Impact of template variations on shape and arrangement of Si/Ge quantum dot arrays

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Templated self-assembly allows the fabrication of quantum dot (QD) arrays for use in nanoelectronic devices. Here, we show the strong dependence of the shape and arrangement of QDs on the template structures. Arrays of etched pits are patterned on Si (100) substrates by extreme ultraviolet interference lithography on which Si/Ge layers are grown in a molecular beam epitaxy system. Single Ge dome clusters or quantum molecules consisting of four Ge hut clusters are obtained by a change of the pit diameter. Both arrays exhibit a narrow size distribution and exact alignment of the dots. In addition, multiple stacking of these arrays is demonstrated. © 2008 American Institute of Physics. [DOI: 10.1063/1.2907196]

Several experiments have demonstrated that templated self-assembly is an efficient and attractive method to create exactly aligned quantum dot arrays (QDAs) with narrow size distribution.^{1–4} Such tailored QDAs are proposed for the development of nanoelectronic devices, such as quantum dot (QD) based field effect transistors,^{5,6} or for implementation in the fields of quantum computation⁷ and spintronics.⁸ QDAs have to be differently designed and manufactured for different applications. Especially, position, periodicity, size, and shape of the QDAs have to be controlled in order to ensure external addressability and identical electronic properties of the QDs. In the templated self-assembly method, pits are etched into the Si surface which then act as nucleation sites during the growth of QDs in an epitaxial deposition process. The control over position, size, and shape of the QDs can be achieved by tuning the growth parameters, such as temperature, amount of material, or deposition rates, as well as the lithography parameters, i.e., periodicity, size, and shape of the created pits. The two fabrication steps, lithography and deposition, are linked because the self-assembly process that occurs during deposition is directly influenced by the template, e.g., larger pit distances imply larger area for collecting atoms by a single pit. Therefore, bigger dots will be observed for patterns with larger periodicities by identical material deposition (assuming that the migration length of the adatoms is larger than the pit distance.⁹). Furthermore, some applications need special arrangements of the QDs, such as building blocks of QD cellular automata.¹⁰ Such QD molecules (QDMs) can also be fabricated by templated self-assembly.^{11,12}

In order to be able to control critical properties of QDs, such as size, shape, and position, it is required to understand the impact of template variations on their growth kinetics. In this letter, we characterize the influence of such variations on the deposition of Si/Ge QDs. A shape and arrangement transition of the QDAs from equally distributed dome islands into QDMs with four hut clusters per prepatterned pit is

found. We discuss the reason for the different alignments. Furthermore, we show that such special dot arrangements can be conserved in vertically stacked Si/Ge multilayers.

The templated Si(100) substrates are prepared by extreme ultraviolet interference lithography (EUV-IL) using the XIL-beamline at the Swiss light source.^{13,14} The EUV-IL technique is based on multiple beam interference of EUV light diffraction masks. Compared to sequential, time-consuming lithography methods, such as e-beam or focused ion beam, EUV-IL allows to create patterns with areas of $2 \times 2 \text{ mm}^2$ within seconds. Our EUV-IL setup has been proven to be capable of fabricating structures with periodicities of less than 30 nm. The created patterns have excellent uniformity, which is a basic requirement for the fabrication of tailored QDAs. For our experiments two-dimensional (2D) pit patterns with a periodicity of $280 \text{ nm} \times 280 \text{ nm}$ were exposed on polymethyl methacrylate (PMMA) coated Si(100) substrates. Template variations were created by using different exposure doses. Subsequently, the PMMA is developed and the pattern is etched into Si via reactive ion etching using CHF_3 , SF_6 , and O_2 . In the next step, the pattern is carefully cleaned with 2:1 $\text{H}_2\text{SO}_4:\text{H}_2\text{O}_2$, deionized water, and a final HF dip to deoxidize the surface. Inside the molecular beam epitaxy (MBE) chamber, the sample is moderately annealed at 500°C for 5 min to prevent smoothening—or even planarization—of pits by thermally activated diffusion of Si atoms. Subsequently, a 50 nm thick Si buffer is grown at 300°C using a growth rate of 0.05 nm/s. Two of the created patterns after deposition of the 50 nm Si buffer are shown in Figs. 1(a) and 1(b). The patterns differ only in the used exposure dose. Clearly, a broadening of the prepatterned pits is observable for the pattern created by a higher exposure dose. In the investigated dose range, a variation of the pit width from 54 to 220 nm is found. Furthermore, the shape of the pits change from round to square with edges along the $[100]$ direction by increasing the exposure dose. These variations significantly affect the profiles of the pits also after the deposition of the 50 nm Si buffer layer. The line scans of Figs. 1(a) and 1(b) (along the red arrows) show that for a lower dose, the pits have V-shaped profiles with (10n) faceted sidewalls, $5 \leq n \leq 7$.

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FIG. 1. (Color online) (a), (b) $1 \times 1 \mu\text{m}^2$ AFM scans of EUV-IL pit patterns with a periodicity of 280 nm after 50 nm Si buffer growth. The patterns differ in the EUV-IL exposure dose: (a) lower dose and (b) higher dose. Overgrowth of the pattern in (a), (b) with 7 ML of Ge results in the formation of (c) QDA with separated dome clusters and (d) QDM with four Ge huts per pit, respectively. (e) Size and height distributions of the QDAs are shown in (c) and (d).

For higher doses, they become truncated with large (001) bottom facets and steeper sidewalls.

The topography after Ge deposition is shown in the atomic force microscopy (AFM) images of Figs. 1(c) and 1(d). For the pattern created with a lower dose, the formation of a multifaceted, single dome cluster per pit is found by the deposition of 6 ML Ge while ramping the temperature from 300 to 500 °C during growth [Fig. 1(c)]. Figure 1(e) shows that the QDA exhibits a remarkably narrow size and height distribution with a mean diameter of $77.8 \text{ nm} \pm 1.2 \text{ nm}$ and a mean height of $9.35 \text{ nm} \pm 0.63 \text{ nm}$. [The distributions in Fig. 1(e) were quantified from several AFM images of the patterned area and a total of approximately 1000 dots were measured for each distribution.] The nucleation at the pit bottom is caused by capillarity driven diffusion of adatoms on the sidewalls into the V-shaped pits,¹⁵ favored by the evolved morphology of the Ge wetting layer on the pit sidewalls.¹⁶ Although the Ge domes are widely separated from each other (the QDA has a dot density of only $1.6 \times 10^9 \text{ cm}^{-2}$), no dots are nucleated outside the pits, which demonstrates an efficient collection of Ge atoms by the prepatterned pits.

FIG. 2. (Color online) (a), (b) The QDAs in Figs. 1(c) and 1(d) were additionally stacked by 5 periods/ 10 periods of Si/Ge layers, respectively (see text for more information about the recipe). The symmetry of the first layer is retained after stacking and a flattening of the Si matrix is observable.

The second pattern [in Fig. 1(b)] fabricated with a higher EUV-IL exposure dose was overgrown with 7 ML of Ge while ramping the temperature from 300 to 460 °C. The deposition leads to the formation of QDMs which consist of four (105) faceted hut clusters per prepatterned pit [Fig. 1(d)] and have the proposed symmetry of a building block for a QD cellular automata.¹⁰ It shows that the dot density can be increased, maintaining the same mask periodicity, by only changing the exposure dose. Consequently, each Ge dot is built up by a smaller number of Ge atoms which is sufficient for Ge hut clusters, but no Ge dome clusters are found. Here, the QDs have a mean diameter of $57.6 \text{ nm} \pm 3.5 \text{ nm}$ and a mean height of $4.6 \text{ nm} \pm 0.4 \text{ nm}$ [Fig. 1(e), in total approximately 4000 dots were measured for the distributions]. It should be noted that the pits formed by the process using the higher EUV-IL exposure dose have a shape of inverted truncated pyramids after buffer layer deposition. The AFM and transmission electric microscope (TEM) [bottommost Ge layer in the TEM image of Fig. 3(b)] investigations indicate that the dots always nucleate at the favored triple intersection of pit sidewalls and the flat (001) bottom facet. QDM formation with five dots per hole is reported in Ref. 12, where the authors found the nucleation of a single dot at the bottom of a pit having the shape of a rather large and deep inverted pyramid for a small amount of Ge deposited. Increasing the Ge amount resulted in the formation of four additional dots located at the pit sidewalls around the initial Ge island.

In additional experiments, we were able to show that the distance between the QDMs (and hence, the inner dot distance of a QDM) can be tuned from 70 to 200 nm by varying the exposure dose. The transition from a QDM with four dots

FIG. 3. (a), (b) Cross-section z -contrast TEM images of 6 period/ 11 period QD stacks deposited on lower/higher exposed EUV-IL patterned Si substrates, respectively.

per pit to a single dot per pit is not abrupt. For example, an intermediate state with two dots per pit is found for patterns created with a medium exposure dose.

Our results demonstrate that QDM arrangements can be obtained by the control of the size and shape of the prepatterned pits. By changing the template structures—from small round pits with a V-type profile to larger squarelike pits with flat bottoms—we obtained a transition of the dot arrangement from a single dot per pit to a QDM.

In addition to primary Si buffer and seeded dot layer the samples were overgrown by stacks of Si/Ge bilayers in order to transfer the 2D dot arrays into a 3D dot arrangement. The pattern of Fig. 1(c) was additionally overgrown by 5 periods of 4 ML Ge at 500 °C separated by 10-nm-thick Si spacer layers while ramping the temperature from 300 to 500 °C. The pattern of Fig. 1(d) was capped by 10 periods of 5 ML Ge at $T=500$ °C separated by 10-nm-thick Si layers at $T=300$ –500 °C. As in a Si/Ge multilayer, the critical thickness of the Ge wetting layers in the second and following period is smaller with respect to the initial Ge layer,¹⁷ a lower Ge amount was used. The AFM scans in Figs. 2(a) and 2(b) taken from the topmost Ge layer show that both arrangements, separated dome clusters as well as QMs, are retained by the Si/Ge stacking. The observed vertical stacking of the dots is induced by strain fields of the underlying Ge dots.¹⁸ Again, a narrow size and height distribution is found for both patterns. The prepatterned pits are not visible anymore after multilayer growth because the growth conditions of the spacer layer cause a flattening of the Si surface, as already observed previously for more densely packed arrays of QDs.² The complete flattening occurs after several periods of Si/Ge layers (see below). Therefore, a slight unevenness (topography) of the Si is still recognizable in Fig. 2(a) (only 6 pairs of Si/Ge layers).

Figures 3(a) and 3(b) show cross sectional TEM images in z -contrast (the dots appear bright in the image) of the created QD stacks. An exact vertical stacking of the Ge dots is found. The cross section was prepared along the $[110]$ direction. Therefore, the observed period is $(280 \times \sqrt{2})$ nm. In the first layer, the nucleation of the QDs in the pits [Fig. 3(a)] and at the intersection of pit sidewalls and flat bottom facet [Fig. 3(b)], respectively, is directly observable. Please note that the dots in the first layer are somewhat bigger because of the higher amount of deposited Ge. Figure 3(b) shows that the dots in the second to seventh layer are still affected from the modulated Si matrix. A slight decrease of the dot diameter by simultaneous increase of the dot height is detectable with an increasing number of Ge layers. From the eighth layer to the top, the Si matrix is completely flattened and a uniform shape of the dots is found. Small Ge agglom-

erations between the dot stacks are found (indicated by black arrows). They could originate from the incomplete flattening of the Si surface, which causes an effective displacement of the intersection of sidewall and bottom facets toward the middle of the pit. These intersection sites serve as further nucleation sites for the Ge adatoms.

In summary, our results demonstrate the importance of template structure on self-assembly processes of QDs. Template variations influence the shape, arrangement, and density of Si/Ge QDAs, which gives us further opportunities for the creation of tailored QDAs. It is worth mentioning that together with the “ability to control” the QD self-assembly process comes the “need to control” the self-assembly process. Therefore, a robust and reproducible lithography process is required in order to be able to obtain the desired QDs.

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