

# Positioning and numbering Ge quantum dots for effective quantum electrodynamic devices

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## 1. Introduction

Recently semiconductor quantum dots (QDs) have been extensively studied for their promising applications on novel nano-optoelectronic devices based on the quantum tunneling process. For practical applications it is indispensable to have a good control on the current-voltage characteristics (CVCs) of these tunneling devices, which depend not only on the material of QDs but also on their shape, size, and location. Even though advanced lithography and patterning techniques are adopted to manipulate the number and position of QDs in a specific region, there still exists a bottleneck on the device fabrication as a result of electron-beam lithography (EBL) resolution limit. Consequently, a reliable fabrication process for generating a precise number of QDs at specified positions in a self-organized manner is urgently developed for quantum electrodynamics.

In this paper, we exploit a selective oxidation method to enable positioning a single Ge QD, double QDs within an oxidized SiGe nano-trench, and even a specific number of QDs within an oxidized SiGe nano-cavity, which are separated from poly-Si electrodes by SiO<sub>2</sub> or Si<sub>3</sub>N<sub>4</sub> tunnel barriers in a self-organized manner. The starting pattern of SiGe to be oxidized is a concave SiGe trench or cavity confined by Si<sub>3</sub>N<sub>4</sub> spacers, rather than an open and convex SiGe nanostructure. Strong nonuniform oxidation rates are induced in the oxidation process of a two-dimensional nanostructure, arising from the dependence of oxidant diffusion on the boundaries and the effects of viscous stress associated with the nonuniform deformation of the oxide. Consequently, pronounced oxidation retardation is observed at the corners of nanostructures. Taking advantages of such unique properties of pattern-dependent oxidation rates and Ge atom segregation/agglomeration during oxidation of SiGe, we are able to modulate Ge QD's position and number by oxidizing SiGe nano-trenches, cavities, or polygons.

## 2. Results and discussions

The fabrication of Ge QDs in nano-trenches and cavities began with Si<sub>3</sub>N<sub>4</sub> and polycrystalline Si layers deposition on top of a Si substrate. After stripe trenches and cavities in various widths were defined using EBL and plasma etching, the nanostructures width was further shrank to  $W = 30 \sim 70$  nm after depositing Si<sub>3</sub>N<sub>4</sub> spacers. Next, poly-Si<sub>0.87</sub>Ge<sub>0.13</sub> was deposited to refill the nanostructures and then directly etched back to remain a 35-40 nm-thick poly-Si<sub>0.87</sub>Ge<sub>0.13</sub> at the nanostructures bottom. The SiGe trenches were oxidized in H<sub>2</sub>/O<sub>2</sub> ambient at 900 °C.

Following complete thermal oxidation, the poly-SiGe layer at

the trench bottom turns into discrete Ge QDs embedded in a dielectric matrix. Figure 1 show the plane-view transmission electron microscopy (TEM) images of oxidized SiGe trenches with Si<sub>3</sub>N<sub>4</sub> spacers, respectively. It is noted that the position of Ge QDs strongly depends on the trench width. For trenches having an initial width of less than 30 nm, spherical Ge QDs ( $9.7 \pm 1.5$  nm) reside randomly either in the center or near the edges of oxidized trenches. On the other hand, for trenches with an initial width of 50 or 70 nm before oxidation, we observed remarkable twin Ge QDs precipitation closely along each boundary between the trench and the nearby Si<sub>3</sub>N<sub>4</sub> spacers, respectively. This reveals the facts that (1) the width of tunnel barriers connecting QDs and poly-Si electrodes is directly determined by the deposited spacer thickness and (2) the distance between QDs is the difference of the trench width and twice of the QD diameter ( $W_{\text{trench}} - 2d_{\text{QD}}$ ). Such double QDs, separated by a thin SiO<sub>2</sub> or Si<sub>3</sub>N<sub>4</sub> tunnel barrier and coupling to each other quantum mechanically, provide an appropriate platform for investigating charge population between them and realizing a charge quantum bit. The number of injected charges into QDs could be controlled by pulsing adjacent poly-Si electrodes; and the electrical control over the charge wave function overlap and the exchange coupling between individual QDs can be manipulated by the top gate bias.

We explain how thermal oxidation converts SiGe trenches to Ge QDs either in the center or along the boundaries of the oxidized region. The scenario is that some segregated Ge atoms precipitate at the interface between the growing oxide and remaining SiGe to form Ge nanocrystals, and some of them diffuse inward and incorporate into the remaining SiGe during the thermal oxidation process, since Si and Ge are totally miscible. This segregation/diffusion process continues till all the SiGe is completely oxidized. This leads to the fact that the location and the number of Ge QDs are predominated by final points where the oxidation process ceases. The characteristic features of a nano-trench oxidation contour are mainly determined by oxidation-induced strain and accessible oxygen diffusion paths. If a trench

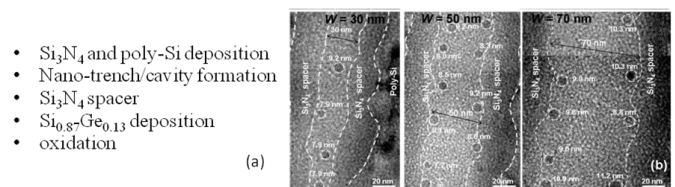


Fig. 1. (a) Process flow and (b) TEM images of Ge QDs formed by thermally oxidizing poly-Si<sub>0.87</sub>Ge<sub>0.13</sub>/poly-Si trenches of various widths with Si<sub>3</sub>N<sub>4</sub> spacers nearby.

width is larger than 30 nm. This results from a reduced oxidation rate at the trench edges in contrast to that at the trench surface owing to stress effects. While for a trench narrower than 30 nm, the endpoint of the oxidation process would be squeezed inwardly at the trench bottom. The refilled SiGe at the trench bottom is concealed by adjacent Si<sub>3</sub>N<sub>4</sub> spacers and bottom Si<sub>3</sub>N<sub>4</sub> layer, which are effective oxidation masks. This indicates that the oxidation process mainly conducts from the trench top surface because of high oxidants concentration for a wide exposure to the oxidant ambient, rather than from the trench sidewalls for limited oxidant exposure.

The aforementioned postulate is further evidenced by oxidizing a variety of poly-Si<sub>0.87</sub>Ge<sub>0.13</sub> polygonal cavities surrounded by Si<sub>3</sub>N<sub>4</sub> spacers, respectively. The Ge QDs precipitate along the peripheral spacer boundaries of oxidized poly-SiGe polygonal cavities with Si<sub>3</sub>N<sub>4</sub> spacers, as shown in Fig. 2. Remarkably Ge QDs exist merely at the corners of triangular, square, and pentagonal cavities as the length of a side is shrank to 120 nm, 70 nm and 55 nm, respectively, while Ge atoms are squeezed into the incenter of triangular and pentagonal cavities as the length of a side is about 50 nm and 30 nm, respectively. Referring to the results in Fig. 2, we note that Ge QDs reside only at the edges of an oxidized SiGe trench if the trench width satisfies the condition of  $30 \text{ nm} \leq W_{\text{trench}} \leq 70 \text{ nm}$ , while QDs present in the trench center if  $W_{\text{trench}} \leq 30 \text{ nm}$ . This suggests that Ge QDs would appear at each corner of a polygon if the radius ( $r$ ) of the corresponding incircle ranges from 15 nm to 35 nm, but a single QD is expected to exist in the center of a polygon if  $r$  is less than 15 nm. It is known that a polygonal area ( $A$ ) equals to the product of its incircle radius and half its perimeter ( $s$ ), i.e.,  $A = \frac{1}{2} r \times s$ . As a consequence, the calculated corresponding length of a side for a triangle, square, and pentagon is 120 (50) nm, 70 (30) nm, and 60 (26) nm, respectively, for  $r = 35$  (15) nm. These estimated geometrical values are in good agreement with our TEM observations in Fig. 2.

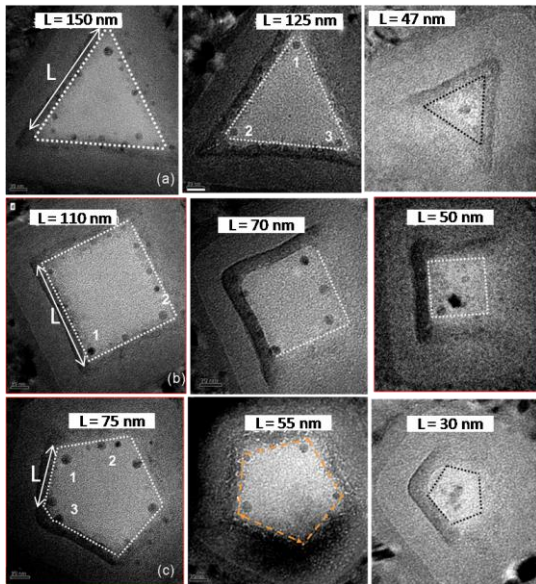


Fig. 2 TEM images of oxidized (a) triangular, (b) square, and (c) pentagonal cavities, refilled with poly-Si<sub>0.87</sub>Ge<sub>0.13</sub>/poly-Si, with Si<sub>3</sub>N<sub>4</sub> spacers nearby. For better TEM observation, the spacer is composed of SiO<sub>2</sub>/Si<sub>3</sub>N<sub>4</sub> (30/20 nm).

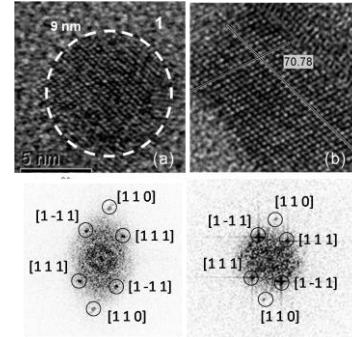


Fig. 3 (a) and (b) HRTEM images and the diffraction pattern .

The experimental results indicate that we can further not only control the exact number of QDs by oxidizing corresponding poly-SiGe polygons with Si<sub>3</sub>N<sub>4</sub> spacers, but also simultaneously position them at either each corner or incenter wherein in a self-organized manner. It has been reported that high efficient SPSs could be produced by coupling QDs into microcavities based on the Purcell effect. However, the precise positioning specific number of QDs in a cavity as well as the coupling the QD's emission wavelength to the cavity resonant mode have been difficult. Our experimental results offer a deterministic approach for placing a single QD or a specific number of QDs within a cavity, which is a prerequisite to fabricating effective QD SETs, SPSs, and low-threshold lasers.

Notably clear lattice fringes in Ge QDs from high-resolution TEM (HRTEM) images in Fig. 3 indicate that these QDs are spherical and in a single-crystalline structure, even after a 900 °C high-temperature oxidation process. No defects such as twin boundaries were observed in the Ge QDs. These Ge QDs were further confirmed to be in a diamond structure by two sets of Ge {111} lattice fringes with an included angle of 70.78° and via fast Fourier-transform (FFT) derived diffraction patterns. The diffraction pattern is similar to that of the main diffraction spots of bulk Ge in a diamond structure. The diffraction spots were indexed with the corresponding diffraction planes. In addition, the Ge QDs are composed of almost pure Ge atoms since the spacing of lattice fringes is rather close to 3.25 Å, the {111} lattice spacing of pure bulk Ge.

### 3. Conclusions

In summary, precise numbering and positioning Ge QDs in an oxidized SiGe polygonal nanocavity is enabled by means of modulating the cavity's geometry and surrounding spacer materials in a self-organized manner. The features of well-resolved oscillatory current spectra due to Coulomb blockade at high temperature in a SHT represent the feasibility of forming a single Ge QD weakly coupled to nanoelectrodes based on our proposed process, which can be easily adapted into the fabrication of embedded circuits. This work demonstrates a deterministic approach for spatial and vertical control Ge QDs in a self-organized way for effective single-electron devices, high efficiency SPSs, and even possible QD molecules or quantum bits.

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