Achieving quantum dot self-assembly at precise pre-defined locations is of vital interest. In this work, a novel physical method for producing germanium quantum dots on silicon using nanoindentation to pre-define nucleation sites is described. Self-assembly of ordered $\sim$10 nm height germanium quantum dot arrays on silicon substrates is achieved. Due to the inherent simplicity and elegance of the proposed method, the results describe an attractive technique to manufacture semiconductor quantum dot structures for future quantum electronic and photonic applications.

Quantum dots (QDs) are one of most promising candidates for use in solid-state quantum computation. In addition, QDs are used in a plethora of photonic and electronic applications. However, in order to achieve any scalable integration into Si based devices, QD ordering is an inevitable requirement$^{1-3}$. The current techniques for fabricating Ge-on-Si QDs either offer unsystematic nucleation, or costly non-efficient methods for defined QDs. Methods that use reactive ion etching or chemical etching have shown to be often cumbersome for accurate QD assembly$^{4-8}$. In order to achieve precise QD assembly in simpler manufacturing arrangements, material self-organization schemes in combination with simple nucleation sites with predefinition is necessary.

Nanoindentation is commonly used as a technique for the study and characterization of the mechanical properties of materials at the nanoscale$^9$. However, as a patterning method for nanostructures, nanoindentation has been proposed and used briefly in recent studies$^{10,11}$. Being a physical process for patterning, nanoindentation has, both, the limitation of its deforming nature and the benefits of being easy and cheap to use. In the present study, we describe a technique that uses nanoindentation to pre-define the nucleation sites for Ge QD’s to reside in. Ordered self-assembly of $\sim$10 nm height Ge QDs arrays on Si is demonstrated.

**Results**

Nucleation site matrices were defined using nanoindentation in order for the Ge QD’s to reside in. Indentation matrices of different sizes and spacing were created on boron doped $1 \times 10^{19}$ cm$^{-3}$ Si wafer. Subsequently, a heating treatment was applied to start the Ge self-assembly process. A schematic of the fabrication process described in this work is shown in Fig. 1a-c. Two matrices of indentation are used: $10 \times 10 \mu m$ and $5 \times 5 \mu m$ matrix (Fig. 2 a and b). The distance between sites in the $10 \times 10 \mu m$ matrix is $\sim$500 nm while in the $5 \times 5 \mu m$ matrix is $\sim$5 $\mu m$. The indentations were made using a corner cube indenter tip. The indents on the surface showed trapezoidal pyramidal shapes after the unloading process. The resulting trapezoidal shapes can be referred to the surface nonhomogeneous plastic recovery after the unloading process$^{12,13}$. A linear load function based on a depth-of-indent feedback loop where the indents reached $\sim$150 nm inside the surface achieves a $\sim$10 nm deep permanent indents after the elastic recovery and plastic deformation.

After the nanoindentation, 1 nm layer of Ge (99.99% pure) was sputtered on the Si wafer with the indentation matrices. The Ge sputtering process was performed under $3 \times 10^{-6}$ Tor vacuum and 25°C temperature with a 0.5 A/s deposition rate. Atomic Force Microscopy (AFM) topography imaging following the Ge deposition confirmed that the Si surface including the indentation grooves was covered homogenously with Ge. After the Ge deposition, the sample was heated to a temperature of 700°C for one hour and then cooled down in a N$_2$ ambient. After the cool down, the surface structure was investigated using AFM imaging. The patterned physical deformations due to the nanoindentations in the Si surface act similar to nucleation sites for the Ge islands. The choice of the higher temperature is based on literature studies of Ge spherical QD formation$^{14,15}$. 


Height data of the Si/Ge surface before and after annealing over a part of the 5 × 5 μm indentation matrix is shown in Fig. 3a–c. The indentation sites along with the other parts of the surface were scanned after the deposition and before the annealing process in (Fig. 3a). The surface topography is identical before and after the sputtering process with no annealing. This indicates that there is no nucleation processes before the heating step. The plot in Fig. 3b shows the formation of higher surface features inside the indentation valleys. The QDs showed to have formed in all of the indentation sites and no place around the indents in the vicinity of the indentation matrix. The QDs formed inside the indentations area and only inside the indentations, varied in size and shape. While the biggest QD was in the center of the indentation with a height of ~10 nm and lateral size of ~300 nm, the other QDs shown in the figure have a range of heights and lateral sizes smaller than the center QD. The resulting structure of indentations with the precise formation of Ge QD’s only inside the indentation grooves is shown in a 3D representation restructured from the height imaging using AFM (Fig. 3c).

Cross sections of the height topography using AFM across the indentations before and after the deposition and annealing processes are plotted in Fig. 4a–b. The image shows a spherical QD structure with the height of ~10 nm and a lateral size of ~300 nm with the cross-section going through the largest QD formed in the center of the indentation groove. While the formed QDs vary in lateral dimensions, their height is roughly in the range of ~10 nm. The analysis shows, with high repetitiveness, results for all the indentation sites and QDs formed.

**Discussion**

The main parameter that governs Ge surface diffusion is the chemical potential, μ, of atoms on the surface. This potential will determine the direction of direction of the Ge diffusion on the surface during the annealing. The Ge atoms diffuse from regions of high chemical potential to regions of lower chemical potential. The chemical potential of a curved surface can be described by a simple continuum model\(^ {16,17}\). This assumes a linear dependence of the surface chemical potential on surface curvature\(,\kappa(x,y)\). Equation (1) describes the change in chemical potential on a surface.

\[
\mu = \mu_0 + \Omega \kappa(x,y) \tag{1}
\]

The first term, \(\mu_0\), is the chemical potential of a flat surface. In the second term, \(\Omega\) is the atomic volume, \(\gamma\) is the surface free energy per unit area and \(\kappa(x,y)\) is the surface curvature. In our case, the surface curvature governs the change in chemical potential on the surface since the layer is fully relaxed due to sputtering. Convex regions will have a positive curvature while concave regions will have a negative curvature relative to the flat surface. Thus, the convex regions have a higher chemical potential than concave regions. As a result, Ge diffuses from convex regions to concave regions or from higher chemical potential to lower chemical potential. In this case, the nanoindentation creates local chemical potential energy minima at the bottom of the valley. Nanoindentation sites are a result of a plastic deformation process that resides after the elastic recovery during the unloading. The plastic deformation creates a collection of dislocations inside the indentation site. Such dislocations and surface deformation prove to be preferential sites for early stage Ge growth\(^ {18}\). As a result, the Ge will diffuse to the bottom of the valley and start to fill the nanoindentation resulting in a Ge QD at that location.

In summary, a simple physical technique for achieving Ge quantum dots on Si with high accuracy is demonstrated. The method requires no chemical processing or lithography. Instead, nucleation sites are created using nanoindentation patterning procedure followed by Ge sputtering and annealing. Using this method an ordered self assembled array of ~10 nm height ordered Ge QDs at pre-defined locations is achieved. Moreover, the achieved results highlight an attractive way to manufacture quantum dot structures for...
future quantum electronics and photonic applications due to its strict precision and simplicity.

**Methods**

**Indentation Patterning.** The indentor used is an Asylum Research MFP-nanoindentor in an MFP-3D AFM setup. The indentor is piezo-electrically driven Z flexure with NPS sensor. The Si wafer was patterned using a cube-corner diamond nanoindentor tip.

Indentations of ~10 nm depth were achieved using a depth-dependent load function, where the indentor tip reaches 150 nm into the Si surface with the speed of 50 nm/sec and unloads with the same speed. The resulting indentations are a function of the surface plastic deformation, which was consistently of ~10 nm in depth. The load function was defined to adjust its load force and only sense the indentation depth till it reaches the maximum defined depth of 150 nm, and then retract with the defined speed. The force range was ~150 μN. The load function used for the indentation process is shown in Fig. 5.

**AFM Imaging.** For imaging, AC-in-Air imaging mode was used. Si based AFM cantilevers with Al reflective coating. The tips radii are ~20 nm according to the manufacturer specifications. The imaging was done over the samples at each stage of the process.

![Figure 3](image3.png)  
**Figure 3** | Formation of QD structures inside indentation sites. (a) indentation sites after growth of Ge 1 nm layer prior to annealing. (b) indentation sites after annealing process with the Ge QDs apparent from the height image. (c) 3D AFM data plot of the matrix QD structure.

![Figure 4](image4.png)  
**Figure 4** | Height profiles of Ge/Si structures inside the indentation sites prior to and after the Ge QD assembly. (a) A cross section of an indentation site after Ge growth prior to annealing. (b) A cross section of an indentation site after growth and annealing of 700° C of Ge structures.
Sputtering. The Ge layer was deposited on the Si surface using Physical Vapor Deposition (PVD). The sputtering was done in argon plasma in vacuum of ~10^{-7} Torr pressure. The Ge target used is of 99.99% purity. The deposition rate of the Ge was 0.2 Å/sec.

Thermal Annealing. The sample heating was carried out in nitrogen ambient. Before the sample mounting, the oven was pre-heated to the required temperature of 700°C. Heat was applied over the sample for one hour before the cooling down process in the nitrogen ambient.

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Author contributions
A.A. has performed the experimental work and wrote the manuscript. Both authors interpreted the results. All authors reviewed the manuscript. A.N. is the mentor of the work performed.

Additional information
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