

Contents lists available at ScienceDirect

# Journal of Crystal Growth



journal homepage: www.elsevier.com/locate/jcrysgro

# Lateral variations in self-assembled InGaAs quantum dot distributions $\stackrel{\scriptscriptstyle \,\mathrm{tr}}{}$

Alexana Roshko<sup>\*</sup>, Todd E. Harvey, Brit L. Hyland<sup>1</sup>, Susan Y. Lehman<sup>2</sup>, Keith D. Cobry<sup>3</sup>

Optoelectronics Division, National Institute of Standards and Technology, 325 Broadway, Boulder, CO 80305-3328, USA

#### ARTICLE INFO

Article history: Received 6 December 2007 Received in revised form 15 June 2009 Accepted 21 June 2009 Communicated by K.H. Ploog Available online 26 June 2009

PACS:

68.65.Hb 81.15.Hi 81.05.Ea

#### Keywords:

A1. Atomic force microscopy

A1. Nanostructures

A1. Self-assembly A3. Molecular beam epitaxy

A3. Ouantum dots

B1. InAs/GaAs

### 1. Introduction

The need to achieve regular in-plane spatial distributions and sufficiently uniform size distributions of self-assembled quantum dots (QDs) has long been recognized as a major challenge for applications [1,2]. Numerous studies have addressed the size uniformity of self-assembled QDs on the local scale (less than 1 mm). Techniques such as low growth rates [3–5], low arsenic pressure [6], "punctuated" or "interrupted" growth [7–9], vertical coupling of stacked dots [10-13] and buried "stressor" dot layers [14–17] have been used to improve the uniformity of dot size (obtain narrower dot size distributions) on a local scale. Arrays of self-assembled dots with lateral ordering have been achieved on high-index GaAs substrates, through balancing the competition between anisotropic surface diffusion and the anisotropic elastic matrix [18-20]. However, whether these translate to more uniform lateral distributions on a large scale (e.g., from one side of a wafer to another) has not been examined.

## ABSTRACT

The lateral uniformity of self-assembled InGaAs quantum dots grown by molecular beam epitaxy (MBE) was assessed as a function of growth conditions. Variations in the dot density and height were determined from atomic force micrographs. Growth rate had a large influence on lateral uniformity. The most uniform dot distributions were grown at low rates, 0.15 monolayers/s (ML/s). Dots deposited at a rate of 1.15 ML/s had large variations in both height and density. These variations decreased as the dot density increased; however, they remained larger than those of dots deposited slowly. The lateral uniformity of dots deposited quickly also improved for the top layer of dots in stacked layers, even though these layers had decreased dot densities. There were negligible differences in the lateral height and density uniformities of dots as functions of continuous versus pulsed growth, wafer diameter and mole fraction of In.

Published by Elsevier B.V.

In most studies, the dot size has been measured indirectly through the photoluminescence (PL) linewidth, which is generally accepted to be inhomogeneously broadened due to lateral dispersion in dot size [21]. There have been reports of narrow linewidths, which correspond to narrow dot size distributions, for a variety of growth modes. However, PL analyzes a relatively small area of a specimen, typically a square millimeter or less. Likewise, studies that have examined the dot size distribution directly, through atomic force or scanning tunneling microscopy, have analyzed areas of several square micrometers. None of these studies has addressed the lateral dispersion in dot size across wafers. Therefore, while the local size dispersion may be quite small, there may be large variations in the average dot size across the specimen.

Similarly, the influence of growth parameters on local dot density has been extensively studied. The density has been found to increase with decreased substrate temperature [22–25] and with increased growth rate [4,23,25,26]. High As flux and a high degree of surface misorientation were also reported to increase the dot density, presumably because they both reduce the cation diffusion length [27]. The dot composition was found to affect the dot density and also to substantially influence the uniformity of the dot size distribution on a local scale [28]. However, again none of these studies examined the lateral variation in dot density at scales larger than several micrometers.

<sup>\*</sup>Contribution of the US government, not subject to copyright. \*Corresponding author.

E-mail address: roshko@boulder.nist.gov (A. Roshko).

<sup>&</sup>lt;sup>1</sup> Currently at University of California, Irvine, CA.

<sup>&</sup>lt;sup>2</sup> Currently at The College of Wooster, Wooster, OH.

<sup>&</sup>lt;sup>3</sup> Currently at University of Colorado, Boulder, CO.

<sup>0022-0248/\$ -</sup> see front matter Published by Elsevier B.V. doi:10.1016/j.jcrysgro.2009.06.040

For device manufacturing it will be important to know whether, although the local size distribution and linewidth remain small, the average dot size and, therefore, the PL peak position vary laterally across full wafers. We have shown previously that substantial lateral variations in both dot height and density can exist across wafers, and that these inhomogeneities are unrelated to variations in the buffer layer morphology, such as variations in the density and distribution of step edges or surface roughness [29]. The causes of the inhomogeneous dot distributions and the means for controlling them have yet to be identified.

In this study, we examined the lateral variations in dot density and height distributions of self-assembled InGaAs quantum dots across GaAs substrates and correlated these with growth conditions. We found large variations in both height and density for dots grown at relatively high rates in a pulsed mode. For these samples, the local dot height varied up to 50% of the average height and the dot density varied up to 85% of the average density across 51 mm wafers. These variations decreased as the dot density increased. They were also reduced for the top layer of dots in 10-layer stacks. Substantially improved uniformity was found for dots grown at low rates in both pulsed and continuous deposition modes. For dots deposited slowly and continuously, the largest variation in height was 20% of the average and the largest variation in density was 27%. The influence of wafer size, growth rate, deposition mode (pulsed or continuous), QD layer thickness, QD layer composition and spacing of stacked dot layers were also studied.

#### 2. Experimental procedures

InGaAs QDs, with nominally 0.45 mole fraction In, and pure InAs QDs were grown by molecular beam epitaxy (MBE) on 51 and 76 mm diameter, Si-doped,  $(100)\pm0.03^{\circ}$  epi-ready GaAs substrates. The MBE system used has been described in detail elsewhere [30]. The temperature variation across a 76 mm wafer in this system at 500 °C was estimated from optical pyrometer and IR camera measurements to be less than 5 °C. The flux uniformity was determined from X-ray measurements of In<sub>0.3</sub>Ga<sub>0.7</sub>As/GaAs superlattices (SLs) grown at the highest and lowest In growth rates used in this study on 76 mm wafers rotated at 10 rpm. For the highest growth rate (In 0.22 monolayers/s (ML/s); Ga 0.50 ML/ s) the maximum difference in SL thickness was 1.0% across the central 30 mm region and 2.0% across the central 50 mm region. The maximum change in indium composition was 0.4% across the central 30 mm region and 1.0% across the central 50 mm region. That the thickness changes more than the composition suggests that the In and Ga fluxes vary in a similar manner, as expected. Similar differences in SL thickness and indium composition were found for the lowest In growth rate.<sup>4</sup> These differences are smaller than the variations in QD height and density, which will be described below.

Prior to QD deposition, the oxide was removed from the substrates at 630 °C under flowing As<sub>2</sub> ( $P_{As2} = 8 \times 10^{-6}$  Pa, or  $6 \times 10^{-8}$  Torr), and a 500-nm-thick GaAs buffer was grown at a rate of ~1 µm/h with the wafer held at 600 °C. This procedure was previously identified as yielding smooth buffer layers, with average root-mean-square (rms) roughness of 0.2 nm [29]. The substrate was then cooled to the QD growth temperature and held

for 5 min, still with an As overpressure, to stabilize the temperature across the substrate.

Dots were grown under a variety of conditions. The deposition rate was varied from 0.069 to 0.22 ML/s for InAs<sup>5</sup> and from 0.077 to 0.89 ML/s for GaAs. The total QD layer thickness deposited ranged from 5.6 to 9.5 ML for the InGaAs dots and was 2 ML for the InAs dots. Wafer rotation speeds of 10 and 40 rpm were used. The growth temperature was varied from 495 to 550 °C and was measured at the center of the wafers by optical pyrometry. Dots were grown either continuously or in a pulsed mode, and dot formation was monitored by reflection high-energy electron diffraction (RHEED).

Pulsed mode QDs were deposited with alternating cycles of 0.25 ML In,  $\sim 5 \text{ s} \text{ As}_2$  ( $P_{\text{As}2} = 8 \times 10^{-6} \text{ Pa}$ , or  $6 \times 10^{-8} \text{ Torr}$ ), 0.31 ML Ga, and another  $\sim 5 \text{ s} \text{ As}_2$ . This sequence was repeated for a total thickness of 5.6 ML and timed relative to the wafer rotation speed, by adjusting the As<sub>2</sub> exposure time, to distribute the material uniformly across the wafer. The wafer rotated  $\sim 612^{\circ}$  for each sequence, completing 17 full rotations during dot deposition. The elapsed time for each sequence was less than 2 min. This method of deposition has been found previously to produce dots with better uniformity on a local scale [21,31].

Because of the difficulty of establishing growth rates corresponding to the nominal composition of In<sub>0.45</sub>Ga<sub>0.55</sub>As, continuously grown QDs were deposited with alternating cycles of InGaAs and GaAs. The growth is called "continuous" because As and group III atoms were deposited simultaneously with no pauses in deposition, only short pauses in the In flux for some samples. The As<sub>2</sub> pressure during the continuous growths was  $3.7 \times 10^{-4}$  Pa ( $3 \times 10^{-6}$  Torr). All samples were cooled to room temperature immediately after deposition was complete.

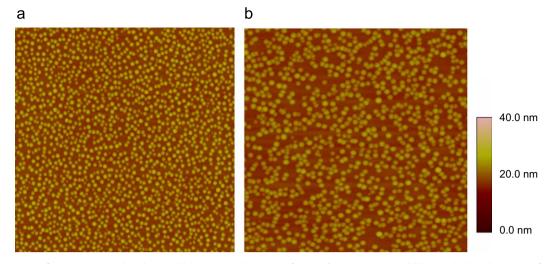
Throughout this paper, samples are labeled "F", "M" and "S", for fast  $(1.15\pm0.05 \text{ ML/s})$ , moderate  $(0.22\pm0.02 \text{ ML/s})$  and slow  $(0.13\pm0.03 \text{ ML/s})$  deposition rates, respectively. Except where noted otherwise, samples with a fast deposition rate were rotated at 10 rpm while samples with moderate or slow deposition rates were rotated at 40 rpm. The labels "P" and "C" are used to designate pulsed and continuous growths.

The QD density and height were determined from atomic force microscopy (AFM) images. Imaging was performed with commercial, pyramidal, etched Si tips in tapping mode under ambient conditions. Images were taken on a uniform array of 81 points (see Fig. 1 in Ref. [29]) which covered the central  $26 \times 26$  or  $40 \times 40 \text{ mm}^2$  regions (or  $\sim 34\%$ ) of the 51 and 76 mm diameter wafers, respectively. The scan size for the AFM images was chosen from a study of the influence of scan size on measured dot density. For  $1 \, \mu m^2$  scans it was found that four adjacent images had relatively large differences in dot density. These local variations diminished with increasing scan size, and the density of dots determined for adjacent images was found to converge at a scan size of  $3 \times 3 \, \mu m^2$ , which was used in this study.

Software which identified local maxima was used to determine the QD positions. The accuracy of the algorithm was confirmed by visual inspection of images generated with the dot positions marked. For each image the average background level was determined and subtracted from the height at each maximum, in order to determine the QD heights. This approach to background subtraction was sufficient because the average rms roughness of the buffers (0.2 nm) is similar to the smallest

<sup>&</sup>lt;sup>4</sup> For the lowest In growth rate (In 0.06 ML/s and Ga 0.14 ML/s) the maximum difference in SL thickness was 1.3% across the central 30 mm region and 2.9% across the central 50 mm region. The maximum change in indium composition was 0.4% across the central 30 mm region and 1.1% across the central 50 mm region.

<sup>&</sup>lt;sup>5</sup> The growth rate for InAs was corrected for deposition onto GaAs, which has a smaller lattice parameter. The correction used was:  $GR_{GaAs} = GR_{inAs}^*(a_{GaAs}/a_{inAs})$ , where GR is the growth rate in (ML/s) and *a* the lattice parameter. This reflects that a monolayer of lattice-matched InAs on GaAs has more atoms per area than a monolayer on InAs.



**Fig. 1.** Typical AFM images of InGaAs quantum dots deposited (a) continuously at a rate of 0.13 ML/s, specimen SC3 and (b) in a pulsed mode at a rate of 1.10 ML/s, specimen FP3, on 76 mm wafers. The images are  $3 \times 3 \mu m^2$ , and the grey scale is 40 nm. The buffer layer on all wafers was quite smooth (average rms roughness 0.2 nm), which facilitated background subtraction for determination of dot heights.

variations in dot height observed. Typical AFM images used for data analysis are shown in Fig. 1.

Although several studies have analyzed the lateral dimensions of In(Ga)As quantum dots based on AFM images [32], we have found that AFM does not give an accurate measure of the dot diameter. In addition, calculations of the influence of QD size on PL linewidth have shown fluctuations in dot height have a larger effect on spectral broadening than dot diameter [13]. Therefore, only dot heights and densities are reported.

#### 3. Results and discussion

The growth parameter found to have the largest influence on the lateral uniformity of the quantum dots was deposition rate. This is evident from plots of dot height and density for three different 76 mm wafers shown in Fig. 2. The dots in contour plots 2 (a) and (b), sample FP3, were deposited in a pulsed mode at a rate of 1.10 ML/s. The deposition rate for the dots in (e) and (f), sample SP3, was nearly an order of magnitude lower, 0.12 ML/s. The dots in (c) and (d), sample MP3, were also deposited at a moderately low rate, 0.21 ML/s. The same range for the contour lines was used for the three plots of dot height (4.5–10.0 nm) and for the three plots of dot density (80–170 dots/ $\mu$ m<sup>2</sup>), to facilitate comparison of the variations between specimens.

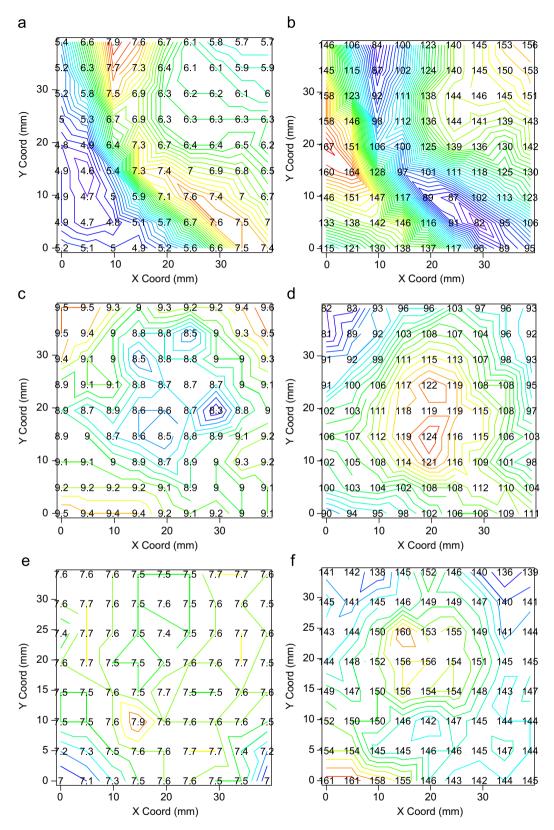
Large lateral variations are evident for the dots grown at the fastest rate, sample FP3, particularly along the diagonal between the bottom left and top right corners of the plots (Fig. 2(a) and (b)). Such large gradients in height and density were typically observed across wafers with fast deposition rates, although the orientation of the gradient varied (see Ref. [29] for more examples). Because the wafers were rotated during growth, it is expected that any temperature gradient across the wafers would be radial. Thus, the gradients in dot size and density probably resulted from transitory inhomogeneities in the material thickness during deposition, which are eliminated by the end of the growth. Since the dots nucleate before the InGaAs deposition is complete, as indicated by the appearance of chevrons in the RHEED pattern part way through the growth, there is a gradient across the wafer in the volume of material deposited at the time of nucleation. That the final InGaAs volume deposited is quite uniform across the wafer has been discussed previously [29]. This accounts for the commonly observed inverse relationship between the dot height and density, which can be seen in Fig. 3(a) and (b).

The dots deposited at low rates, samples MP3 and SP3 in Fig. 2(c) through (f), have smaller lateral variations in height and density than the dots deposited quickly, and the variations are primarily radial from the center of the wafer. Again, the lateral distributions of dot height and density are inversely correlated, similar to those in the less uniform higher deposition rate specimens, indicating that the total volume of InGaAs deposited is laterally more uniform than the dot distributions. The radial distributions suggest the influence of thermal and/or beam flux gradients, both of which can produce radial gradients on rotating wafers. As described in the experimental section, the flux in the growth system used has a maximum variation of 3%. This is significantly less than the maximum variations in the distributions of slowly deposited dots (see Tables 1 and 3 below) and suggests that temperature gradients are largely responsible for the radial distributions, which is not surprising since it is well known that QD density varies rapidly with growth temperature [22-25].

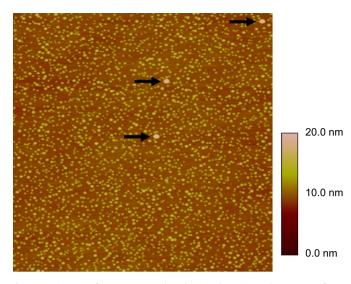
It is interesting that growth rate has such a large influence on lateral uniformity of QDs especially since, as described in the experimental section, there was no significant change in the flux uniformity with growth rate for the MBE system used. The increased lateral uniformity of dots formed at low deposition rate is, however, similar to what has been found on a local scale in previous studies where lower growth rates resulted in narrower dot size distributions [3–5].

The statistics for the three samples in Fig. 2 and also for a fourth sample, SC3, grown continuously at a low rate similar to that used for the pulsed sample, SP3 (shown in Fig. 2(e) and (f)), are given in the top of Table 1 (76 mm wafers). This table gives the average and standard deviation of the dot heights and densities from the 81 images analyzed for each sample. It also gives the absolute difference between the maximum and minimum values determined from the images, and the difference as a percentage of the average.

As expected based on the contour plots, the standard deviations and maximum differences in dot height and density across the central  $40 \times 40 \text{ mm}^2$  region of the wafer are largest for the sample with the fastest deposition rate, FP3. The standard deviation in dot height was 15% of the average height, and the standard deviation in density was 18% of the average. The



**Fig. 2.** Contour plots for dots on three wafers (76 mm diameter) grown in the pulsed mode at different rates: (a) height and (b) density of dots deposited at 1.10 ML/s, FP3; (c) height and (d) density of dots deposited at 0.21 ML/s, MP3 and (e) height and (f) density of dots deposited at 0.12 ML/s, SP3. The dots grown at the fastest rate have large variations in both height and density across the wafer along one axis (bottom left corner to top right corner of the plot). Dots grown more slowly have smaller radial variations, which are indicative of temperature gradients across the wafers. Heights are in nm and densities are in dots/µm<sup>2</sup>. The contour lines of dot height are plotted at intervals of 0.1 nm; dot density lines are at intervals of 2 dots/µm<sup>2</sup>.



**Fig. 3.** AFM image of InAs quantum dots deposited continuously at a rate of 0.06 ML/s, specimen SCIn, showing three large quantum dots amongst many much smaller dots. The image is  $3 \times 3 \,\mu\text{m}^2$ ; note the grey scale is 20 nm (unlike Fig. 1, where it is 40 nm).

#### Table 1

Influence of deposition rate and wafer size on dot uniformity.

Sample	QD h	(nm)		QD density (/µm <sup>2</sup> )						
	Avg	σ	$\sigma$ % avg	Δ	⊿ % avg	Avg	Σ	$\sigma$ % avg	Δ	⊿ % avg
76 mm wafers-central $40 \times 40$ mm <sup>2</sup> region										
FP3	6	1	15	3.3	53	126	23	18	85	67
MP3	9.0	0.3	3	1.2	14	104	10	9	43	42
SP3	7.5	0.2	2	0.9	12	147	6	4	25	17
SC3	8.8	0.4	4	1.8	20	183	6	3	26	14
51 mm wafers-central $26 \times 26 \text{ mm}^2$ region										
FP2	9.0	1.0	12	3.6	40	165	27	16	142	86
MP2	8.5	0.3	3	1.6	18	125	13	10	78	63
SC2	6.2	0.5	7.6	1.7	27	362	8	2	46	13

Sigma is the standard deviation of the 81 points used to determine the average:  $\sigma = (\sum (x - x)^2 / (n - 1))^{1/2}$ .

Delta is the difference between the maximum and minimum values of the 81 points.

Samples labeled "F", "M" and "S" had fast (1.15 $\pm$ 0.05 ML/s), moderate (0.22 $\pm$ 0.02 ML/s) and slow (0.13 $\pm$ 0.03 ML/s) deposition rates, respectively. The labels "P" and "C" designate pulsed and continuous growths.

All samples were grown at 530  $^\circ\text{C},$  except MP3 and SC2, which were grown at 480 and 520  $^\circ\text{C},$  respectively.

All samples were rotated at 40 rpm, except MP2 which was rotated at 10 rpm.

\* The large variation in QD height for sample SC2 was caused by streaks in the AFM images.

maximum differences in height and density in the central region of this wafer were 53% and 67% of the averages, respectively.

These are large variations, particularly for commercial production of optical devices. For manufacturing, consistent dot size will be required to maintain reproducible wavelength response, and consistent dot density will be necessary for reproducible device sensitivity or brightness. Based on a simple height-cubed estimate of the dot volume, we have previously calculated that a 40–50% change in dot height would translate to a ~100 meV (~124 nm) shift in energy (wavelength) response for InGaAs QDs [29]. A 60% variation in dot density would be expected to cause a roughly 60% change in device responsivity, assuming device saturation is possible.

Decreasing the growth rate by a factor of five (compare MP3 with FP3) substantially improved the height uniformity; however, the density uniformity did not change as significantly. The

Fabl	e	2	

Dot statistics for fast pulsed growths arranged in order of increasing dot density.

Sample	QD height (nm)					QD density (/µm <sup>2</sup> )				
	Avg	σ	$\sigma$ % avg	Δ	⊿ % avg	Avg	σ	$\sigma$ % avg	Δ	⊿ % avg
FP1	7.5	1.0	13	4.1	55	169	31	18	150	89
FP2	9.0	1.0	12	3.6	40	165	27	16	142	86
FP4	5.1	0.5	9	2.4	47	287	17	6	74	26
FP5	5.6	0.6	10	1.6	29	371	20	5	92	25
FP6	3.9	0.2	5	1.0	25	494	24	5	105	21
Stacked dots										
FPS10	11.6	0.4	4	2.0	18	54	3	5	16	29
FPS7	7.5	0.3	5	1.7	22	160	7	4	31	19

All samples were rotated at 10 rpm.

Stacked dots had 10 dot layers

FPS10 had 10 nm GaAs spacer layers.

FPS7 had 7 nm spacer layers.

standard deviation in density as a percentage of the average was reduced by only a factor of two, and the maximum deviation changed even less. It is surprising that the height uniformity improved more than the density uniformity when the deposition rate was decreased, considering that the dot density is directly related to the volume of the InGaAs layer deposited, while the dot height is related to the volume to the  $\frac{1}{3}$  power, assuming that the dot shape does not change across the wafer. However, this was observed for both 76 and 51 mm wafers (MP3 versus FP3 and MP2 versus FP2).

Decreasing the growth rate further, by an order of magnitude, caused a considerable enhancement of both height and density uniformity. This was true for dots grown both continuously and in a pulsed mode, SP3 and SC3.

Similar results were found for dots grown on 51 mm wafers (also shown in Table 1). Dots deposited quickly, FP2, had large standard deviations and maximum variations in height and density in the central  $26 \times 26 \text{ mm}^2$  region. As observed for the larger wafers, reducing the growth rate by a factor of five improved the height uniformity substantially, but the density uniformity changed by less than a factor of two, for sample MP2. The greatest lateral uniformity of dots, in terms of density, was obtained by depositing at a very low rate, 0.13 ML/s, for sample SC2. The larger variation in the QD height measured in this sample was caused by streaks in many of the AFM images, which artificially increased the dot height distribution. For this sample the deposition was continuous but, based on the results for the larger 76 mm wafers, we expect similar uniformity would have been obtained for pulsed growth specimens.

From the results in Table 1 it can be seen that there was no significant difference between the uniformity of dots grown on 51 and 76 mm wafers. In addition, the uniformities of dots grown at low rates were similar for both pulsed and continuous depositions (samples SP3 and SC3).<sup>6</sup> This last result differs from local scale studies which found improved QD uniformity with pulsed deposition [21,31].

For specimens with fast deposition rates and pulsed growth the dot uniformity increased with dot density. Data for specimens with a deposition rate of  $1.15\pm0.03$  ML/s and a variety of dot densities are shown in Table 2. Here, it can be seen that the standard deviations in both dot height and density were smaller

<sup>&</sup>lt;sup>6</sup> A comparison between pulsed and continuous deposition was not made for the high growth rate since it was anticipated that for continuous deposition the material distribution across the wafer would not be uniform. With no pauses in the growth, the time required to deposit the dot layer at the higher rate would have been ~5 s, in which time the wafer would have rotated less than 360°.

percentages of their respective averages in samples with higher densities, and the maximum variations in dot height and density in these samples as percentages of their averages were lower as well.

This was not the case for dots grown slowly  $(0.13 \pm 0.03 \text{ ML/s})$  and continuously. As can be seen in Table 3, the standard deviations in dot height and density as percentages of the averages for these samples were independent of dot density and small in all cases, ranging from 2% to 4%. The maximum variations as a percentage of the average were also independent of dot density and relatively small,  $\leq 27\%$ . For these samples the variations in height and density, as measured by the standard deviation and the maximum difference, were always smaller fractions of the average than those in samples with fast deposition rates (compare  $\sigma\%$  avg and  $\varDelta\%$  avg in Tables 2 and 3).

Dot composition had a negligible effect on the lateral dot uniformity. Statistics for a specimen with pure InAs dots, deposited slowly and continuously at 500 °C, are given at the bottom of Table 3, sample SCIn. The deposition rate for these dots was 0.06 ML/s, roughly half that of the InGaAs dots, because only one group III element was being deposited. From the statistics it is apparent that the lateral uniformities of the dot height and density in this sample are quite similar to those of the InGaAs dots deposited slowly.

This was true in spite of a greater dispersion in the size of the InAs dots on a local scale. AFM images of specimen SCIn reveal a low density of large dots (see Fig. 3), which are approximately 4 times as tall as the other InAs dots, and occur less frequently than

Table 3

Dot statistics for slow continuous growths arranged in order of increasing dot density.

Sample	QD height (nm)					QD density (/µm <sup>2</sup> )					
	Avg	σ	$\sigma$ % avg	Δ	⊿ % avg	Avg	σ	$\sigma$ % avg	Δ	⊿ % avg	
SC1	10.2	0.4	4	1.7	17	114	3	3	13	11	
SC4	7.9	0.2	3	0.9	11	134	4	3	21	16	
SC3	8.8	0.4	4	1.8	20	183	6	3	26	14	
SC40	7.4	0.2	2	0.9	12	277	9	3	42	15	
SC10	6.4	0.2	3	1.0	16	286	10	3	44	16	
SC5	8.3	0.1	2	1.0	13	339	14	4	92	27	
SC6	7.8	0.1	2	0.8	10	366	16	4	90	24	
SC7	7.1	0.3	4	1.3	18	368	15	4	70	19	
InAs dot	s										
SCIn <sup>a</sup>	4.7	0.1	2	0.5	10	334	16	5	67	20	

All samples were rotated at 40 rpm, except SC10 which was rotated at 10 rpm. <sup>a</sup> Pure InAs dots. 1 per 1000 dots. Histograms of all the measured dot heights for sample SCIn and sample SC5, which has a similar density of InGaAs dots, are shown in Fig. 4. The number of dots at each height is plotted on a log scale so that the small number of large dots on sample SCIn is visible. Previous studies have also found pure InAs QDs to be less uniform on a local scale than InGaAs dots [28] and to have multimodal distributions on a local scale [33]. It is interesting that, in spite of the greater local variations in InAs dot size, the lateral uniformity is similar to that of InGaAs QDs. That the uniformity does not vary with dot composition also suggests that In re-evaporation was not an issue for these samples.

Stacking dots was found to improve the uniformity of the top layer of dots in a set deposited quickly (1.15 ML/s) in a pulsed mode. The statistics for the top layer of dots on two wafers with 10 dot layers are given at the bottom of Table 2. The samples, FPS7 and FPS10, had 7- and 10-nm-thick GaAs spacer layers between the InGaAs dot layers, respectively. For both samples the standard deviations in the height and density of dots in the top layer are 4–5% of the average. This is quite close to the variations found for single layers of dots deposited slowly and continuously (see Table 3), and substantially better than that of single layers deposited quickly. This improved lateral uniformity of the top layer is consistent with previous observations of enhanced uniformity of stacked dots on a local scale [34,35].

Also in agreement with local scale studies of stacked dots [34,35], the top layer of dots in both of the stacked wafers had lower dot densities than a single layer of dots grown under nominally the same conditions, specimen FP4. In addition, the dot density decreased as the thickness of the spacer increased. As described earlier, the height and density uniformity of dots deposited quickly in a pulsed mode improved with increased density. However, in this case the dots in the top layer of a stack have lower densities but better uniformity than a single dot layer deposited under similar conditions (compare with FP4), and they have vastly better uniformity than single dot layers with similarly low densities (compare, for example, with FP1 and FP2). The uniformity of dots grown slowly may also be improved by stacking, however, this was not examined.

Under the growth conditions studied, the number of monolayers of InGaAs deposited was found to affect the dot height but not the dot density. This was most clearly demonstrated by specimens SC2 (Table 1) and SC7 (Table 3). These samples were grown under nominally identical conditions, except with different amounts of InGaAs, 6.8 and 9.5 ML, respectively, yet their dot densities were nearly identical, 362 and 368  $\mu$ m<sup>-2</sup>. For these two wafers there was less than a 2% difference in dot density for a 40% change in the amount of InGaAs deposited.

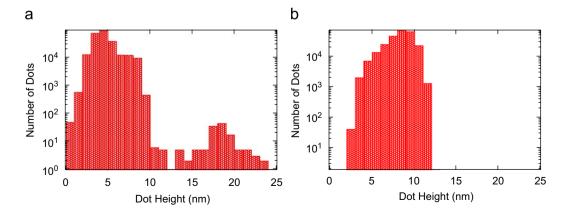


Fig. 4. Histograms of all the dot heights from 81 AFM images of samples (a) SCIn and (b) SC5. The data are plotted on a log scale to make the small number of very tall dots in sample SCIn visible.

From this it appears that the density of dots was determined earlier in the growth process, when fewer monolayers of InGaAs had been deposited, and that it reached a saturation value (for the given set of growth conditions, including temperature, flux, etc.) before 6.8 ML of InGaAs were deposited. As more material was added the existing dots grew larger, but new dots did not form. That new dots were not nucleated as more material was deposited is consistent with the fairly narrow dot size distributions found even for tall dots (see statistics for SC1 SC3 and SC5). Similar results have been obtained in studies of InAs QDs [25,36]. In those studies, the dot density increased rapidly as a function of InAs coverage up to  $\sim 1.75$  or 2 ML. For greater InAs coverage the density of the ODs was saturated and independent of the amount of InAs deposited. We expect saturation to occur at greater coverage for InGaAs dots, because of the reduced strain and increased wetting layer thickness compared with those for InAs dots.

On a local scale ( $\sim 0.5 \,\mu\text{m}^2$ ) it has been found that 2 ML of InAs grown at a very low rate ( $\leq 0.01 \text{ ML/s}$ ) produced QDs that are larger and more uniform in size but lower in density than 2 ML grown at 0.1 ML/s [3]. Similar narrowing of InAs QD size distributions, on a local scale, for growth rates  $\leq 0.01 \text{ ML/s}$  has been observed in other studies as well [4,5]. It is possible that the lateral uniformity of QDs could also be improved further by growing at even lower rates than the lowest rate (0.12 ML/s) used in this study; however, it is anticipated that temperature and flux gradients will become limiting factors.

#### 4. Summary

Deposition rate had a large influence on lateral uniformity of QDs. InGaAs quantum dots deposited at low rates had the smallest lateral variations in size and density across 51 and 76 mm diameter wafers, for both continuous and pulsed growth modes. As the deposition rate was increased, from 0.13 to 1.15 ML/s, the lateral dispersion in dot height and density also increased. Dots grown slowly had radial distributions, probably caused by gradients in temperature and/or flux across the wafers. For dots deposited quickly, the variations were diagonal or lateral rather than radial and they decreased with increasing dot density. The uniformity also improved for the top layer of stacked dots grown at a high rate, even though these had low densities. For dots with fast growth rates, even the most uniform specimens had larger lateral dispersion than dots grown slowly. The uniformity of dot distributions on 51 and 76 mm diameter wafers was similar. Changing the dot composition from 0.45 to 1.0 mole fraction In did not affect the lateral uniformity of dots deposited slowly, even though the InAs dots were less uniform on a local scale.

#### Acknowledgement

The authors thank Kris Bertness for helpful discussions and assistance with measurements of temperature variation across wafers.

#### References

- [1] Q. Xie, A. Madhukar, P. Chen, N.P. Kobayashi, Phys. Rev. Lett. 75 (1995) 2542.
- [2] V.-G. Stoleru, D. Pal, E. Towe, Physics E 15 (2002) 131.
  [3] R. Murray, D. Childs, S. Malik, P. Siverns, C. Roberts, J.M. Hartmann, P.
- Stavrinou, Jpn. J. Appl. Phys. Part 1 38 (1999) 528.
- [4] Y. Nakata, K. Mukai, M. Sugawara, K. Ohtsubo, H. Ishikawa, N. Yokoyama, J. Cryst. Growth 208 (2000) 93.
- [5] P.B. Joyce, T.J. Krzyzewski, G.R. Bell, T.S. Jones, S. Malik, D. Childs, R. Murray, Phys. Rev. B 62 (2000) 10891.
- [6] K. Yamaguchi, K. Yujobo, T. Kaizu, Jpn. J. Appl. Phys. Part 2 39 (2000) L1245.
   [7] I. Mukhametzhnov, Z. Wei, R. Heits, A. Madhukar, Appl. Phys. Lett. 75 (1999) 85
- [8] S. Saravanan, H. Shimizu, J. Cryst. Growth 289 (2006) 14.
- [9] F. Patella, M. Fantoni, F. Arciprete, S. Nufris, E. Placidi, A. Balzarotti, Appl. Phys. Lett. 78 (2001) 320.
- [10] Y. Sugiyama, Y. Nakata, T. Futatsugi, M. Sugawara, Y. Awano, N. Yokoyama, Jpn. J. Appl. Phys. Part 2 36 (1997) L158.
- [11] Q. Xie, A. Madhukar, P. Chen, N.P. Kobayashi, Phys. Rev. Lett. 75 (1995) 2542.
- [12] G.S. Solomon, J.A. Trezza, A.F. Marshall, J.S. Harris, Phys. Rev. Lett. 76 (1996) 952
- [13] A. Endoh, Y. Nakata, Y. Sugyama, M. Takatsu, N. Yokoyama, Jpn. J. Appl. Phys. Part 1 38 (1999) 1085.
- [14] I. Mukhametzhanov, R. Heitz, J. Zeng, P. Chen, A. Madhukar, Appl. Phys. Lett. 73 (1998) 1841.
- [15] S. Krishna, J. Sabarinathan, K. Linder, P. Bhattacharya, J. Vac. Sci. Technol. B 18 (2000) 1502.
- [16] E.C. Le Ru, P. Howe, T.S. Jones, R. Murray, Phys. Rev. B 67 (2003) 165303.
- [17] Z. Mi, P. Bhattacharya, J. Appl. Phys. 98 (2005) 023510.
- [18] G.S. Solomon, Appl. Phys. Lett. 84 (2004) 2073.
- [19] Zh. Wang, Sh. Seydmohamadi, J.H. Lee, G.J. Salamo, Appl. Phys. Lett. 85 (2004) 5031.
- [20] M. Schmidbauer, Sh. Seydmohamadi, D. Grigoriev, Zh.M. Wang, Yu.I. Mazur, M. Hanke, R. Köhler, G.J. Salamo, Phys. Rev. Lett. 96 (2006) 66108.
- [21] R.P. Mirin, J.P. Ibbetson, K. Nishi, A.C. Gossard, J.E. Bowers, Appl. Phys. Lett. 67 (1995) 3795.
- [22] K. Shiramine, T. Itoh, S. Muto, T. Kosaki, S. Sato, J. Cryst. Growth 242 (2002) 332.
   [23] O. Suekane, S. Hasegawa, M. Takata, T. Okui, H. Nakashima, Mater. Sci. Eng. B
- [23] U. Suekane, S. Hasegawa, M. Takata, T. Ukui, H. Nakashima, Mater. Sci. Eng. B 88 (2002) 158.
   [24] H. Gaira, K. Nichi, S. Sugay, Appl. Divis. Lett. 74 (1000) 1224.
- [24] H. Saito, K. Nishi, S. Sugou, Appl. Phys. Lett. 74 (1999) 1224.
- [25] T.J. Krzyzewski, P.B. Joyce, G.R. Bell, T.S. Jones, Phys. Rev. B. 66 (2002) 201302.
   [26] P.B. Joyce, T.J. Krzyzewski, G.R. Bell, T.S. Jones, S. Mailk, D. Childs, R. Murray, Phys. Rev. B. 62 (2000) 10891.
- [27] J.-I. Chyi, Mater. Sci. Eng. B 75 (2000) 121.
- [28] H. Li, Q. Zhuang, Z. Wang, T. Daniels-Race, J. Appl. Phys. 87 (2000) 188.
- [29] A. Roshko, T.E. Harvey, S.Y. Lehman, R.P. Mirin, K.A. Bertness, B.L. Hyland, J. Vac. Sci. Technol. B 23 (2005) 1226.
- [30] T.E. Harvey, K.A. Bertness, R.K. Hickernell, C.M. Wang, J.D. Splett, J. Cryst. Growth 251 (2003) 73.
- [31] D.L. Huffaker, D.G. Deppe, Appl. Phys. Lett. 73 (1998) 520.
- [32] S.-K. Choi, J.-M. Jang, S.-H. Yi, J.-A. Kim, W.-G. Jung, Proc. SPIE 6831 (2008) 683119.
- [33] H. Kissel, U. Müller, C. Walther, W.T. Masselink, Y.I. Mazur, G.G. Tarasov, M.P. Lisitsa, Phys. Rev. B 62 (2000) 7213.
- [34] J. Tersoff, C. Teichert, M.G. Lagally, Phys. Rev. Lett. 76 (1996) 1675.
- [35] B. Lita, R.S. Goldman, J.D. Phillips, P.K. Bhattacharya, Appl. Phys. Lett. 74 (1999) 2824.
- [36] F. Patella, F. Arciprete, M. Fanfoni, V. Sessi, A. Balzarotti, E. Placidi, Appl. Phys. Lett. 87 (2005) 252101.