Regimented placement of self-assembled Ge dots on selectively grown Si mesas

G. Jin,[a] J. L. Liu, and K. L. Wang
Device Research Laboratory, Electrical Engineering Department, University of California at Los Angeles, Los Angeles, California 90095-1594

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The control of positioning of self-assembled dots is critical for the fabrication of regimented arrays in signal processing applications. We report the controllable positioning of self-assembled Ge dots using selectively grown Si mesas as a template. The dependence of the dot arrangement on growth temperature and Ge thickness has been investigated. The experimental results show ability to control the positioning of Ge dots based on energetically preferential nucleation. The Ge dot growth on Si mesas is demonstrated to be a promising way to realize the placement of regimented arrays of self-assembled dots and even a single dot. This technique can be extended to other heterostructure growths. © *2000 American Institute of Physics.* [S0003-6951(00)04124-3]

Self-assembled Ge dots have attracted extensive interests due to their appealing properties and potential applications.[1] It is well known that self-assembly is spontaneous and the spatial distribution is random. Recent research interests in dot-based applications, such as cellular automata[2] and other signal processing applications,[3] require the controlled placement of self-assembled dots. Thus, how to control the spatial distribution of self-assembled quantum dots becomes an essential issue in self-assembly studies.

Many techniques have been introduced in attempt to realize the control of the spatial distribution of self-assembled dots. For example, growth on tilted substrates demonstrated one-dimensional (1D) and two-dimensional (2D) arrangements of the dots with some success (of limited ordering).[4,5] Growth on the samples with intended dislocations[6] and defects[7] as a template could form preferential nucleation sites for ordering. Ge dots were also formed on patterned substrates prepared by electron beam[8] and nanoprinting techniques.[9] Most recently, one of the most effective approaches,[10,11] using selective epitaxial growth (SEG) to form Si mesas as a template for Ge dot growth, has been reported to realize ordered 1D dot arrays. However, it is far from the desired control of the dot positioning for practical applications.

In this letter, we report the placement of regimented 2D arrays of self-assembled Ge dots on patterned Si (001) substrates. It is found that based on the preferential nucleation, the positioning of self-assembled Ge dots can be freely controlled by varying the growth conditions, such as the growth temperature, and Ge and Si thickness. Discussions are given to help understand the dot placement.

Si (001) substrates were first thermally oxidized to form about 400-nm-thick SiO_2. Then, by using conventional photolithography, Si windows were formed with their edges oriented along the ⟨110⟩ directions. The patterned Si (001) substrates were chemically cleaned and dipped in a diluted HF solution to form a hydrogen-terminated surface before being loaded into vacuum chamber. The growths were carried out in a molecular beam epitaxy system with a Si_2H_6 gas source and a Ge Knudsen cell source. The base pressure in the growth chamber was about 5×10^{-10} Torr and the main residual gas was hydrogen. The pressure during the Si SEG was in the range of 10^{-6} Torr. After thermal cleaning, about 120 nm Si was selectively grown in the exposed Si windows at 660 °C. Si mesas with sidewall facets were formed. Details on the facet formation in the SEG process can be found elsewhere.[12,13] After the Si growth, Ge was deposited at growth temperatures from 500 to 700 °C and a growth rate of about 0.01 nm/s. After Ge growth, the samples were removed from the vacuum and the silicon oxide was etched away for atomic force microscopy (AFM) study. All the AFM scanning was parallel to an ⟨110⟩ direction with a contact mode.

Figure 1(a) shows a 2D AFM image of self-assembled Ge dots on the SEG Si mesas. The bright regions are Si mesa networks, which were formed after the selective epitaxial growth.[12,13] The dark regions correspond to the original SiO_2 mask regions. The darkness, corresponding to a low region, is due to the removal of the SiO_2 masks for AFM measurements. The original Si windows are oriented in ⟨110⟩ direc-

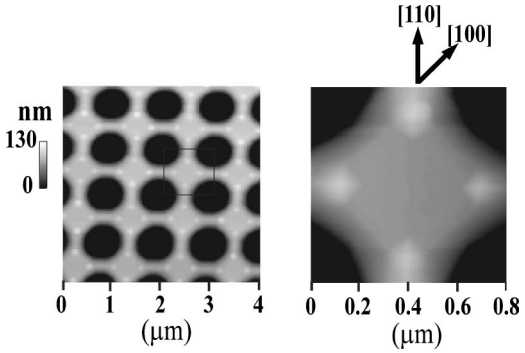

FIG. 1. (a) 2D AFM image of a 2D array of Ge self-assembled dots on pregrown Si mesas. The growth temperature is 600°C and the Ge thickness is equivalent to 0.8 nm. The average dot height is 12 nm. (b) Magnified view of the dot arrangement on a (boxed) unit cell of Si mesas in Fig. 1(a). The dots are square-based pyramids, which are located at the corners and whose base squares are oriented in ⟨100⟩ directions.

[a]Electronic mail: gjin@ee.ucla.edu

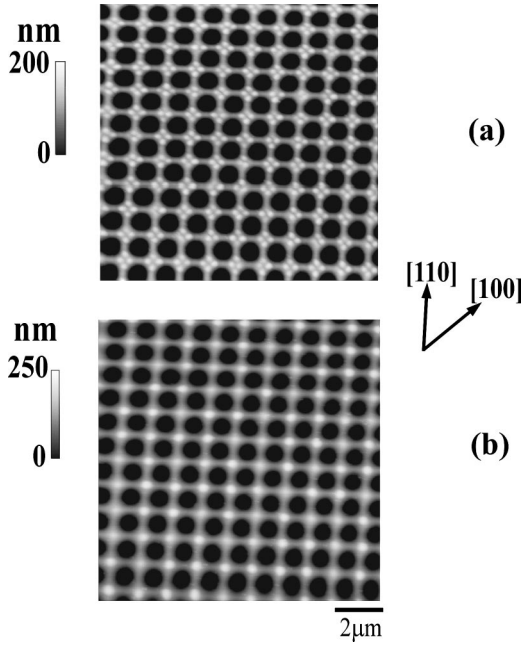

FIG. 2. 2D AFM images of 2D arrangement of Ge dots with 1.6 nm Ge (a) growth temperature is 650°C, four Ge dots are arranged on each unit of Si mesas and (b) growth temperature is 700°C, only one Ge dot is arranged on each unit of Si mesas.

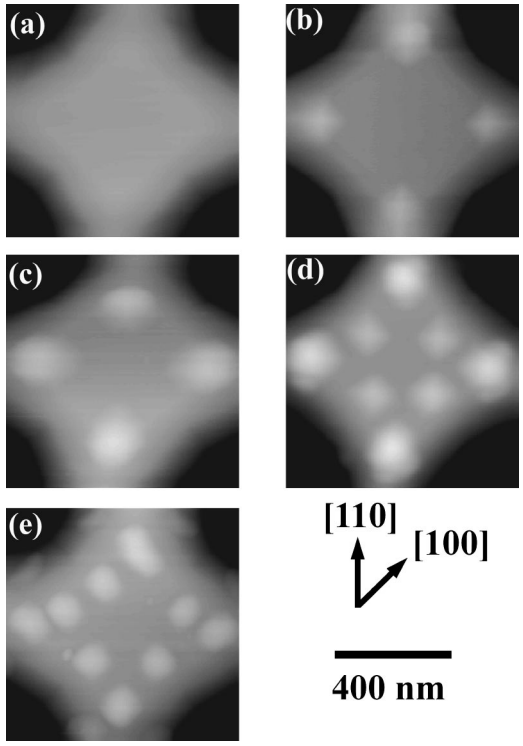

FIG. 3. Dependence of the arrangement of Ge dots on Ge thickness. Several configurations of arrangement can be seen with different Ge thickness. The Ge thickness deposited is equivalent to (a) 0.4, (b) 0.8, (c) 1.1, (d) 1.3, and (e) 1.7 nm, respectively.

tions, however, the base lines of the sidewalls of the Si mesas after SEG process are not oriented in ⟨110⟩ directions, but in ⟨100⟩ directions. The change of the base line orientation may be attributed to the result of the competitive growth on different facets.[13] A 0.8-nm-thick Ge was deposited at a temperature of 600 °C, resulting in pyramid-shaped Ge dots with an average height of 12 nm, as shown in Fig. 1(a). It is interesting to note that there are four Ge dots on each unit cell of the Si mesas and the arrangement of Ge dots shows ordered 2D arrays. Figure 1(b) is a magnified view of the boxed unit cell. There are four Ge dots located at each corner of a Si mesa and the central region is free of Ge dots. The base squares of the dots are oriented in ⟨100⟩ directions. The formation of Ge dots at the corners is due to the energetically preferential nucleation.[10,11]

In order to understand the effect of temperature on 2D arrangement of Ge dots, we investigated the dependence of 2D arrangement on growth temperature. For comparison, the growth temperature was the only variable while the other growth parameters were kept fixed. The Ge amount deposited was equivalent to 1.6 nm. Figures 2(a) and 2(b) present the AFM images of the samples with the growth temperatures of 650 and 700 °C, respectively. As we see, four dots prefer to locate at the corners of each Si mesa at the temperature of 650 °C as shown in Fig. 2(a). As the temperature increases from 650 to 700 °C, we found that only one dot on each Si mesa is formed on each Si mesa at 700 °C [Fig. 2(b)], and the top (001) facet of Si mesas becomes smaller than that at the temperature of 650 °C. Even though the other growth conditions were the same, we observed that the mesa height increased with the increase of the growth temperature. This is due to the mass transfer from the sidewalls of Si mesas.[12] At higher temperature, the mass transfer is more pronounced, leading to the increase of the height and the reduction of the mesa tops (001). Meanwhile, the dot size becomes larger with the increase of the temperature.[14] Therefore, only one Ge dot was formed on each small top (001) facet.

In order to gain insight into the formation of ordered dot arrays, we have also performed the experiments to study the dependence on Ge amount deposited. Figure 3 shows 2D images of one unit mesa cell [similar with Fig. 1(b)]. The growth temperature was kept at 600 °C for all of these samples. When the Ge amount is 0.4 nm (about 3 ML, 1 ML=0.143 nm), no Ge dots are seen [Fig. 3(a)], suggesting layer-by-layer growth at this thickness. With the Ge thickness of 0.8 nm, four Ge dots are formed at the corner sites of each Si mesa [Fig. 3(b)]. The formation of Ge dots means that Ge growth on the Si mesas [with the top (001) dimension of ~0.5 μm] is still in the Stranski–Krastanov growth mode and the critical thickness is about 0.5 nm, in agreement with that of the growth on planar Si(001) substrates. As the thickness increases to 1.1 nm, the result is similar to that with 0.8 nm Ge, i.e., only four dots are located at the corner sites of each Si mesa. The difference is that the Ge dots with 0.8 nm Ge are square-based pyramids [Fig. 3(b)], but all the dots with 1.1 nm Ge are domes, whose size is larger [Fig. 3(c)]. With further increase of deposited Ge, the second set of Ge dots are observed, which are located between the corner dots along the edges [Fig. 3(d)]. The dot placement is suggested to be associated with the strain energy distribution, which will be discussed later. The second set of Ge dots are square-based pyramids, which are different from the dome dots at the corners. These pyramids occur at the early stage of their evolution, i.e., that they have not undergone the shape transformation.

Based on our experimental observations, we discuss the

mechanism associated with the Ge dot arrangement using an energetic analysis. At the early stage of Ge deposition, the growth is in layer-by-layer mode on Si SEG mesas. It is well known that strain energy increases with the increase of Ge due to the lattice mismatch between Si and Ge. Thus, Ge dot formation is to reduce the strain energy. Ge atoms tend to nucleate at the sites with a minimum total energy. The corner sites are the energetically preferred sites as they are susceptible to the strain relaxation, thus the nucleation is easier to take place at the corner sites. After the formation of the Ge dots and with the increase of the dot size, the strain energy at the corner sites increases, and then the corner sites are no longer the energy-preferred one. On the other hand, the other sites along the mesa edges may become the favorable even though they are not preferred in the beginning of the growth. Therefore, Ge dot is expected to form in the middle site of the edge, which is consistent with the observation of the two sets of the Ge dots on each Si mesa. This mechanism is also applicable to the growth in other heterostructure growth, such as InAs on GaAs.

In summary, 2D regimented arrays of self-assembled Ge dots on pregrown Si mesas have been investigated. Several configurations of arrangements on the Si mesas have been observed. Our results show the ability to position self-assembled Ge dots and even single dot by controlling growth conditions based on energetically preferential nucleation. This confirms our previous prediction that the placement of Ge dots could be controlled at will on pregrown Si mesas. Finally, the Ge dot growth on Si mesas is demonstrated to be a promising way to realize the regimented arrays of the dots and even the control of a single dot.

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