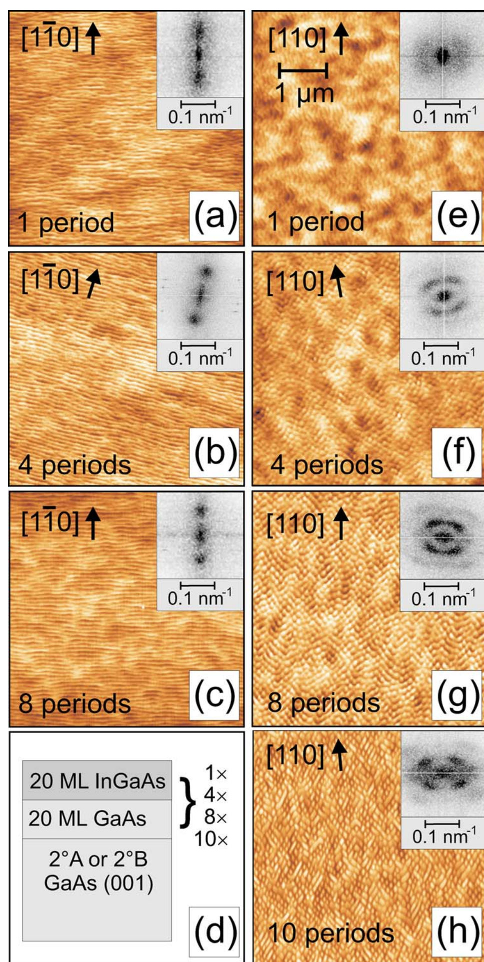

FIG. 1. (Color online) Atomic force micrographs (5×5 μm^2) for two orthogonal substrate miscuts {2°B [(a)–(c)] and 2°A [(e)–(h)]}. The various superlattices contain either one [(a) and (e)], four [(b) and (f)], eight [(c) and (g)], or ten periods of 20 ML $In_{0.19}Ga_{0.81}As$/20 ML GaAs (h), see schematic sample setup (d). The insets depict corresponding fast Fourier transforms (FFTs). Please note that the scales therein directly accord with the x-ray scattering patterns in Figs. 2 and 3.

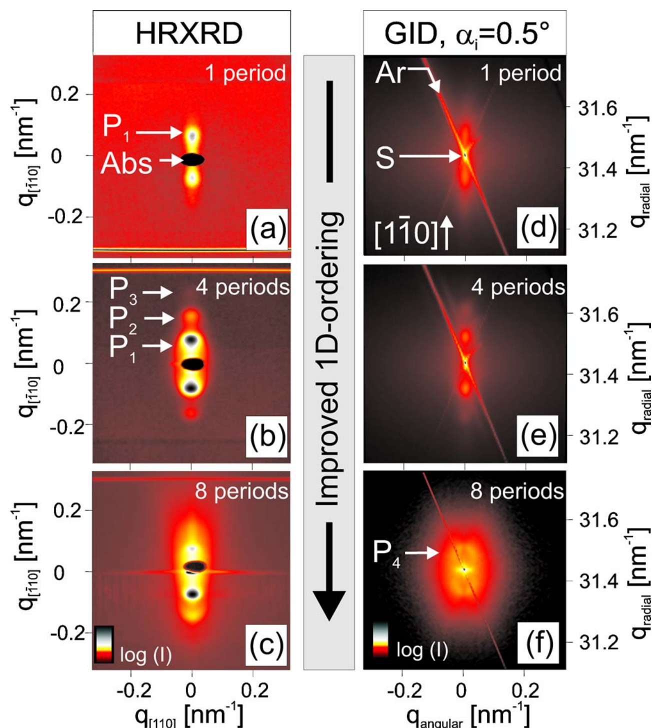

FIG. 2. (Color online) Diffusely scattered intensity near the (004) reflection [(a)–(c)] at $q_{\text{vertical}}=44.0$ nm^{-1}. An absorber (Abs) weakens highly intense scattering in the center of the image. (d)–(f) show the vicinity of the (220) GID reflection (S). The intensity distributions were measured at a single layer of InGaAs QDs (a) and (d) and various multilayer stacks containing four [(b) and (e)] and eight periods [(c) and (f)] on 2°B miscut GaAs(001), respectively. Several scattering satellites P_1 to P_4 probe the lateral ordering, (Ar) denotes a scattering artefact.

radiation becomes a mandatory requisit to measure the scattered x-ray intensity. We have first applied grazing incidence diffraction (GID) near the (220) reciprocal lattice point in order to probe the in-plane elastic strain and the lateral step bunch and QD ordering—both, in particular, near the surface, since the chosen angle of incidence of 0.5° figures only sligthly above the critical angle for total external reflection. Respective measurements were performed at beamline ID10B at the European Synchrotron Radiation Facility (ESRF), Grenoble using an x-ray energy of 8 keV. A positional sensitive detector placed 500 mm behind the sample enables an angular resolution along different exit angles α_f. Furtheron, an additional Si(111) crystal analyzer had to be used in order to enhance the angular resolution parallel the sample surface to about 0.03 nm^{-1}.

For the same set of samples, we have applied high resolution x-ray diffraction (HRXRD) at beamline BW2 (HASYLAB) as a complementary technique to GID, using the identical x-ray energy of 8 keV. Since the information depth in HRXRD gets an order of magnitude higher, this probes now lateral ordering throughout the entire stack. A two-dimensional charge coupled device, which simultaneously records the intensity along a curved area in reciprocal space, was utilized for recording 3D resolved scattering patterns. However, since we require *in-plane* intensity distributions (in order to compare them with respective GID measurements) respective sections through the 3D distribution have been extracted.

The atomic force micrographs in Fig. 1 clearly prove a different lateral ordering scenario for the 2°B [Figs. 1(a)–1(c)] and 2°A [Figs. 1(e)–1(h)] samples. The highly anisotropic (2×4) surface reconstruction on GaAs(001) induces anisotropic surface diffusion of the adatoms, which obviously favors step bunching in the first case [Figs. 1(a)–1(c)] and a two-dimensional alignment of QDs in the other [Figs. 1(e)–1(h)]. While the average inter-step-bunch (respectively, inter-QD) distance does not significantly change if the number of vertical layers increases (see insets in Fig. 1), the topmost surface morphology becomes a more pure one-dimensional (1D) [or two-dimensional (2D), in case of 2°A miscut substrates] pattern, which is proved by corresponding fast fourier transforms (FFTs). Corresponding FFTs are shown as insets in Fig. 1. It is worth noticing that the number and height of the bunched steps do not vary with the number of periods. While AFM exclusively probes the topmost surface HRXRD and GID, Fig. 2 (2°B) and Fig. 3 (2°A miscut samples), probe via different information depths the evolution of lateral ordering through the vertical stacks.

Both techniques yield a single ordering satellite P_1 in Figs. 2(a) and 2(d) for the 2°B single layer sample, similar to the FFT in Fig. 1(a). However, in contrast to that the scattering at both 2°B, vertical stacks with four and eight periods in Figs. 2(b), 2(c), 2(e), 2(f) exhibit a subsequently evolving 1D

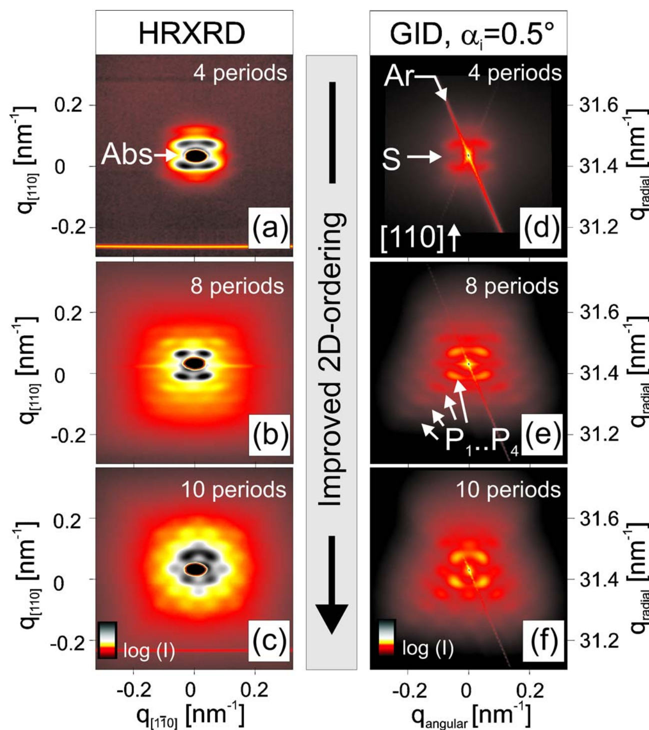

FIG. 3. (Color online) Diffusely scattered intensity near the (004) reflection [(a)–(c)] at q_{vertical}=44.0 nm^{-1}. An absorber (abs) weakens highly intense scattering in the centre of the image. (d)–(f) show the vicinity of the (220) GID reflection (S). The intensity distributions were measured at InGaAs/GaAs QDs multilayer stacks containing four layers [(a) and (d)], eight layers [(b) and (e)], and ten periods [(c) and (f)] on 2°A miscut GaAs(001), respectively. Several scattering satellites P_1-P_4 probe the lateral ordering. (Ar) denotes a scattering artefact.

ordering by the appearance of additional lateral ordering satellites $P_1 \ldots P_3$ along the $[\bar{1}10]$ direction. The in-plane GID pattern in Fig. 2(f), and, in particular, the presence of peak P_4, may be related to the 2D morphology at the tails of each bunch.

A similar trend of subsequently evolving lateral (however, 2D) ordering with increasing number of layers in a vertical stack was found for the samples on 2°A miscut substrates, Fig. 3. While the sample with just 4 periods shows a comparatively weak 2D pattern with lateral ordering satellites up to the first order around the substrate (S) peak [Figs. 3(a) and 3(d)], the in-plane patterns depict additional satellites up to fourth order ($P_1 \ldots P_4$) in case of 8 and 10 periods [Figs. 3(b), 3(c), 3(e), and 3(f)].

In conclusion, we have probed single and multilayer stacks of InGaAs/GaAs on 2°A and 2°B miscut GaAs(001) with direct atomic force microscopy, high resolution x-ray diffraction, and grazing incidence diffraction. We could show, that due to the vertical stacking, the lateral ordering on both orientations improves with increasing number of periods. While the surface on 2°B tends to form highly unidirectional 1D step bunches, the 2°A orientation yields highly ordered 2D QD patterns.

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