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Comparison of structural re-organisations observed on pre-patterned vicinal Si(1 1 1) and Si(1 0 0) surfaces during heat treatment

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Abstract

The creation of distinct, periodically structured vicinal Si(1 1 1) and (1 0 0) substrates has been studied using scanning tunnelling microscopy at various temperatures. The vicinal Si(1 1 1) and (1 0 0) surfaces transform under heat treatment in a self-organised way into flat and stepped regions. Optical and electron beam lithography is used to produce a regular hole pattern on the surfaces, which interferes with the temperature-driven morphological changes. The step motions are strongly influenced by this pre-patterning. Pre-patterned Si(1 1 1) surfaces transform into regular one-dimensional (1D) and two-dimensional (2D) morphologies, which consist of terraces and arrangements of step bunches and facets. On pre-patterned Si(1 0 0) substrates different re-organisations were observed where checkerboard-like 2D structures are obtained.

Keywords: Pre-patterning; Vicinals; Two-dimensional reorganisation; Template formation

1. Introduction

There is a great technological interest in the controlled preparation of structured silicon surfaces as potential templates for the growth of periodic nanostructures [1]. Many different approaches to surface structuring have been described and can be divided into two principal groups. One approach takes exclusively advantage of the surface re-constructions induced through intrinsic properties of the material to control the growth of adsorbates [2,3]. Another approach relies on nanofabrication by scanning probe microscopy [4] and focused ion beam patterning [5]. These methods suffer from the limited area (∼100 nm × 100 nm), which can be structured. Other preparation schemes rely on subsurface stressors, which interfere with the surface through elastic fields [6]. Also, an aligned array of etched holes on vicinal Si(1 1 1) was used to modify the Si surface morphology successfully [7,8]. Hasegawa et al. [9] used such tailored step configurations to illustrate the influence

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of the atomic steps on the surface conductivity. Much less work has been reported on the patterning of Si(1 0 0), where the aim was mainly to obtain large, step-free surfaces [10–12]. In our work, we combine the lithography approach of Ogino et al. [7,8] with our prior work on the local atomic re-arrangements of concave silicon surfaces, which were prepared by dimple grinding [13–15]. In the present paper, we focus on first results obtained for different pattern orientations with respect to the $[1\,1\,\bar{2}]$ direction for Si(1 1 1). We present a example of the resulting morphologies after annealing a pre-patterned Si(1 0 0) surface. A more detailed study, which discusses the influence of the heat treatment on the surface re-organisation will be published elsewhere [16].

2. Experimental

Regular hole patterns have been created on slightly misoriented 2 inch silicon (1 1 1) and (1 0 0) wafers by either optical lithography or by electron beam

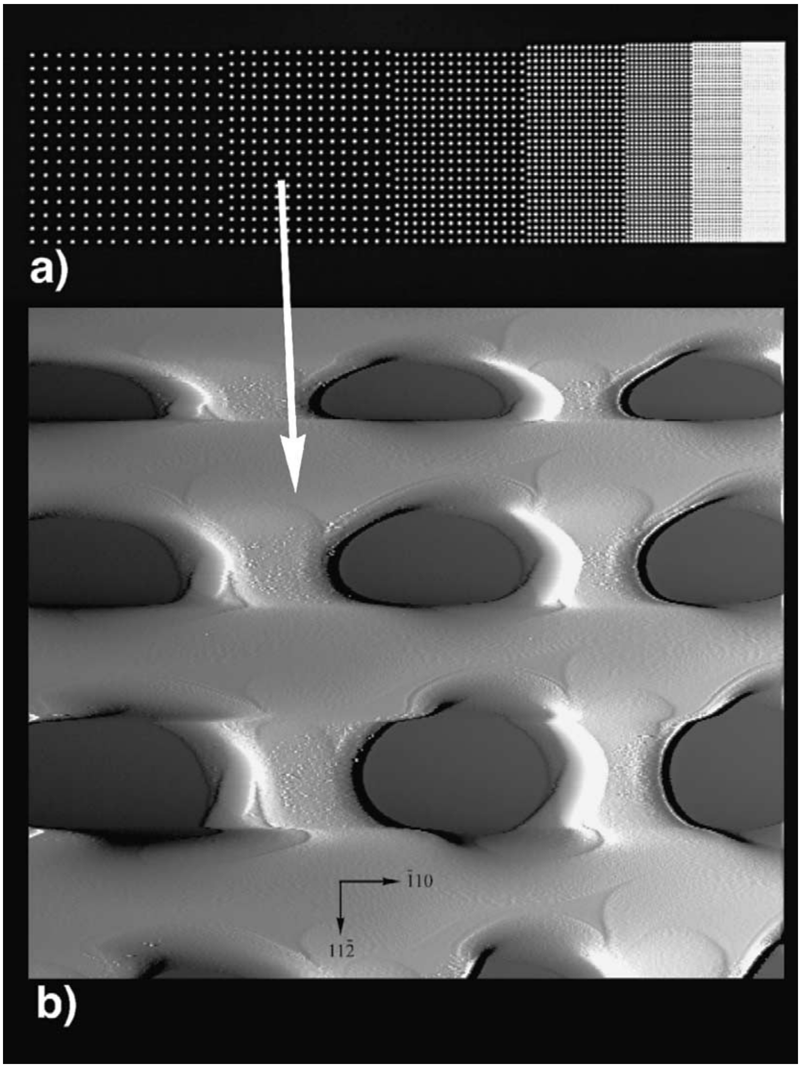

Fig. 1. (a) Sketch of one hole region in the mask used for the optical lithography experiments (see text for more details). (b) A typical hole pattern in Si(1 1 1) obtained with the mask in (a) after heating the sample to 1200 °C for 10 s, four times. Imaged area: 7.4 μm × 8.2 μm.

lithography. For the former, a specially designed mask was used. It was composed of 16 distinct areas with rectangular hole-arrays, each characterised by a given hole diameter and seven different hole spacings. The hole diameter ranges from 1 μm to 30 μm over the 16 hole-arrays. Fig. 1(a) shows a sketch of the hole array. This gives rise to arrays with increasing hole density from one side of the patterned zone to the other. The transfer of the hole pattern from the mask to the silicon surfaces was done by optical lithography followed by either wet chemical etching in HNO_3, HF and CH_3COOH or by reactive ion etching (RIE) with SF_6. The depth of the etched holes could be varied between 200 nm and 1 μm, depending on the etch process. An example for the etched hole pattern on Si(1 1 1) is presented in Fig. 1(b), where the transition from a hole distance of 27 μm (left side) to 18 μm (right side) for a 9 μm diameter hole-array is shown. The cross-sectional profile for each hole is close to rectangular after RIE and close to concave-shaped

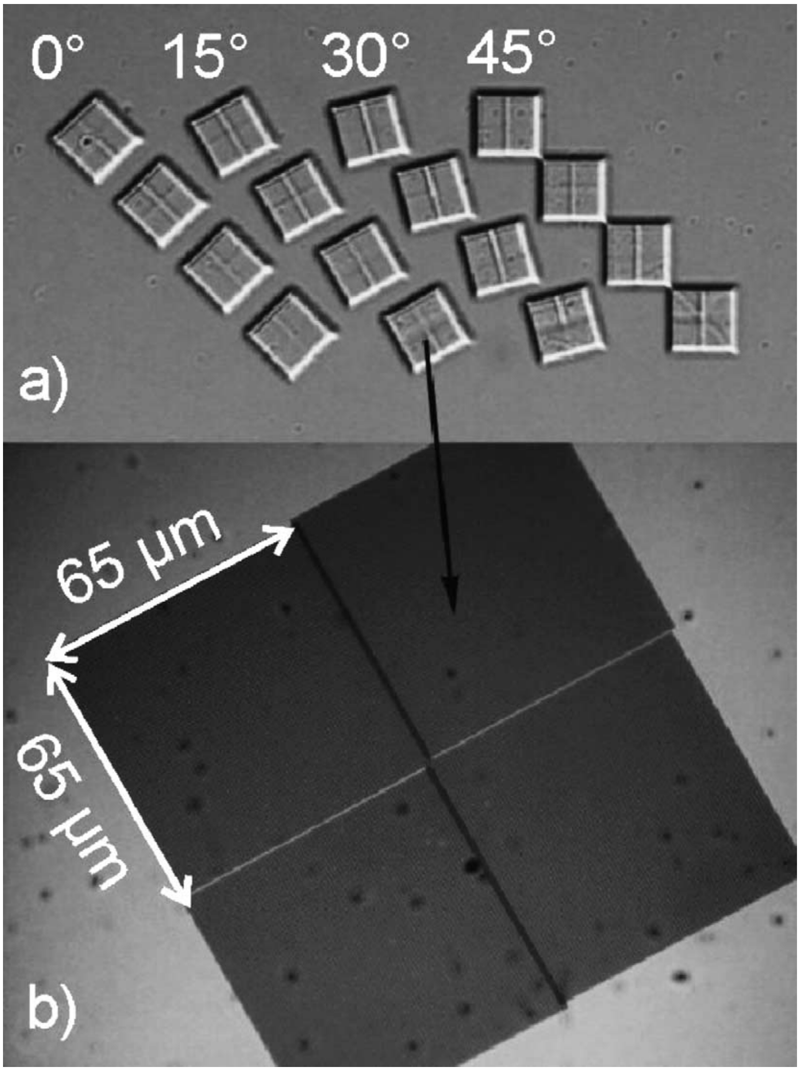

Fig. 2. Hole pattern obtained on Si(1 1 1) with electron beam lithography. (a) Etched regions with varying orientation in the (1 1 1) plane. The labels give the deviation from the $(1\,1\,\bar{2})$ direction. (b) Zoom into one patterned region (see text for more details).

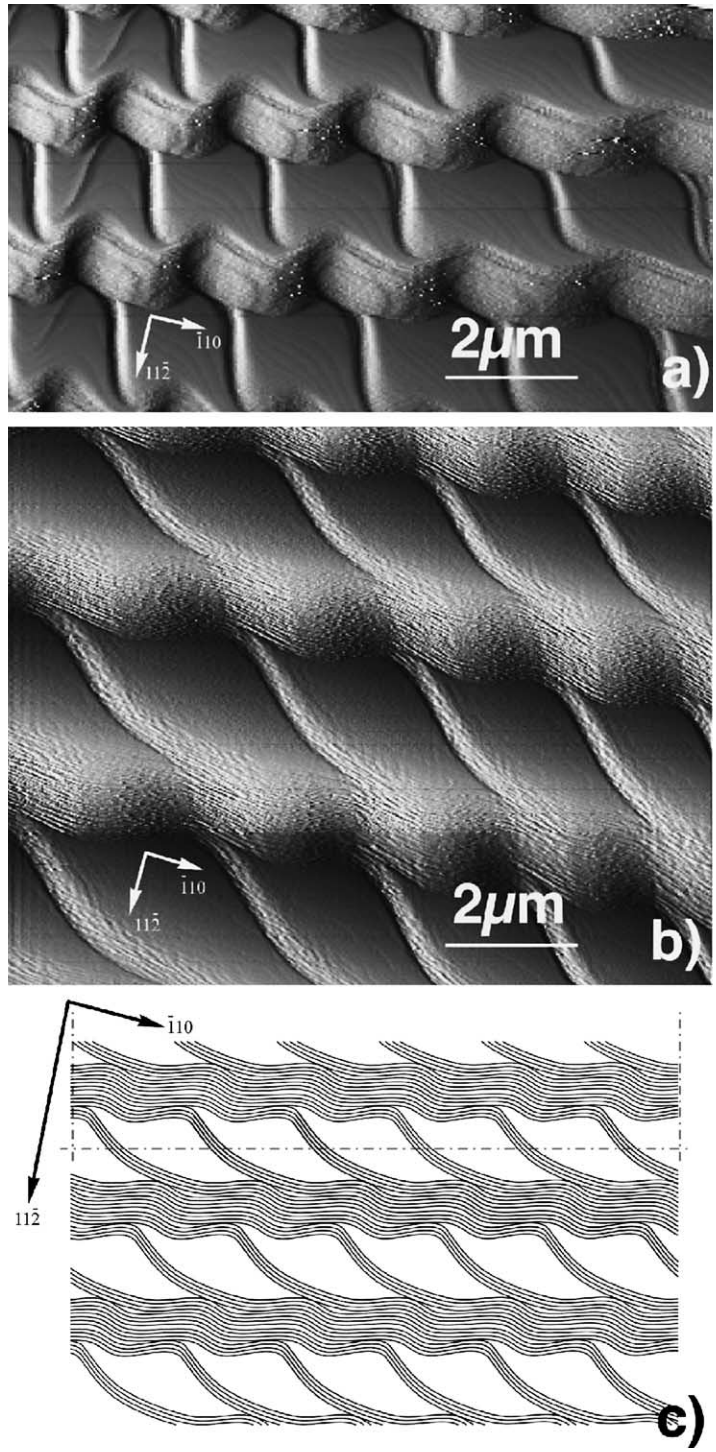

Fig. 3. STM-images taken at high substrate temperature on patterned Si(1 1 1) surfaces. (a) Sample heated 90 s at 1200 °C and 5 min at 1100 °C. (b) Same sample heated additional 5 min at 1100 °C. (c) Schematic drawing of the observed 2D re-organisation.

after chemical etching, as checked by scanning electron microscopy observations of cross-sectional samples. Fig. 1(b) shows one patterned area after heating to 1200 °C for 10 s, four times. The hole pattern of Fig. 1(a) was either oriented along $[1\,1\,\bar{2}]$, or rotated in-plane by an angle between 15° and 45°, as shown in Fig. 2(a). Sub-micrometer hole sizes were produced by electron beam lithography, with typical diameters and pitches (hole spacings) of 500 nm, for an overall period of 1 μm in the hole-array. The patterned areas consist of squares of 65 μm side length, as shown in Fig. 2(b). The Si samples (*n*-type, resistivity 0.5 Ω cm–1 Ω cm) used in this work are misoriented by 1.5° towards the $[1\,1\,\bar{2}]$ direction for Si (1 1 1), 1.5° in the [1 1 0] direction for Si (1 0 0). Samples of 5 mm × 15 mm, cut within the different hole-arrays, were pre-cleaned in methanol prior to mounting on a Ta holder in the STM chamber. They were resistively heated by a d.c. current, which flows perpendicular to the misorientation direction. The samples were carefully outgassed at 300 °C, and then at 600 °C for 12 h and subsequently flashed to 1200 °C in intervals of 15 s. Morphological changes were studied in situ by STM, either after cooling down to room temperature or in a variable temperature STM for temperatures between 500 °C and 900 °C. The experimental methods used for this study have been presented in more detail elsewhere [17].

3. Results and discussion

Depending on the azimuthal misorientation direction, vicinal Si(1 1 1) surfaces re-organise under UHV-heat treatment into two different morphologies. When misoriented along the $[1\,1\,\bar{2}]$ direction, small terraces separated by mono- and triple-layer high steps are formed. For misorientations along the opposite $[1\,1\,\bar{2}]$ direction, vicinal surfaces transform into larger terraces, separated by step bunches. The present results were obtained on the latter misorientations, and step bunch formation was expected and is found in the experiments. Regular hole pattern with holes of 2 μm diameter and varying pitches were etched into the Si(1 1 1) surface. An angle of about 30° was introduced between hole alignment and the crystallographic $[1\,1\,\bar{2}]$ direction in the (1 1 1) plane. The as-prepared surface was heated under UHV by passing a direct current through the sample for different time intervals. Characteristic morphological re-organisations have been obtained. Fig. 3 shows two STM images obtained on this surface in a region with a pitch width of 2 μm. The observed morphology can be explained by an overlap of the initially etched hole pattern and the temperature-driven morphological re-organisation. We observe a tendency of the surface to form step bunches along the $[1\,1\,\bar{2}]$ direction, which are separated by rather flat terraces. The image in (a)

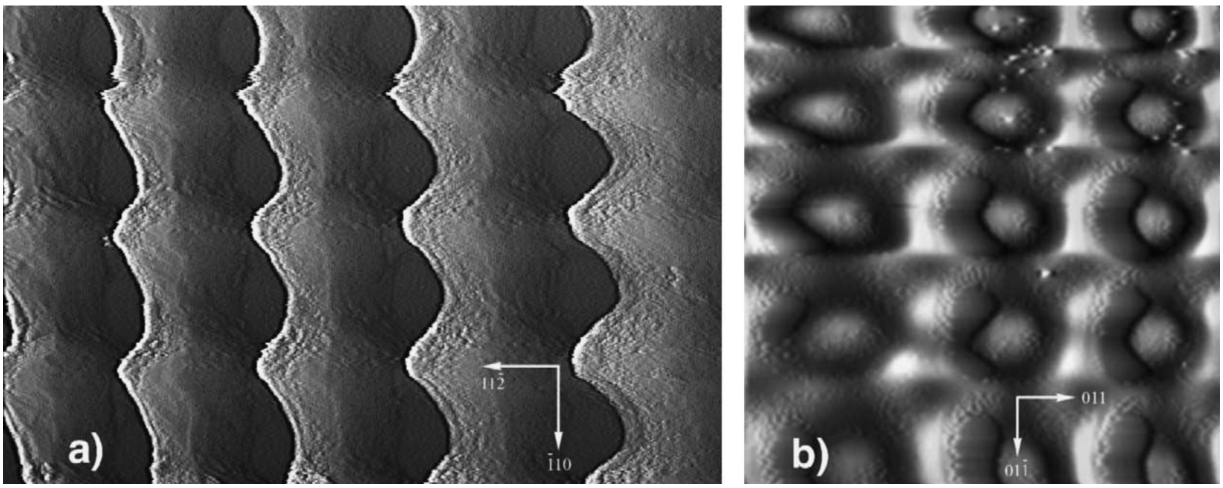

Fig. 4. Comparison between patterned Si(1 1 1) in (a) and Si(1 0 0) in (b) surfaces. Imaged area in (a) is 7.8 μm × 5.2 μm and in (b) is 3.8 μm × 3.8 μm.

was obtained after a heat treatment of 90 s to 1200 °C and an anneal to 1100 °C for 5 min, the image was taken at 600 °C. The wavy bunches indicate the location of one part of the inner wall of the pre-patterned holes. After this initial heat treatment, the surface shows still a strong zig-zag corrugation along the step bunches. The original step bunch formation gives rise to minor step bunches crossing the terraces. Additional heating of the same sample to 1100 °C for another 5 min smoothens the in-plane corrugation of all step bunches, major and minor ones, as is shown in Fig. 3(b) (image taken at 600 °C). Fig. 3(c) shows a sketch with lines representing the step bunches.

Further experiments were performed with the hole pattern aligned parallel to the $[1\,1\,\bar{2}]$ direction. The obtained morphological changes give rise to 1D re-organisations of the surface in flat terraces of uniform width separated by straight step bunches. Fig. 4(a) reproduces an image of the surface re-organisation at an early stage. The STM image was recorded at 600 °C, and was obtained after direct current heating of the sample to 1200 °C for 40 s, followed by annealing to 1000 °C for 1 min. The re-organisation has not yet reached the final configuration. For these heating conditions, the terrace separation is already very regular, but the step bunches are not yet straight, but still heavily corrugated. We compare this result to a patterned Si(1 0 0) surface shown in (b). This surface was flashed several times to 1250 °C. A very different re-organisation is obvious. The initial 2D hole pattern persists. At the location of the pre-patterned holes, deep bumps are found, which give rise to a checkerboard-like up-down morphology.

4. Summary and outlook

The preparation of patterned Si(1 1 1) and Si(1 0 0) surfaces with periodic structures of adjustable repetition length is possible. The periodic structures can extend in one or two dimensions. Due to the interference of the imposed hole pattern and the ‘‘natural’’ re-organisation of the surfaces, reduced periodicities can be obtained. In the contrary to Si(1 1 1) surfaces, re-organisations on Si(1 0 0) surfaces always showed a 2D periodicity. These results will be described in more detail elsewhere [16].

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