

## BRIEF REPORTS AND COMMENTS

This section is intended for the publication of (1) brief reports which do not require the formal structure of regular journal articles, and (2) comments on items previously published in the journal.

### Effect of surface pretreatment and substrate orientation on the characteristics of InAs quantum dots on Si and SiO<sub>2</sub> substrates

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The surface morphology of InAs quantum dots (QDs) on undoped Si (100) shows a strong dependency on surface pretreatments, with as much as 30% difference in island density with different size distributions. Lowering the V/III fluxes ratio (58:1 to 10:1) effectively suppresses the average height (47%) and increases density (75%) of QDs. While changing the growth temperature (220–300 °C) increased the average height by 62% on chemically etched Si surfaces, nucleation on the SiO<sub>2</sub> surface was less sensitive to surface preparation. The authors also observe that the specific substrate orientation and annealing at 500 °C do not significantly impact the morphology of QDs. © 2007 American Vacuum Society. [DOI: 10.1116/1.2739568]

#### I. INTRODUCTION

InAs quantum dots (QDs) on Si substrates have potential applications such as Si-based optoelectronic devices.<sup>1–3</sup> But exploiting Si substrates has not been vigorously pursued in comparison to the growth of InAs QDs on GaAs substrates. In addition to the greater lattice mismatch between InAs and Si (11%) in comparison to InAs and GaAs (7%), one difficulty with growth on Si is the effective *in situ* removal of the Si oxide prior to growth. The Si native silicon oxide can be removed by a high temperature treatment above 850 °C.<sup>4</sup> In addition, there are some chemical treatments reported that may decrease the temperature of native oxide removal and decrease surface contaminations.<sup>4–6</sup> The purpose of this study is to advance the understanding of the impact of surface preparation and termination on the nucleation of InAs QDs on Si and SiO<sub>2</sub> substrates, which will provide a sound foundation for development of devices and their optimization.

We present the characteristics of InAs QDs on Si with different chemical pretreatments, substrate orientations, V/III fluxes ratios, temperatures of substrate during growth, and annealing conditions after QD formation. Additional comparisons of InAs QD formation on a SiO<sub>2</sub> terminated Si substrate will be made to understand the nucleation and nanoscale transport that govern the evolution of the InAs film. All growths were carried out using a Riber 2300 solid source molecular beam epitaxy MBE (SSMBE) equipped with a valved cracker source for arsenic. High resolution atomic

force microscopy (AFM) measurements and image analyses are used for characterization of the resultant QD size distribution.<sup>6</sup>

#### II. CHEMICAL PRETREATMENT EFFECT

##### A. Chemical pretreatments, deoxidation, and growth conditions

Four different chemical pretreatments (Table I) were applied to undoped Si (100) wafers. Samples were loaded in the introduction chamber of Riber 2300 solid source MBE (SSMBE) system within 1 h. After outgassing in the introduction chamber for 1 h at 200 °C, samples were moved to growth chamber which was kept at a pressure of 10<sup>-11</sup> torr before deoxidation. The thermal deoxidation temperature (~720 °C) was determined by the observation of clear 2 × 1 reflection high-energy electron diffraction (RHEED) patterns, which were saturated in 10 min at the temperature. After deoxidation, samples were cooled down to a growth temperature of 290 °C, then soaked only with tetramer arsenic beam for 1 min. At the end of the soaking, indium shutter was opened for the following 12 s, while the beam flux ratio (V/III) was 45:1. The temperature was lowered to room temperature immediately after closing shutters.

##### B. Deoxidation results

During the exposure in open air between the chemical pretreatment and loading, the growth of a very thin native oxide layer on surface is hard to avoid. Using spectroscopic ellipsometry (SE), we found that thickness of the native oxide is ~1 nm without chemical etching and ~0.5 nm with

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TABLE I. Summary of chemical pretreatments.

[Trichloroethylene, 5 min → acetone, 5 min → methanol, 5 min → DIW rinsing, 2 min] then:	
(1)	HF 1%, 4 min → DIW rinsing, 2 min
(2)	HF 1% + HCl, 4 min → DIW rinsing, 2 min
(3)	[{H <sub>2</sub> O <sub>2</sub> :H <sub>2</sub> SO <sub>4</sub> (3:5), 1 min} + {HF:H <sub>2</sub> O(1:10), 1 min}] × 3 → HF dipping, 4 min <sup>a</sup>
(4)	HNO <sub>3</sub> (98 °C, 10 min) → 4H <sub>2</sub> SO <sub>4</sub> + 1H <sub>2</sub> O <sub>2</sub> + 0.01% HF(90 °C, 10 min) → DIW rinsing, 2 min <sup>b</sup>

<sup>a</sup>Reference 7.<sup>b</sup>Reference 5.

treatment 1 (see Table I) before thermal deoxidation. We also confirmed our thermal deoxidation temperature ( $\sim 720$  °C) in separate experiments by increasing the temperature up to 1150 °C (pyrometer reading) and observing no change in the RHEED pattern. This is lower than previously reported values, and there are only  $\pm 25$  °C difference between samples with different chemical pretreatments.

### C. AFM results

Very dense QDs were observed for all treatments: (1)  $1.71 \times 10^{11}$ , (2)  $1.57 \times 10^{11}$ , (3)  $1.44 \times 10^{11}$ , and (4)  $1.89 \times 10^{11}$  cm<sup>-2</sup>. The shapes of the dots were isotropic hemispheres, but height distributions showed differences, as in Fig. 1. The maximum difference in average height was  $\sim 70\%$  between treatments (2) and (4).

### III. SUBSTRATE (*hkl*) EFFECT

Si (100), (111), (211) wafers were cleaned similar to (1) in Table I; the HF 1% dip was 2 min for this set. After outgassing in the introduction chamber, samples were heated up to 800 °C in the growth chamber for deoxidation. Clear unchanging RHEED patterns for each (*hkl*) were observed at 800 °C over 10 min. Once the temperature was lowered to 230 °C, a dimeric arsenic beam soaked the substrates for

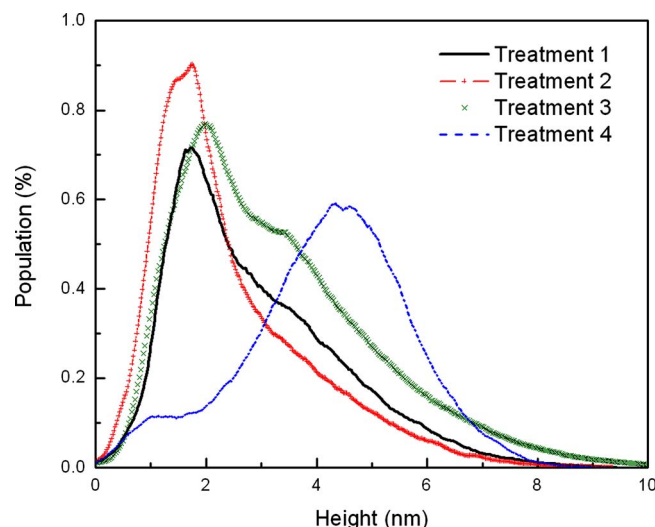
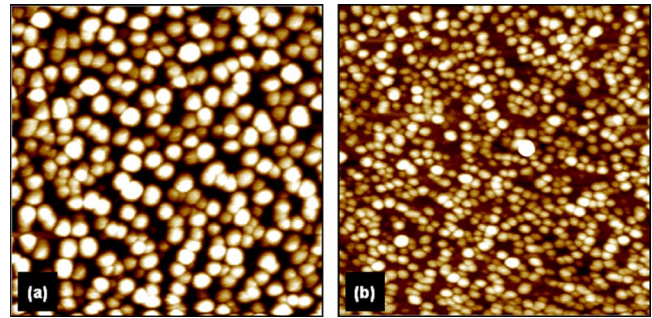


Fig. 1. AFM height distributions for different chemical pretreatments.

Fig. 2. AFM images of InAs quantum dots on Si (211): using flux ratio (V/III) (a) 58:1 and (b) 10:1 (scan area=500×500 nm<sup>2</sup>, scale is 5 nm).

2 min. Following this the indium shutter was opened for 15 s with a beam flux ratio (V/III) of 58:1. AFM measurements showed dense hemispherical QDs for all (*hkl*), with no significant difference in size and density among the samples.

### IV. BEAM FLUX RATIO (V/III) EFFECT

Two Si (211) wafers were prepared as described in Sec. III. One sample was grown with a beam ratio (V/III) of 58:1, and the other one was grown with 10:1. AFM analysis showed a 47% decrease in average height and a 75% increase in density with lowering the V/III ratio (see Fig. 2).

### V. GROWTH TEMPERATURE EFFECT

A set of samples cut from a Si (100) wafer with an intentionally grown, high-quality SiO<sub>2</sub> layer (thickness  $\sim 1$  μm) was cleaned with trichloroethylene, acetone, and methanol for 5 min each, with a final de-ionized (DI) water rinse. Additional chemical etching with HF 10% for 2 min followed by another DI water rinse was applied to another set of samples from the same wafer. Two pieces of Si wafer—one with SiO<sub>2</sub>, the other with a final HF etch—were mounted on one block in order to apply the same growth conditions on both surfaces. Samples were outgassed in the introduction chamber, then heated to the growth temperature in the growth chamber without a thermal deoxidation step. Two different growth temperatures, 220 and 300 °C, were applied with 2 min of dimeric arsenic soaking plus 90 s of growth under a beam ratio (V/III) of 29:1. Similar dense hemispherical InAs QDs were grown on both surfaces. The average heights were found on the SiO<sub>2</sub> surface to be 34% lower for 220 °C and 61% lower for 300 °C. With higher growth temperature, average heights were increased by 62% for the HF etched surface and 35% for the SiO<sub>2</sub> surface.

### VI. ANNEALING EFFECT

The same sample preparation and growth conditions at 300 °C previously from Sec. V were applied to some samples with an additional annealing step under an overpressure of dimeric arsenic. Annealing for 5 min at 460 or 500 °C did not affect the morphology of the InAs QDs.

## VII. SUMMARY AND CONCLUSIONS

*Ex situ* chemical surface preparations were applied and compared in this study in order to lower the thermal deoxidation temperature and study its effect on epitaxy. SE (spectroscopic ellipsometry) revealed the thicknesses of the native oxide on the silicon surface with and without chemical pretreatment,  $\sim 10$  and  $\sim 5$  Å, respectively. Interestingly, the density and average height of QDs were changed by as much as 31% and 70% as a consequence of the pretreatment.

We found that the orientation of the substrates (*hkl*) did not significantly affect the QD properties while different beam flux ratios (V/III) changed the average height (47%) and density (75%) significantly. The growth temperature had more impact on the results for the chemically etched Si surfaces than for the SiO<sub>2</sub> surface, which produced smaller QDs for all temperature conditions.

Investigations of the electrical and optical characteristics of the QDs for different growth conditions are currently underway.

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<sup>1</sup>T. Mano, H. Fujioka, K. Ono, Y. Watanabe, and M. Oshima, *Appl. Surf. Sci.* **130–132**, 760 (1998).

<sup>2</sup>L. Hansen, F. Bensing, and A. Waag, *Jpn. J. Appl. Phys., Part 1* **38**, 6219 (1999).

<sup>3</sup>G. E. Cirlin *et al.*, *Mater. Phys. Mech.* **1**, 15 (2000).

<sup>4</sup>A. Ishizaka and Y. Shiraki, *J. Electrochem. Soc.* **133**, 666 (1986).

<sup>5</sup>H. Okumura, T. Akane, Y. Tsubo, and S. Matsumoto, *J. Electrochem. Soc.* **144**, 3765 (1997).

<sup>6</sup>P. E. Thompson, M. E. Twigg, D. J. Godbey, K. D. Hobart, and D. S. Simons, *J. Vac. Sci. Technol. B* **11**, 1077 (1993).

<sup>7</sup>Z. M. Zhao, O. Hul'ko, H. J. Kim, J. Liu, B. Shi, and Y. H. Xie, *Thin Solid Films* **483**, 158 (2005).