Materials Today: Proceedings 47 (2021) 1617–1620

Contents lists available at ScienceDirect

Materials Today: Proceedings

journal homepage: www.elsevier.com/locate/matpr

Silicon nanodots via sputtering of Si(111)-7×7 surfaces and post-annealing

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ARTICLE INFO

Article history:
Available online 19 May 2021

Keywords:
Self-organized epitaxial nanostructure
Nanodots
Scanning tunneling microcopy

ABSTRACT

Using *in-situ* scanning tunneling microscopy (STM) we have investigated the evolution of Si(111)-7×7 surfaces, prepared under ultrahigh vacuum condition, upon Ar^+ ion sputtering and subsequent annealing. We have monitored how the surface atomic arrangement changes upon annealing of the sputtered Si (111)-7×7 surface. Sputtering renders the Si(111)-7×7 surface amorphous and rough. Annealing at 500℃ causes no recrystallization, although the surface roughness is reduced. When the sample is annealed at 600℃, recrystallization starts producing short-range orders. Flat-top nanoislands with a height distribution appear. The top surface of these nanoislands is ordered; most islands have Si(111)-7×7 surface reconstruction, while there are also islands with other surface reconstructions, such as 5×5, 2×2 etc. Smaller silicon nanodots grow at the edges of these flat-top islands.

Selection and peer-review under responsibility of the scientific committee of the National Conference on Recent Advances in Functional Materials-2020.

1. Introduction

Surface patterning in nanoengineering with the help of ion beam irradiation is an active field of research for understanding the surface dynamics at the atomic scale and also for the nanodevice fabrication in the industries [1–3]. Ion beam sputtering is a powerful tool in the generation of diverse nanoscale surface topographies by spontaneous self-assembled mechanisms [2,4–7]. These nano-scale patterns have immense applications in the area of optical devices, suitably reconstructed substrates for scanning tunneling microscopy (STM) studies, photonic devices, good oxidation surface, and strain-free structured templates [1,6–8]. Fabrication of nano-dots by keV ion irradiation holds promising applications because of the possibility of the generation of large-area well-arranged arrays of dots in a single technological step [2,9,10]. When a solid is exposed to energetic ion beam irradiation, the surface remains in a non-equilibrium state and as a consequence, different atomistic surface processes become active [11]. The nontrivial competition among these mechanisms causes roughening or smoothening of the solid surface and generates diverse self-organized topographies on it depending on irradiation parameters such as ion beam incidence, fluence and energy [10,12]. Ghose et al. reported on the surface and subsurface defect formation due to low energy (200 eV) Ar^+ ion beam irradiation on the Si(111)-7×7 surface [13]. Zandvliet et al. investigated the effect of Ar^+ ion beam (3 keV) impact on the Si(111) surfaces, with a fluence $\leq 10^{12}$ ions cm^{-2} [14]. One can investigate the surface atomistic processes of the ion beam irradiated surface, by applying temperature, pressure etc. To investigate the atomistic processes on the ion-beam-irradiated surface at atomic scale, in-situ experimental arrangement is inevitable, which is not frequently accessible. An alternative approach to address this issue might be utilization of the ion beam sputtering in a molecular beam epitaxy (MBE) chamber and in-situ investigation of the atomistic processes

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https://doi.org/10.1016/j.matpr.2021.04.319
2214-7853/
Selection and peer-review under responsibility of the scientific committee of the National Conference on Recent Advances in Functional Materials-2020.

with the application of temperature, pressure etc. Ar^+ ion has been used regularly for the cleaning of semiconductor, metal surfaces for growth of epitaxial nanostructures. In the present work, the effect of 500 eV Ar^+ ion beam sputtering on a clean Si(111)-7×7 surface and subsequent atomic scale processes have been investigated using the Ar^+ ion gun in the MBE chamber and in-situ STM investigation via post-sputtering annealing of the sample. The sputtering was carried out at room temperature.

2. Experimental details

The experiments were performed in an ultra-high vacuum (UHV) molecular beam epitaxy (MBE) growth chamber, which is connected with a UHV variable temperature scanning tunneling microscopy (VT-STM) system. The UHV chambers of MBE and STM are interconnected and the samples are transferred from one to other in situ. The base pressure in the MBE chamber was 5.2×10^{-11} mbar and in the STM chamber was 2.3×10^{-10} mbar, during the experiment. P-doped, n-type Si (111) wafer with resistivity of 10–20 Ω-cm was used. Atomically clean Si(111)-7×7 surfaces were prepared by degassing the Si(111) substrate at ~700 °C for about 14 h, and then flashing the substrate at ~1250 °C for one minute. The sample was then quickly brought down to ~780 °C. Kept at this 780 °C for 30 min, the sample was then slowly allowed to cool down to room temperature (RT). After checking the cleanliness and surface reconstruction in the STM chamber, the sample was transferred back to MBE chamber where it was subjected to a 500 eV Ar^+ ion sputtering. During the Ar^+ ion sputtering of the Si (111)-7×7surfaces, the pressure in the MBE chamber rose to 1.0×10^{-5} mbar due to insertion of Ar gas. Following the Ar^+ ion sputtering of the Si(111)-7×7surface, the sample was transferred to the STM chamber for atomic resolution imaging measurements. Subsequently the sample underwent thermal annealing at temperatures of ~500 °C and ~600 °C sequentially for 1 h in each case.

3. Results and discussion

3.1. Effect of Ar^+ ion sputtering on clean Si(111)-7×7 surfaces

Fig. 1(a) shows the STM image of a clean Si(111)-7×7 surface. The atomically resolved image of 12 adatoms of the faulted and upfaulted half units were checked (not shown here). Fig. 1(b) shows the STM image of the 0.5 keV Ar^+ ion sputtered Si(111)-7×7 surface. From the STM image, it is clear that the surface has become amorphous from atomically clean crystalline one. STM investigation reveals that there is no atomic periodicity on the atomic scale on the surface (not shown here). Fig. 1(c) shows the STM image of the ion-sputtered Si(111)-7×7 surface following annealing at ~500 °C. The surface roughness has decreased upon annealing of the Ar^+ ion sputtered Si(111) surface. However, no recrystallization of the surface is observed.

3.2. Effect of annealing at 600 °C

When the 500 °C-annealed sputtered sample is further annealed at 600 °C, new features appear on the surface. Fig. 2 (a-c) show STM images of the 600 °C-annealed sample. After annealing at 600 °C flat top nanoscale Si island formation is observed. Atomically resolved STM investigation reveals that the islands are crystalline with well-ordered surfaces. Higher magnification STM images in Fig. 3(d-f) show that the top surface of each island possesses atomically well-ordered reconstructed arrangements. Different plateaus contain different reconstructions, such as 7×7, 5×5, 2×2 [Fig. 3(d-f)]. However, the dominant surface reconstruction is 7×7. At the edges of the flat top plateaus, formation of various features like atomic chains, staircases and Si nanodots are observed (Fig. 3). It is obvious that at this annealing temperature, although the surface is recrystallized, the long-range order with 7×7 reconstruction of the original surface is not recovered. The recrystallized surface is still quite rough compared to the original surface. D. Gupta *et. al* reported the epitaxial recrystallization of the amorphized Si(111) surface [15,16] by annealing the Si(111) samples at 850 °C. The Ar^+ ion fluence in the range 1×10^{17} to 5×10^{17} ions cm^{-2} were used for the amorphization of the Si (111). The authors did not make any investigation at the atomic scale. Si(111) surface has a 7×7⇌'1×1′ phase transition at 870 °C. The high temperature '1×1′ phase is a disordered structure. Near the phase transition temperature, although 7×7 is the predominant structure, regions of the surface with 11×11, 9×9, $\sqrt{3}\times\sqrt{3}$, 2×2, c(2×4) and even $\sqrt{7}\times\sqrt{7}$ R19° reconstructions were observed [17,18]. Also on disordered flat trenches on a Si(111)-7×7 surface, small patches of 11×11, 9×9, c(5×$\sqrt{5}$), c(4×4) and 2×2 structures were observed [19]. When there is no adsorbate present, the surface atomic density and the substrate temperature play a crucial role in the reconstruction of the Si(111) surface. When the oxide-layer-removed clean Si(111) surface is slowly cooled down from above the order–disorder transition temperature, the surface atoms rearrange in 7×7 units from the step edges and proceed on the terrace. But if the surface is quenched much faster to below the transition temperature, the surface atoms rearrang in stable as well as metastable reconstructed surface units. *Yang et al.* [17] reported a number of metastable surface reconstructions namely, 2×2, 9×9, 11×11, c(2×4), $\sqrt{3}\times\sqrt{3}$ etc. on the Si(111) surface, which possess higher surface atomic densities compared to the energetically stable 7×7 surface reconstruction. (d-f) High resolution STM images reveal that the top surfaces of the silicon nanoislands reconstruct predominantly in the (7×7) structure, but there are also (5×5) and (2×2) reconstructions. The inset in Fig. 3(c) shows the height profile along the marked line with an arrow.

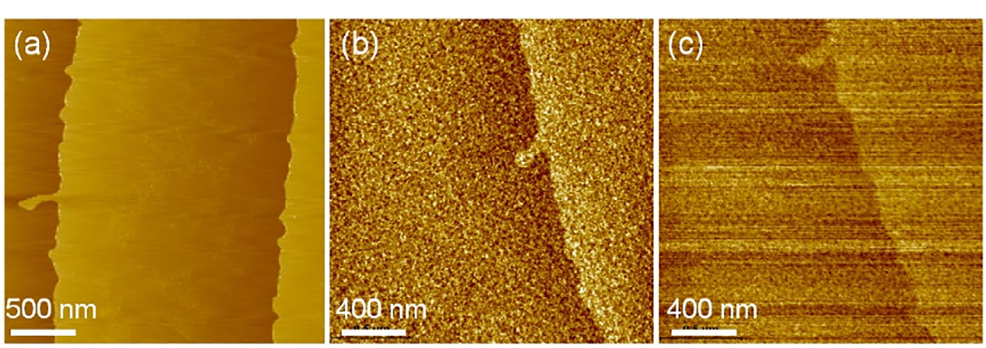

Fig. 1. STM images: (a) of an atomically clean Si(111)-7×7 surface, (b) of the sputtered surface, (c) of the surface of the sputtered sample, annealed at 500C.

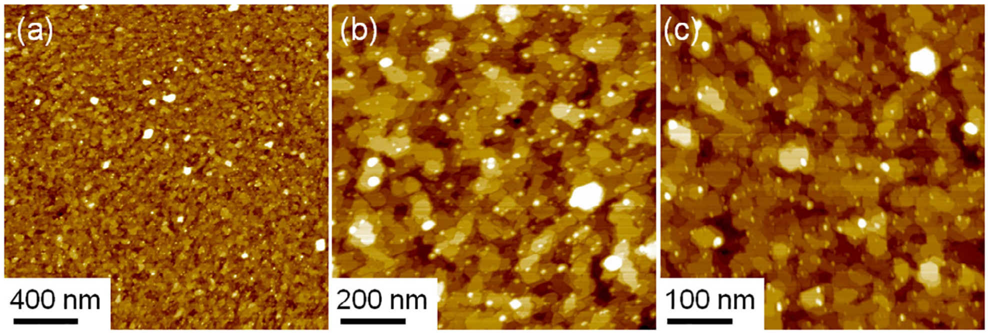

Fig. 2. STM images from (a) 2000×2000 nm^2, (b)1000×1000 nm^2 and (c) 100×100 nm^2 areas of the sputtered Si(111) surface after annealing at 600C.

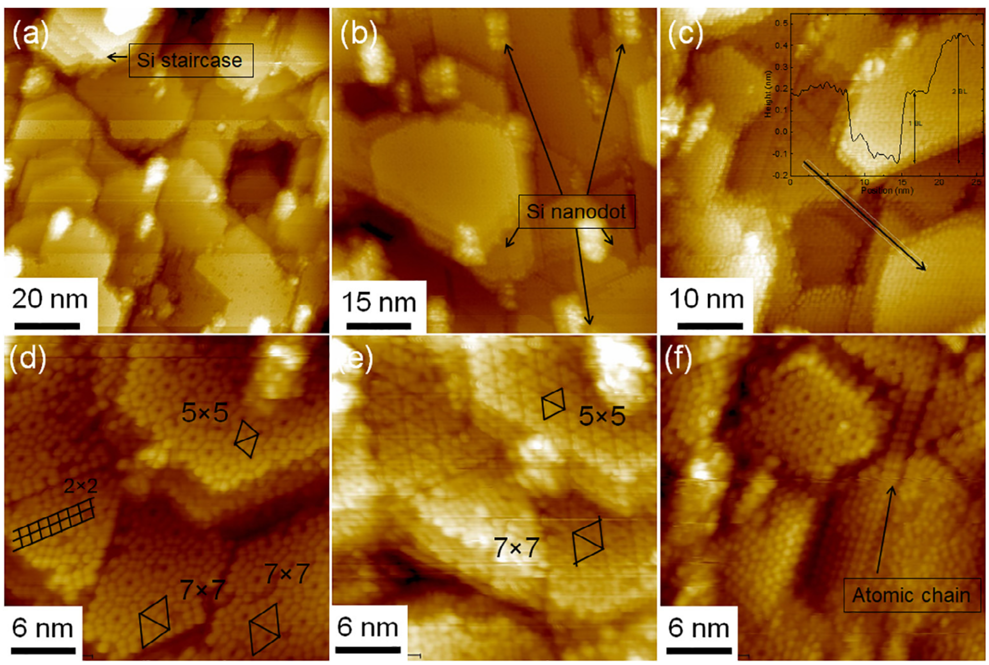

Fig. 3. (a-f) STM images of the Ar$^+$ sputtered Si(111) surface after annealing at 600C. (d-f) High resolution STM images reveal that the top surfaces of the silicon nano-islands reconstruct predominantly in the (7×7) structure, but there are also (5×5) and (2×2) reconstructions. The inset in Fig. 3(c) shows the height profile along the marked line with an arrow.

In recent years, the effect of highly energetic (a few keV) Ar$^+$ ion beam irradiation on the Si(111) surfaces has been investigated by a number of groups using ion sources in ion accelerators [3,5,15,20]. *D. Vidya et. al.* [20] reported the depth-controlled amorphization of Si(111) substrates using 80 keV off-normal Ar$^+$ ion beams. Subsequent annealing of the irradiated Si(111) samples at 800 °C produces depth-varying buried amorphous layer due to defect recrystallization and damage recovery [20]. However, the present work focuses on in-situ investigation of low energy (500 eV) Ar$^+$ ion beam sputtering effect on the clean Si(111)7×7 surface resulting in a flat amorphous Si surface. Subsequent annealing of this amorphous Si surface has produced crystalline Si(111)7×7 plateau island decorated with nanodots. The advantage of this investigation is that this approach sheds light on the atomic scale phenomena on the ion beam irradiated Si(111) surface and how this irradiated surface can be used to produce self-organized Si nanodots.

4. Conclusion

The effect of Ar$^+$ ion sputtering of the atomically clean Si(111)-7×7 surfaces has been investigated. The 0.5 keV Ar$^+$ ion sputtering transforms the crystalline silicon surface into rough amorphous surface. Upon annealing of the sputtered Si(111) sample at 500 °C, the surface remains amorphous although the surface roughness decreases. Annealing at 600 °C transforms the Si(111) surface, into silicon islands with flat-top plateaus which display short range orders. The silicon islands stabilize at different heights and consequently the surface roughness remains much higher compared to the original unsputtered surface. The top surface atomic arrangement of the silicon islands or plateaus is predominantly 7×7, although many islands have other surface reconstructions like 5×5, 2×2 etc. This has similarity with surface reconstructions observed around the transition temperature of the thermally induced 7×7⇌'1×1' order–disorder phase transition on the Si(111) surface. The formation of Si nanodots on the edges of the flat-top silicon islands, narrow atomic chains and staircase structures have been observed.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

JCM and DD were supported by CSIR Fellowships [09/080 (0674)/2009-EMR-I] and [09/080(0725)/2010-EMR-I] respectively. This work was carried out in BND's laboratory when JCM, DD, RB, AR and BND were all at Indian Association for the Cultivation of Science, Kolkata, India.

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