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Flower-Like Distributed Self-Organized Ge Dots on Patterned Si (001) Substrates

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Self-organized Ge dots were obtained utilizing ultra high vacuum chemical molecular epitaxial growth of Ge on electron beam lithographically patterned Si (001) substrates. The dimensions of these etched Si mesa are 65/23/200 nm in diameter/ height/period. The sizes and arrangement of the Ge dots were characterized by scanning electron microscopy and atomic force microscopy. The Ge dots have an average base width of 10 nm and the size is quite uniform. Due to the energetically favorable sites, the Ge dots tend to form homocentrically along the Si mesa edge, and their distribution is flower-like. [DOI: 10.1143/JJAP.42.L718]

KEYWORDS: self-organized Ge dots, ultra high vacuum chemical molecular epitaxy, electron beam lithography, scanning electron microscopy, atomic force microscopy, mesa

Advances in fabrication tools and material growth techniques, self-assembled nanostructures, such as Ge and InAs dots or islands on semiconductor surfaces,¹⁻⁴⁾ have been extensively studied by their potential in the electronic and optoelectronic device applications.5-7) The self-organized quantum dots (QDs) grown by Stranski-Krastanov (SK) mode are considerable promising candidates for use in quantum devices due to their ease in fabrication and their defect-free, high-quality properties. Many efforts on the selft-organized ODs have been carried out to investigate the nature and the mechanism of the dots.^{8,9)} However, QDs grown by SK growth mode are usually distributed randomly on the growth surface and suffer from fluctuations in size and strain in a random manner. Also, the size fluctuation may result in large inhomogeneous broadening in the energy spectrum. This seriously limits potential-device applications of QDs. Therefore, controlling the size uniformity and spatial arrangement of the self-organized dots has been attracting many interests. To improve the Ge dot density and size uniformity, carbon-induced Ge dots¹⁰⁾ and boronreconstructed surface¹¹⁾ for the formation of Ge quantum dots have been proposed. To control the spatial distribution of the dots, a variety of growth methods were experimented. Xie et al. showed the vertical alignment of InAs Quantum box islands by stacking growth of the multi-layers of dots.¹²⁾ Lee et al. grew the InAs QDs on predesigned mesas with added strain layer, leading to "one dot on one mesa" relationship.¹³⁾ Jin et al. demonstrated one-dimensional selfalignment of Ge islands on the (110)-oriented ridges of the Si strip mesas and positioning of Ge islands at the corners of square Si mesas with selective eptiaxial growth (SEG) method.¹⁴⁾ Kitajima et al. succeeded in fabricating twodimensional alignment of Ge islands on lithographically patterned Si (001) surfaces with the smallest Si mesa of 140 nm, resulting in a "one island on one mesa" arrangement.¹⁵⁾ However, the dots size stated above is still too bigger to be considered as QDs. For effective quantum confinement, it is necessary to form smaller dots of about a few tens of nanometers. From previous theoretical simulations of heteroepitaxy on patterned substrates,^{16,17)} when the lateral dimensions of the patterns are smaller than 100 nm, the strain energy in the epilayer can be significantly reduced, resulting in the vertical growth of Ge dots on top of the Si

mesas. So far, there is no report on using Si mesa less than 100 nm for the formation of Ge dots to realize the spatial distribution of the Ge dots.

In this letter, we report self-organized Ge dots obtained utilizing ultra high vacuum chemical molecular epitaxial (UHVCME) growth of Ge on electron beam (EB) lithographically patterned Si (001) substrates. The sizes of these etched Si mesas were less than 100 nm. As characterized by scanning electron microscopy (SEM) and atomic force microscopy (AFM), the Ge dots have an average base width of 10 nm and the size is quite uniform. Also, the Ge dots tend to form homocentrically along the Si mesa edge and the distribution is flower-like. Experimental details of the Ge dots growth are described below.

Figure 1 summarizes the process flow for the fabrication of the Ge dots on the patterned Si mesas. First, the six inch



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Fig. 1. Process flow for the fabrication of self-organized Ge dots on patterned Si (001) substrates.

p-type Si (001) wafer was degreased with acetone (ACE) and isopropyl alcohol (IPA), and rinsed in the de-ionized water. After blowing dry with nitrogen and baking in air, the Si substrate was coated with a 120 nm thick polymethylmethacrylate (PMMA) as EB resist and then baked at the temperature of 180°C. After exposing at the Leica EBML300 EB direct writing system at 40 KeV and developing in methyl isobutyl ketone (MIBK):IPA (1:3) for 100 s and rinsing in an IPA for 30 s, well-defined 100 nm PMMA trenches with 200 nm period were formed. The Si substrates with PMMA trenches were then etched using the PMMA trenches as the etching mask. The Si etch was performed in TEL 5000 Oxide Etcher with gas mixtures of argon, methane and oxygen. After plasma etching, the wafer was dipped in ACE to remove the residual PMMA. Finally, the Ge dots were grown on the etched Si mesas by UHVCME system under the growth temperature of 550°C with GeH₄ flow rate of 5 sccm. The growth time was 15 s and the corresponding nominal thickness of Ge layer was approximately 30 Å.

Figures 2(a)-2(f) show several typical arrangements of self-organized Ge dots on patterned Si mesas. By controlling the EB exposing dose and etching time, the formed Si mesas have dimensions of 65/23/200 nm in diameter/height/period as can be seen in Figs. 3(a) and 3(b). Compared to the schematic drawing of Fig. 1, the real profile of etched Si mesa looks like the mountain, which is due to the lateral plasma etching. It has been predicted that for growth of Ge dots on small Si mesas with sizes of less than 100 nm, there



Fig. 2. SEM pictures of typical arrangements of flower-like distributed self-organized Ge QDs on patterned Si mesas with the mesa dimensions of of 65/23/200 nm in diameter/height/period.





Fig. 3. AFM images of (a) three-dimensional 3×3 Si mesas array and (b) cross-sectional analysis of one Si mesa, resulting in the size of 65 nm.

will be some novel phenomenon occurred on the top of the mesas.^{16,17)} In this work, we found that when the size of the mesa was 65 nm, the lateral growth of Ge dots seemed to be effectively suppressed and the size of the Ge dot became very small, leading to an averge base width of 10 nm. Furthermore, the uniformity of these dots has also been improved and many Ge dots can be grown on the top of the small Si mesas to form the flower-like distribution. From SEM image in Fig. 2(a), there is no "one dot on one mesa" relationship that was observed in ref 15. Instead, about ten Ge dots were homocentrically grown along the Si mesa edges. The mesa edges are more favorable for dots nuceation than other sites. This result is consistent with the previous works of Jin et al.¹⁴⁾ and Kitajima et al.¹⁵⁾ In addition, the Ge dots initially formed at the mesa edge may change the strain distribution.¹⁴⁾ Furthermore, when the mesa sizes were less than 100 nm, the edge effect may take an important role on the strain distribution of the mesa. By the above two mechanism, we also found other arrangements of Ge dots on Si mesa as can be seen in Figs. 2(b) to 2(f). These dots tend



Fig. 4. Three-dimensional AFM image of flower-like distributed selforganized Ge dots on patterned Si mesas.

to be symmetrically arranged on the top of these Si mesas. From SEM image of Fig. 2(f), the dot density of Ge on Si surface was estimated to be 3×10^{10} cm⁻².

In order to measure the uniformity of Ge dots on Si surface, the phase-mode of AFM were used, which can differentiate areas with different properties such as viscoelasticity or mechanical properties on a sample. Figure 4 is the three-dimensional image of the flower-like distributed self-organized Ge dots on patterned Si mesas. From this figure, uniform Ge dots were indeed formed on the top of the patterned Si mesas.

In summary, the self-organized Ge dots were grown by UHVCME on the electron beam lithographically patterned Si (001) substrates. The diameters of these mesas were less than 100 nm. These small Si mesas may suppress the lateral growth of Ge dots, resulting in an average Ge dot base width of only 10 nm. Due to the energetically preferred sites and edge effect, the Ge dots were formed homocentrically along the Si mesa edges. Their distribution is flower-like. Several typical arrangements of Ge dots were observed in our experiments. With optimal growth conditions, the uniform Ge dots with dot density of 3×10^{10} cm⁻² were formed on the top of the Si mesas.

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