Controlled Formation of Atomic Step Morphology on Micro-patterned Si (100)

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Micro scale features are fabricated on Si (100) surfaces using lithographic techniques and then thermally processed in an ultra high vacuum (UHV) environment. Samples are flash heated at 1200 °C and further annealed at 1050 °C for 18 hours. The surface morphology was examined using an atomic force microscopy (AFM). The process resulted in the formation of symmetric, reproducible step-terrace patterns with very wide atomically flat regions exhibiting highly reproducible step-terrace morphology. 25 µm lithographically patterned cells spontaneously transform into a symmetric formation marked by step bunches pinned by pyramidal structures separated by wide atomic terraces. The pyramidal features are visible using a conventional optical microscope and are to be used as fiducial marks to locate nanoscale features fabricated on the atomically flat terraces.

The practical fabrication of novel silicon-based atomic structures has two essential requirements, the ability to control the step terrace patterns on silicon surfaces at the atomic-scale and the need to locate these devices in tools other than those used to fabricate the structures. Realistic wafer-scale integration of atomically controlled nanostructures with conventional silicon-based microelectronics technology requires atomic scale control of step-terrace morphology at the die or wafer scale where the positions of all atoms on a Si-wafer substrate are controlled by design[1]. Self-organized step-terrace patterns can serve as nanometer scale templates and are already becoming a necessity for atomically precise manufacturing[2]. Fabrication of quantum dot nanostructures is an example of an area of significant interest that requires atomically flat surfaces to be used as substrates[3,4]. To achieve repeatable atomic surface templates, one needs to have a predictive understanding of the evolution of step-terrace patterns on the surface from a fundamental surface science point of view.

The BCF-theory of crystal growth describes crystal surfaces by steps of single monolayer height and the evolution of the surface described by the diffusion of adatoms on terraces[5]. Several other parameters also play into the dynamics and evolution of atomic step patterns such as the desorption of atoms into the surrounding environment, and atomic attachment-detachments at the step edges. All of these are directly related to the sample temperature. The mechanical surface stress due to the sample clamping etc. has also been shown to influence the atomic step patterns[6]. Apart from those processes that are intrinsic to crystal surfaces, it possible to control the dynamics externally by patterning or texturing the surface at the micrometer scale. Published reports show wide atomic scale terraces can be achieved by annealing patterned surfaces with micrometer scale features[7,8].

In the results reported here, we achieve 'control' of the step-terrace patterns within lithographically defined areas by taking advantage of electromigration effects where the ad-atoms drift in the direction of the electric field during a resistive heating process and the consequent step bunching of the surface[9]. The combination of these effects drive the system toward self-organized states, characterized by the appearance of ordered step-terrace patterns on the vicinal surfaces which can be exploited in the development and fabrication of nanotechnology devices. In this letter we report the *controlled* formation of highly reproducible step-terrace morphology with wide terraces across the cells in a patterned Si (100) surface by high temperature processing. The control of the step-morphology is achieved by optimizing the design characteristics of the surface patterns and subsequent processes.

Standard lithographic procedures in combination with dry (plasma) etching were used to create micrometer sized patterns having 25 µm and 50 µm squares in two different orientations on a Si (100) substrate. The resulting patterns are mesas (plateaus) separated by trenches 1 µm deep and ~2 µm wide. Standard RCA cleaning procedures were used before loading the samples into the UHV chamber for high temperature annealing. The samples were mounted on an Omicron [10] sample holder and degassed at 650 °C for more than 12 hours by resistive heating. Thermal processing of the samples was carried out in an ultrahigh vacuum (UHV) at 2×10^{-10} mbar.

We have completed two sets of experiments where in one case the samples were only flashed at 1200 °C for a few seconds and in the other case they were annealed at 1050 °C for more than 18 hours. This temperature is sufficiently high to enable significant diffusion and allow the surface to approach equilibrium. Care was taken to keep the samples below the roughening silicon transition temperature (1150-1200 °C)[11]. The samples were then examined *ex situ* using an intermittent contact mode ambient AFM (Veeco Metrology[10]) in a controlled temperature (21.0°C ± 0.1°C), humidity (45% ± 5%), and clean air (class 1000) environment. The samples were stored in a controlled environment while transporting from the UHV facility to the ambient AFM.

Fig. 1 shows an AFM image after a 1200 °C flash heating. The figure shows a considerable amount of material transport on the surfaces. The 1 µm deep trenches were reduced to 0.6 µm after the 1200 °C flash[12]. The area within the patterns has transformed into the symmetric formation of step terrace patterns. The step-terrace morphology is very reproducible across various cells in the patterned area. The morphology achieved is an outcome of the kinetics that drive the surface to a minimum surface energy configuration. Details of the theory and mass transport (equilibrium crystal shapes, ECS) have been

previously studied and reported in the literature[13]. The size and orientation of the micro patterns etched on the surface dictate the resulting morphology.

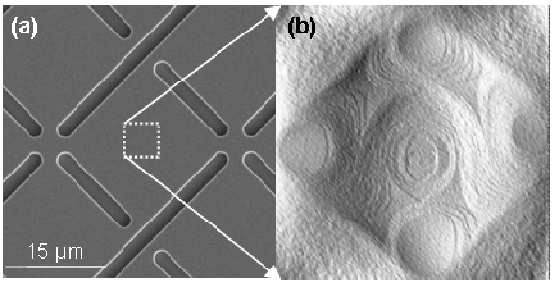

FIG. 1: SEM micrograph of the patterned surface and an AFM image (6µm × 6µm) taken after 1200 °C flash heating inside one of the cells marked by the box. The atomic scale ordering of the steps are nearly identical in the center of each cell on.

The etched micrometer scale patterns underwent considerable deformation via diffusion, sublimation and re-adsorption of surface atoms. Self organization of the step terrace patterns appears to occur on the constrained layer of adatoms within the micro-patterns transforming the surface to this final pattern. The pyramidal structure in the center is an outcome of the interactions between neighboring steps and the confined geometry during the kinetic process[14]. Previous simulation studies show self organization of vicinal steps in the presence of stress[15]. In those reports the surface is under tensile stress parallel to the dimmers and under compressive stress in the perpendicular direction[16]. Step formation occurs to relax the strain associated with the atomic dimers on the reconstructed surface[17]. Etched patterns serve as boundaries to confine the dynamics promoting self organization within individual cells.

It is not yet known if this state corresponds to a global equilibrium or if the surface is frozen at a meta- stable state as the temperature is reduced rapidly after the 1200 °C anneal. The structures were reproducible across different cells within the pattern and also across other samples under identical thermal processing conditions.

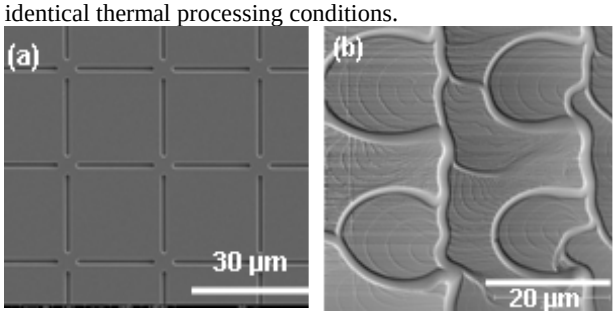

FIG. 2. (a) Scanning electron micrograph of the 25 µm square pattern. (b) Corresponding AFM image after 1050 °C anneal of 18 hours. The current direction is from left to right.

In the second set of experiments the samples were annealed at 1050 °C for an extended period of time (4 hours – 18 hours) following the initial flash heating at 1200 °C. Figs 2(b) and 3 (b) shows the AFM images corresponding to the etched patterns shown in Figs 2(a) and 3(a) respectively. The images demonstrate consistent self-ordering of step-terrace patterns in all the cells. The direction of current flow in both images is from left to right. The area outside the etched patterns has also undergone step bunching, although there is no specific ordering.

The data show that the step flow patterns are directly associated with the lithographically etched micro-scale feature. Each of the cells has transformed into regions with regular pattern of equidistant step train. The steps are organized in the form of 'arcs' connecting the step bunches. The lines shown by arrows in both images 4b and 4b are atomic steps (conventional D_A type double steps). These steps are separated by atomically flat regions.

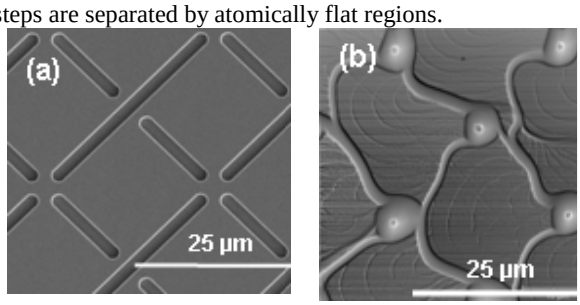

FIG. 3. (a) Scanning electron micrograph of the 25 µm square at 45 degrees. (b) Corresponding AFM image after 1050 °C anneal of 18 hours. The current direction is from left to right.

The total z- scale corresponding to the Fig. 3b is less than 50nm and that of Fig. 4b is less than 100nm. All the trenches originally had an etch depth of 1µm before annealing. These data suggest that there is a considerable amount of mass flow reducing the height difference of the sample. The micro scale patterns cause different mass transport kinetics due to thermodynamics effects in combination with electromigration effects from the heating current.

The Si (100) vicinal surface reconstructs with alternating 1×2 and 2×1 terraces causing a diffusion anisotropy. It has been shown that the diffusion parallel to the dimmer rows is faster resulting in anisotropic diffusion[18]. The step-step interactions (generally repulsive) originating from the intrinsic stress cause the surface to reach an equilibrium state[16]. This prevents the unconstrained surface from forming step bunches. In addition, the adatoms do not cross a step in the upward direction, a case generally referred to as the Ehrlich-Schwoebel (ES) barrier. This is the reason why ordinarily one does not see step bunching in flash heated samples. However, with extended period annealing at high temperatures the steps become permeable to the adatoms overcoming the ES Barrier leading to the upward crossing of the steps and gaining large diffusion lengths[19]. The direct current heating induces a preferential diffusion direction to the adatoms. The combination of these two effects causes the step bunching where all the steps are swept aside and accumulated leaving atomically flat terraces between them. For short annealing times on the order of minutes, the effect of electromigration is not very prominent; however the effect becomes substantial with extended annealing times of several hours. In this case the adatoms are supplied from the step edges of the lower terrace and the wide terraces tend to broaden further[20].

The lithographically etched micro-scale patterns on the sample serve as pinning sites for the steps to accumulate and bunch leaving the entire area within the patterns atomically flat with only very few steps. By lithographic process, the steps are confined within the smaller regions which help to remove the random fluctuations (usually associated with very long steps) and facilitate the formation of regular step arrays. It can be seen from Fig. 4 that the ordered step-flow patterns are originated in the deep trenches in the pattern. There is enough material flow into the trenches from the plateaus to reduce the height difference. As material flows, the trenches widens with the formation of atomically flat regions. The flat regions of the trenches undergo a process similar to a silicon epitaxial growth where layer by layer growth occurs.

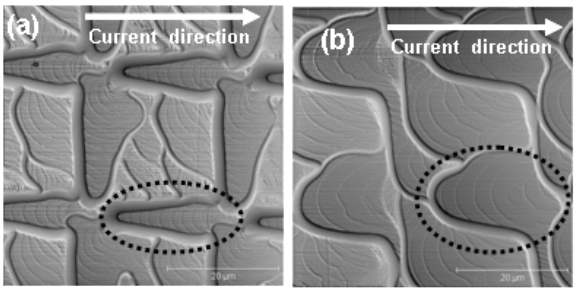

FIG.4: AFM images showing the evolution of the step-terrace patterns originating from the trenches as shown by the dashed circles. The images are taken at two different times during the annealing.

Although there are wide atomically flat regions, we do not observe the repeated step-flow patterns in the 50 µm patterns. This indicates that the size of the patterns is an important factor as it serves to constrain the mass flow and promotes a self organized morphology of step-terrace patterns.

The surfaces that we obtain by this process are ordered at the atomic scale. We have used a UHV Omicron[10] STM to image several samples under nearly identical conditions. As can be seen in Figure 5 the surfaces undergo dimmer row reconstruction and there are only a few vacancies and defects present as the surfaces evolve layer by layer resembling an epitaxial growth process.

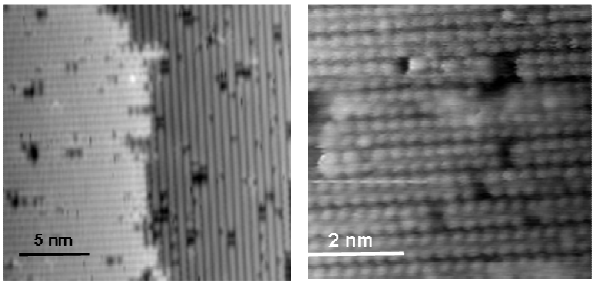

FIG.5: High resolution image of the samples acquired using scanning tunneling microscope. Figures show long range atomic scale ordering with dimmer row reconstruction.

It has been shown that for larger miscut angles (>5°), the surface contains predominantly double-layer steps[21]. It is also possible to have coupling of adjacent steps due to anisotropic diffusion caused by the direct current heating[22]. As shown in Fig.4, we have observed that the wide terraces are always separated by double steps. Pairing of steps leave the large terraces with the same orientation 1×2 or 2×1 (D_A type or D_B type). Our data consistently show that the area between the step-bunches has the same orientation because of the double steps, making the surface ideal for large area atomic scale manufacturing purposes[23]. The 1×2 and 2×1 orientation can be switched by reversing the current direction in agreement with data reported earlier[24].

In summary we have shown that microscale patterns on the surface facilitate the dynamics of the surface to attain a very ordered step-terrace pattern. The surface is constrained within the boundaries defined by the pre-fabricated patterns. Electromigration of adatoms is accomplished by annealing the surfaces at 1050 °C for an extended period of time. The process results in reproducible step-terrace patterns with long-range atomically ordered surfaces.

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