Phenomenology of iron-assisted ion beam pattern formation on Si(001)

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New Journal of Physics 13 (2011) 073017 (20pp)
Received 29 March 2011
Published 12 July 2011
Online at \url{http://www.njp.org/}
doi:10.1088/1367-2630/13/7/073017

Abstract. Pattern formation on Si(001) through 2 keV Kr\textsuperscript+ ion beam erosion of Si(001) at an incident angle of $\vartheta = 30^\circ$ and in the presence of sputter co-deposition or co-evaporation of Fe is investigated by using \textit{in situ} scanning tunneling microscopy, \textit{ex situ} atomic force microscopy and electron microscopy. The phenomenology of pattern formation is presented, and experiments are conducted to rule out or determine the processes of relevance in ion beam pattern formation on Si(001) with impurities. Special attention is given to the determination of morphological phase boundaries and their origin. Height fluctuations, local flux variations, induced chemical inhomogeneities, silicide formation and ensuing composition-dependent sputtering are found to be of relevance for pattern formation.

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1. Introduction

Ion beam erosion is an efficient and versatile technique for creating regular nanoscale surface patterns over large, wafer-size areas. Such structured surfaces may be used as templates for subsequent nanostructure fabrication (e.g. in nanomagnetism [1–5]) or show altered physical properties of interest (e.g. diminished reflectivity for optical surfaces [6–8]).

In past years, it became obvious that trace amounts of impurities—stemming from the ion source itself or sputter deposited from the sample surroundings—influence pattern formation, even for such simple situations as noble gas ion erosion of Si [9–12]. Subsequently, it was found that no patterns develop on Si surfaces in a large angular range of ion incidence $0^\circ \leq \theta \leq 50^\circ$ with respect to the surface normal if no impurities are present or co-deposited [13–15]. All patterns observed in this angular range—in the past, often erroneously interpreted in terms of pure ion beam erosion mechanisms—are just a consequence of unintentional co-deposition of a non-volatile species during erosion, resulting in a two-component surface system. These findings not only prepare the ground for careful and clean ion beam erosion experiments, which may serve as an empirical basis for a theoretical description of single-component ion beam erosion, but also provide a strong parameter to tune patterns and to develop new ones, qualitatively different from the patterns observed in a one-component situation [15–17].

In recent years, also pattern formation theory has discovered the relevance of multicomponent systems for expanding the opportunities and our understanding of ion beam patterning [18, 19]. Even co-deposition situations are now analyzed theoretically [20, 21].

Here, we investigate the phenomenology of pattern formation on Si(001) using sputter co-deposition of stainless steel as a function of temperature. We take advantage of the fact that in the deposition geometry used, gradients result in the concentration of co-deposited stainless steel species and of primary noble gas ions reflected from the sputter target to the sample surface. These gradients give rise to a pattern manifold as a function of sample location in a single-shot experiment. The experiments extend our previous room temperature work on sputter co-deposition [15]. It is the goal of our analysis to separate and distinguish the most relevant factors for ion-beam-induced pattern formation with co-deposition. Knowledge of such factors is a prerequisite for an adequate modeling of this situation. Key findings described below relate to the effect of chemical reactions on pattern formation, to critical fluctuations causing a pattern bifurcation and to local flux variations of the co-deposited species causing its inhomogeneous lateral distribution.
2. Experimental

The experiments were performed in a variable temperature scanning tunneling microscopy (STM) apparatus [22] with a base pressure of $< 6 \times 10^{-11}$ mbar. It is equipped with a differentially pumped fine focus ion source, a Faraday cup, a load-lock for sample transfer and a low-energy electron diffraction system and allows full sample temperature control during ion exposure and STM imaging.

After the removal of the protective adhesive tape, the Si sample was mounted at its edges to the sample holder through three tungsten leaf springs. Prior to insertion to the load lock, dust was blown off with a He gas beam. Samples cleaned this way display a roughness of $\leq 0.2$ nm, after removal of the native oxide through a normal incidence ion fluence of $F = 2 \times 10^{20}$ ions m$^{-2}$. For the sputter co-deposition experiments, a piece of angle iron made from stainless steel (Fe 84% and Cr 13%) was mounted on the Si(001) sample by clamping it together with the Si wafer under two of the three tungsten leaf springs. The stainless steel piece exposed an area 1.4 mm high and 6 mm wide to the ion beam, vertical with respect to the Si sample. For reproducibility and prior to each experiment, this area received a finish with P180 sand paper and supersonic rinsing in propanol. For brevity, we will refer to the co-deposited material in text and figures frequently as ‘Fe’. A 2 keV Kr$^+$ ion beam with a full-width at half-maximum of $\approx 1$ mm was then scanned over the sample and the steel target, resulting in simultaneous erosion and metal deposition on the Si sample. The ion beam was impinging at an angle $\vartheta = 30^\circ$ with respect to the surface normal onto the Si sample and with an angle $\vartheta_t = 60^\circ$ onto the stainless steel target (compare figure 1(a)). The scanned area was 4 mm in the direction parallel to the stainless steel plate. In the direction along the $x$-coordinate as indicated in figure 1(a), the ion beam was covering the exposed area of the stainless steel piece and a distance of almost 6 mm on the sample. Without the stainless steel piece the scanned area would have been from $x = -1.2$ mm to $x = 5.8$ mm. The time averaged flux of primary Kr$^+$ ions arriving at the Si sample was $\Phi_{Kr} = 3.4 \times 10^{17}$ ions m$^{-2}$ s$^{-1}$. Within the entire scan area the uniformity of the ion flux was better than 10%; within the area 0.5 mm away from the edges of the scan area it was better than 5% as measured by the movable Faraday cup. Prior to and after each experiment, the ion flux was checked by the Faraday cup. The ion fluence to the sample was $F = 5.9 \times 10^{21}$ ions m$^{-2}$. Note that fluxes and fluences are specified as values on the sample plane and not in a plane normal to the ion beam, as frequently specified. Due to differential pumping during ion exposure, the working pressure was below $1 \times 10^{-7}$ mbar. After ion exposure, the pressure dropped quickly into the $10^{-11}$ mbar range.

We also performed co-evaporation experiments using the same stainless steel material as for sputter co-deposition. The ion species, energy, fluence and flux were identical to the ones used for sputter co-deposition. In these experiments, no stainless steel plate was mounted on the Si sample. Instead, stainless steel was co-evaporated from an e-beam evaporator during ion exposure. The evaporator was calibrated through a quartz crystal micro balance and controlled during evaporation through the evaporation temperature measured by a thermocouple directly attached to the evaporating stainless steel plate. The amount of co-evaporated material is characterized by the atom-to-ion arrival ratio $\Phi_{Fe}/\Phi_{Kr}$.

After preparation, the samples were imaged in situ by STM at room temperature or at 140 K for the low-temperature erosion experiment. Additionally, the sample topography was analyzed ex situ by atomic force microscopy (AFM), scanning electron microscopy (SEM) and 400 keV transmission electron microscopy (TEM). The surface structure was investigated
Figure 1. (a) Sketch of the sputter co-deposition setup. The gray shading of the upper layer of the substrate schematically visualizes the concentration of sputter co-deposited atoms; dark: high concentration; light: low concentration (see text). (b) Maximum iron concentration $c_{Fe}$ measured during SIMS depth profiling as a function of distance $x$ from the stainless steel plate after 300 K sputter co-deposition. The line is a guide to the eye.

ex situ by grazing incidence x-ray diffraction (GI-XRD). The surface chemical composition was analyzed by time-of-flight secondary ion mass spectrometry (ToF-SIMS) using 15 keV Ga$^+$ and 250 eV O$_2^+$ as well as with high lateral resolution by electron energy loss spectroscopy (EELS) within a dedicated STEM operated at 100 keV acceleration voltage. Quantitative image analysis was conducted by using WSxM [23]. Sputter yield calculations for Si and Fe–Si compounds were performed with TRIM.SP [24].

3. Results

As a prerequisite for the discussion of the temperature-dependent ion erosion experiments, we recall some basic consequences of the deposition geometry already presented in a previous publication [15], where this geometry was used for room temperature sputter co-deposition. As shown in figure 1(a), the primary ion beam hits the sample at an angle $\vartheta = 30^\circ$ with respect to the sample surface normal. At the same time, the stainless steel target is hit at an angle $\vartheta_t = 60^\circ$ with respect to the local surface normal of the target. Sputtered Fe and Cr atoms leave the target with a broad angular distribution and the most probable emission angle of $\vartheta_t \approx 37^\circ$ (compare figures 6(a) and (c) of [15] for the angular and energy distributions). These atoms give rise to a substantial flux of arriving atoms on the sample. This flux diminishes with increasing normal distance $x$ from the stainless steel plate. In addition, primary Kr$^+$ are scattered at the target surface. They leave the target surface with substantial energy, a rather narrow angular
distribution and a most probable emission angle $\theta_1 = 68^\circ$ (compare figures 6(b) and (c) of [15] for the angular and energy distributions). Consequently, in the region adjacent to the stainless steel plate with $0 \leq x \leq 1$ mm and indicated as (I) in figure 1(a), a large flux of scattered Kr atoms and sputtered atoms arrives at the sample, in addition to the primary Kr$^+$. Although the concentration of sputter co-deposited atoms measured after room temperature ion exposure is in this area significant (compare figure 1(b)), the surface remains rather smooth with a roughness $\sigma \approx 0.3$ nm, comparable to the initial roughness of the wafer, but slightly larger [15]. In the region indicated as (II) in figure 1, with $x > 1$ mm the flux of Kr$^+$ scattered from the target is absent. Consequently, at $x \approx 1$ mm the concentration of co-deposited Fe displays a sharp rise to $c_{\text{Fe}} \approx 0.2$ in figure 1(b), a sharp rise in roughness and onset of pronounced pattern formation after room temperature ion exposure. The concentration of the sputter co-deposited material, roughness and pattern fade with increasing distance $x$ from the stainless steel plate.

Figure 2 gives an overview of the temperature-dependent morphologies as a function of $x$. The three rows correspond to the erosion temperatures of 140, 300 and 440 K (from bottom to top); the four columns correspond to distances $x \approx 0.5$ mm, $x \approx 2.5$ mm, $x \approx 3$ mm and $x \approx 4.5$ mm (from left to right). It is obvious from figure 2 that the patterns in each column are similar, irrespective of the temperature during ion exposure: the first column with $x \approx 0.5$ mm (figures 2(a), (e) and (i)) represents a flat surface with randomly distributed holes. The roughness of this hole pattern is low with $\sigma \approx 0.25$ nm. At 440 K, the hole pattern is somewhat more pronounced and has a slightly higher roughness of $\sigma \approx 0.35$ nm. The depth of a hole is 1–1.5 nm as is visible in figure 2(m), which displays the height profile along the line in the inset of figure 2(a). The second column with $x \approx 2.5$ mm (figures 2(b), (f) and (j)) represents a pattern of short ripple segments with wave vector parallel to the projection of the ion beam on the sample surface. The roughness of the ripple patterns shown is $\sigma \approx 3$ nm, with a typical corrugation between valleys and ridges of 10 nm. The ripple wavelength is around $\lambda = 50$ nm and varies only weakly with temperature. As is apparent from the height profile along the line in figure 2(f) shown in figure 2(n), the ripples are asymmetric with a steep flank facing the incoming sputter co-deposition flux. Typical angles between the local surface normals of the facet and the primary ion beam are $65^\circ$ (facet facing the sputter co-deposition flux) and $15^\circ$ (facet facing the primary ion beam). The third column with $x \approx 3$ mm (figures 2(c), (g) and (k)) represents dot patterns. The roughness of the dot patterns is much lower than that of the ripple patterns and is $\sigma \approx 1$ nm. The dot density increases slightly with temperature corresponding to a decrease of the typical dot–dot spacing from 45 nm at 140 K to 40 nm at 440 K. Also the dots are asymmetric with a steep flank facing the incoming sputter co-deposition flux. The height profile along the line in figure 2(c) is shown in figure 2(o). Typical angles between the local surface normals of the dot facets and the primary ion beam are $50^\circ$ (facet facing the sputter co-deposition flux) and $20^\circ$ (facet facing the primary ion beam). Based on our SIMS measurements for the 300 K sample, the Fe concentration of the sputter co-deposited material in the surface layer has reduced from its peak value at the onset of the ripple pattern range of $c_{\text{Fe}} \approx 0.2$ to $c_{\text{Fe}} = 0.01–0.02$ (compare figure 1(b)). The fourth column with $x \approx 4.5$ mm (figures 2(d), (h) and (l)) represents a flat surface without pattern. The roughness is $\sigma \approx 0.2$ nm and has a comparable value as would be obtained for ion erosion without sputter co-deposition.

Figure 3 represents the result of a systematic roughness analysis based on AFM topographs measured ex situ. Within the AFM it was possible to vary the $x$-coordinate systematically with high precision at the disadvantage of lower resolution and postoxidation of the wafer. It is apparent that the roughness has for all three temperatures a pronounced and broad maximum
Figure 2. STM topographs of Si(100) taken after a fluence of $\approx 5 \times 10^{21}$ ions m$^{-2}$ 2 keV Kr$^+$ in the sputter co-deposition geometry. The temperature of ion exposure is fixed for each row: (a–d) 440 K, (e–h) 300 K and (i–l) 140 K. The distance $x$ normal to the stainless steel plate and the height difference $\Delta z$ spanned by the image contrast between white and black are indicated above the columns. Note the large differences in $\Delta z$ for the four columns. The black and white arrows in (d) visualize the projected directions of incoming sputtered Fe atoms and of the primary Kr$^+$, respectively. The topograph size is always 500 nm $\times$ 500 nm; the size of the insets in (a), (e) and (i) is 100 nm $\times$ 100 nm. Height profiles along the lines in the inset of (a) and in (f) and (c) are shown in (m), (n) and (o), respectively.

reaching $\sigma = 10$–20 nm in the $x$-range between 1 and 2.5 mm, i.e. in the $x$-range where the ripple patterns form. Some scatter in the roughness data is visible around $x \approx 1$ mm, caused by the abruptness and inhomogeneity of the pattern changes in this range (see below).
Figure 3. The roughness $\sigma$ as a function of normal distance $x$ to the stainless plate for the erosion temperatures indicated (see text). The lines are a guide to the eye.

Figure 4. AFM topographs after sputter co-deposition at 140 K taken ex situ at 300 K at the following positions: (a) $x = 1.12$ mm, (b) $x = 1.14$ mm, (c) $x = 1.15$ mm and (d) $x = 1.45$ mm. The directions of the incoming co-deposited Fe and Kr$^+$ projected onto the surface are indicated in (d). The height difference $\Delta z$ spanned by the image contrast between white and black is 100 nm for all images. The topograph size is always $3 \mu$m $\times$ $3 \mu$m.

From figure 3, it is apparent that until now we have not yet presented any topographic data from the rough region with $1 \text{ mm} \leq x \leq 2.5 \text{ mm}$. The reason is simply that—due to the roughness in this range—STM topographs free from tip artifacts are almost impossible to record. To discuss this $x$-range, we therefore refer to AFM and SEM data. Figure 4 displays a sequence of AFM topographs with increasing $x$-coordinate after ion exposure at 140 K and measurement at 300 K. By comparison of topographs taken in situ at 140 K by STM and taken ex situ at 300 K by AFM in the smoother regions with $x \leq 2.5$ mm, we find no differences of significance due to the annealing to room temperature and the exposure to ambient conditions. The sequence of AFM topographs shows that the transition from the hole pattern to the ripple pattern is not gradual and smooth, but abrupt and inhomogeneous. It does not have a single sharp boundary, but rough and flat patches coexist for a narrow range of $x$-coordinate values.
Figure 5. SEM images after sputter co-deposition at 440 K. Panels (a)–(c) are top view images taken at (a) $x = 1.12$ mm, (b) $x = 1.19$ mm and (c) $x = 1.24$ mm. The numbers in (a) refer to different stages of the pill bug evolution (see text). Panels (d) and (e) are tilt-view SEM images. The directions of the incoming co-deposited Fe and Kr$^+$ are indicated in (a) (projected onto the surface) and in (d). Image widths are (a) 4.4 $\mu$m, (b, c) 2.8 $\mu$m and (d, e) 2 $\mu$m.

Figure 4(a) with $x = 1.12$ mm displays on a flat hole pattern background with a very low roughness of $\sigma = 0.2$ nm (similar to that seen in figure 2(i)) the rise of characteristic rippled objects. The rippled structure of such an object mimics the segmented exoskeleton of a pill bug, a brief term used below for this feature. Moving towards larger values of the $x$-coordinate, the concentration of the pill bugs increases in figure 4(b) with $x = 1.14$ mm until they percolate in figure 4(c) at $x = 1.15$ mm. Only small patches of the flat hole pattern are left. Figure 4(d) displays an AFM topograph of the fully developed ripple pattern taken at $x = 1.45$ mm.

The morphological phase transition from the flat area with shallow depressions to the rough ripple pattern is temperature independent. The sequence of SEM topographs of a sample after sputter co-deposition at 440 K shown in figure 5 displays precisely the same pattern transition as the sequence of AFM topographs of figure 4 after sputter co-deposition at 140 K. With increasing $x$-coordinate, the flat hole pattern starts to become interrupted by oval-shaped pill bugs (figure 5(a)), which become denser and start to coalesce (figure 5(b)) until a full ripple pattern forms (figure 5(c)). We stress here that for both temperatures there is no coexistence of different evolution levels within the same area. The transition always displays, with increasing $x$-coordinate, a monotonic increase in the pill bug concentration until they merge to form a ripple pattern. The $x$-position of the transition line depends slightly on the $y$-position, which is likely the result of the finite size and the irregularities of the Fe target. Figure 5(a) highlights also the different stages of pill bug evolution. The numbers (1)–(4) indicate pill bugs of increasing...
size containing three to six well-developed ridges, respectively. Number (5) suggests that pill bugs tend to nucleate at surface irregularities. In the presented case (5), it appears that multiple nearby nucleation centers caused the formation of three overlapping pill bugs. The tendency for the preferred nucleation of pill bugs at irregularities is supported by the observation of a higher density of pill bugs at a surface scratch (not shown).

The tilt-view SEM images in figures 5(d) and (e) make it plain why AFM measurements display roughness variations at the onset of the ripple regime: not only is the transition from the flat hole pattern to the ripple regime abrupt and inhomogeneous, but the depth of the ripple trenches is very large as well. Their depth is considerably underestimated by AFM due to tip limitations. Therefore the roughness data shown in figure 3 have to be considered with care, especially in the range $1 \text{ mm} \leq x \leq 2 \text{ mm}$. It is visible from the tilt-view SEM images that the ripple ridges are bounded by steep walls almost parallel to the incoming ion beam. The ripple pattern visible in the tilt-view SEM image of figure 5(e) displays also a considerable height variation along the ridges, correlated along the $x$-coordinate. These variations are consistent with the idea that the ripple pattern emerges through coalescence of growing, initially isolated pill bugs.

Figure 6 highlights the features of individual pill bugs. The two AFM topographs of figures 6(a) and (b) show two pill bugs fabricated at 440 and 140 K, respectively. Their common features are: (i) pill bugs have a characteristic extension of 600 nm along the $x$-coordinate and 300 nm normal to it. (ii) Their maximum height above the flat background is 100–150 nm. (iii) As shown in the profile of figure 6(c), their height linearly slopes down from its maximum value at their head (large $x$-coordinate) towards their tail (small $x$-coordinate). (iv) The segmenting of their exoskeleton is uniform with a characteristic wavelength that is slightly temperature dependent. It is 65 nm at 140 K and 75 nm at 440 K. (v) The head of a pill bug usually consists of a short ridge directed precisely towards the ion beam (compare the SEM image in figure 5(d)), i.e. with an angle of 30° with respect to the surface normal. As is visible in figure 5(d), the segments of the exoskeleton are in fact ridges separated by deep trenches. The ridges are inclined towards the ion beam, similar to the pill bug head. (vi) The pill bugs cause depressions in their surroundings. This is especially visible in figure 6(a), where the trenches between the ridges extend into the flat region with a depth of 5–10 nm.
There are slight differences between pill bugs fabricated at different temperatures. Most obvious, pill bugs prepared at 440 K are more regular and oval-shaped, while the pill bugs created at 140 K are less regular with a tendency to a triangular appearance. At 440 K, the ridges, trenches and their extension into the surroundings are slightly curved, with an imaginary center of curvature on the $x$-axis somewhere well behind the pill bug head. The trenches extending into the flat area and the depressions in front of the pill bug head remind us of patterns caused by the drag of an object through a highly viscous material. The pill bugs appear to be moved backwards, i.e. in the direction of decreasing $x$-coordinate.

Figure 7 summarizes the discussion on the phenomenology of patterns in a morphological phase diagram. We distinguish four fundamental types of surface areas as a function of the $x$-coordinate: the flat hole regime with negligible roughness within region (I) exposed simultaneously to scattered Kr$^+$. Within region (II), three regimes are found: the rough ripple regime, the dot regime, and the flat surface. The transition between the latter three morphological phases is continuous: with increasing $x$-coordinate, the ripple pattern gradually transforms to a dot pattern, which eventually vanishes as the roughness approaches the limiting value of about 0.2 nm. Therefore, these pattern transitions are not quantitatively defined, but depend slightly on visual judgment. In contrast, the transition between holes and ripples is not gradual but locally abrupt with a narrow zone of increasing ripple area fraction, i.e. of increasing pill bug concentration, which causes their coalescence to a ripple pattern. The location of this phase transition is strictly temperature independent, while for the other transitions a tendency for a shift towards lower $x$-coordinate with increasing temperature is visible.

Figure 8(a) is a cross-sectional bright-field TEM image of a ripple pattern at $x \approx 2.5$ mm created at 300 K; figure 8(b) is a high-resolution micrograph of the area indicated by the white box in figure 8(a). The brightness of the amorphous layer varies due to the concentration variation of the sputter co-deposited material. The darker patches result from the stronger electron scattering of the elements Fe and Cr with higher atomic number. It is apparent that the sputter co-deposited material has its highest concentration on the facet facing the incoming
Figure 8. (a) Cross-sectional bright-field TEM image of ripples generated by sputter co-deposition at 300 K. The dashed white lines highlight the ion beam amorphized Si layer. The area enclosed by the white box is shown as a high-resolution TEM image in (b). The circle in the valley at the origin of the left arrow and the circle close to the ripple ridge at the origin of the right arrow indicate the selected areas where EELS spectra were taken, which are displayed in (c) and (d), respectively. The arrows labeled Kr and Fe give the directions of the primary ion beam and the sputter co-deposited material flux, respectively.

Particle flux as indicated by the black arrow in figure 8. This interpretation is confirmed by EELS spot analyses (beam diameter: 1 nm) of a bright location in the amorphous surface layer (circle at the origin of the left arrow in figure 8(a)) and a dark location (circle at the origin of the right arrow in figure 8(a)), which are represented in figures 8(c) and (d), respectively. The EELS spectrum in figure 8(c) displays a strong oxygen O-K core loss peak and a Fe-L$_{2,3}$ ionization edge of considerably lower height. In contrast, the EELS spectrum of figure 8(d) is dominated by the peak of the Fe-L$_{2,3}$ shell ionization, an additional Cr-L$_{2,3}$ loss peak is visible and the loss peak of the O-K shell ionization is diminished. Apparently, the enhanced concentration of the sputter co-deposited material on the facet facing the stainless steel plate in turn reduces also post oxidation.

In the high-resolution TEM image of figure 8(b), the crystalline structure of the substrate is visible. In addition, within the Fe-rich parts of the amorphous layer, small crystalline patches are visible, which we interpret as indications of nanocrystalline Fe silicide.

4. Discussion

Pattern type and sequence of patterns observed with increasing $x$-coordinate are temperature independent. The pattern transition positions are only a weak function of temperature. Roughness differences for the same $x$-position are lower than a factor of two. Wavelength or
dot spacings agree to within 20%. Based on these facts we conclude that thermally activated processes are largely irrelevant for pattern formation under sputter co-deposition conditions. Any thermally activated process relevant for pattern formation at room temperature would either decrease or increase its frequency by orders of magnitude through either lowering or increasing the temperature by about 150 K as pursued here. As thermally activated processes of diffusion or reaction are irrelevant for the pattern formation with sputter co-deposition, it is probable that the same holds for ion-induced pattern formation on Si(001) in the absence of co-deposition. Slight temperature dependences just outside the experimental scatter are present in our data. The shape of the pill bugs changes and becomes more regular and the pattern transition lines within region (II) shift toward smaller $x$ with increasing temperature. Both effects may be due to thermally activated processes just becoming visible at 440 K (although not yet relevant) or due to a physical effect with a weaker dependence on temperature than an activated process. One plausible speculation is to assume that the slight changes observed are due to a decrease in viscosity of the amorphous layer with temperature.

The largely temperature-independent pattern formation agrees with the findings of Gago et al [25]. Based on their temperature-dependent dot pattern investigations in the range of 300–525 K after normal incidence sputtering with unintentional impurity sputter co-deposition, they state: ‘In the low-$T$ range $\leq$ 400 K, the pattern is not significantly affected by $T$ . . . ’. Also, Ozaydin-Ince and Ludwig [12] find, in their normal incidence Mo seeding studies in the entire temperature range from 20 to 300 °C, only moderate changes of roughness and wavelength by less than a factor of two. These statements do not fully coincide with a subsequent normal incidence study by Sánchez-García et al [26], who find already between 300 and 400 K a significant change in their hole pattern roughness by a factor of three. It may well be that these differences are linked to the exact nature of the sputter co-deposited material. But all three studies agree on the absence of pronounced or even qualitative changes of patterns with temperature in the temperature range around room temperature.

Within region (II), where the concentration of sputter co-deposited steel gradually vanishes to zero with increasing $x$-coordinate, smooth pattern transitions from ripples to dots to a flat surface are observed. One may therefore speculate that these pattern transitions are caused by the decreasing ratio of arriving sputter co-deposited Fe versus primary Kr$^+$. To test this hypothesis we conducted dedicated experiments, where we controlled the concentration, energy and angle of incidence of the arriving co-deposited material. In these experiments, stainless steel was evaporated onto the Si sample while it was ion beam eroded. The stainless steel plate and consequently sputter co-deposition were absent. As shown in figure 9(a) the ion beam direction was not changed compared to the sputter co-deposition situation, and Fe impinges at an angle of incidence of 75° with respect to the surface normal. Despite being well defined, this angle is a rough approximation to the angular distribution of incidence for Fe in the sputter co-deposition experiments for $x = 1$–4.5 mm. The sequence of patterns with decreasing flux ratio $\Phi_{\text{Fe}}/\Phi_{\text{Kr}}$ represented by figures 9(b)–(d) is the same as the sequence obtained within region (II) with increasing $x$-coordinate in sputter co-deposition. This similarity is highlighted by the insets of figures 9(b)–(d), which are taken from the 300 K sputter co-deposition experiment. Even the asymmetric shapes are similar. This is highlighted by a height profile through dots in figure 9(c), which is shown in figure 9(e). We conclude that the details of the angular and energy distributions of the impinging Fe are irrelevant, as long as the global direction of Fe impingement and the ion beam parameters are the same. We find here the flux ratio $\Phi_{\text{Fe}}/\Phi_{\text{Kr}}$ to be a key parameter of pattern selection.
In a recent publication, Zhang et al [17] claim that the saturation Fe coverage is decisive for the pattern selected in a sputter co-deposition situation (termed surfactant sputtering by them). The authors find that for 5 keV Xe\(^+\) ion exposure and simultaneous Fe sputter deposition, a saturation coverage is quickly reached and established for fluences of 1 \times 10^{21} \text{ ions m}^{-2}. Assuming that the saturation coverage grows monotonically with the flux ratio \(\Phi_{\text{Fe}}/\Phi_{\text{ion}}\), this finding agrees with our observations derived from both sputter co-deposition and co-evaporation. However, below it will be shown that the flux ratio (or the saturation coverage) is only a good parameter for the description of pattern formation, as long as the other variables are fixed, specifically the directionality of the impinging Fe particles with respect to the ion beam.

We now turn to one key issue of this paper, namely the origin of the pattern phase transition between regions (I) and (II), i.e. the transition between a flat surface marginally patterned through irregular shallow depressions (holes) and a rippled surface with a large roughness in excess of 10 nm and a corrugation of the order of 100 nm. Disregarding for the moment the real space aspect of this transition, we note that it coincides with a transition from simultaneous exposure of Si(001) to direct and scattered energetic Kr atoms for \(x \leq 1 \text{ mm}\) in region (I) to exposure only by direct Kr\(^+\) in region (II). This transition is unambiguous from the erosion step between regions (I) and (II) as measured by optical interferometry (compare figures 7(a) and (b) of our previous publication [15]). Despite the additional bombardment by scattered Kr atoms, in region (I) plenty of Fe is present. The concentration \(c_{\text{Fe}} \approx 0.1\) is much higher than for \(x \approx 3 \text{ mm}\), where a pronounced dot pattern is observed. We are left with the conclusion that to understand this difference, we have to consider the directionality of the energetic particles with respect to the Fe flux rather than the concentration (or saturation coverage) of Fe. Specifically, we hypothesize that if a component of the energetic particles comes from a similar direction as the Fe flux, the tendency for pattern formation is strongly diminished.

To test this hypothesis, we again conducted controlled co-evaporation experiments. In figure 10, we compare three co-evaporation experiments with a flux ratio \(\Phi_{\text{Fe}}/\Phi_{\text{Kr}} = 0.04–0.05\),
Figure 10. (a) Dependence of pattern formation on the directions of deposition flux and ion beam. (a–c) Sketches of deposition geometry indicating the angles of deposition flux and ion beam with respect to the surface normal. (d–f) STM topographs after co-evaporation during a fluence of $6 \times 10^{21}$ ions m$^{-2}$ of 2 keV Kr$^+$ at 300 K. The atom-to-ion flux ratios are (d) $\Phi_{Fe}/\Phi_{Kr} = 0.04$, (e) $\Phi_{Fe}/\Phi_{Kr} = 0.05$ and (f) $\Phi_{Fe}/\Phi_{Kr} = 0.04$. Height differences $\Delta z$ spanned by the image contrast are (d) 56 nm and (e, f) 2 nm. Image size is always 1 µm × 1 µm.

which differ only in the incidence angles of the 2 keV Kr$^+$ and of the Fe as sketched in panels (a)–(c). Figure 10(d) is identical to figure 9(b) where the ion beam and Fe flux enclose an angle $\alpha = 105^\circ$ and a faceted ripple pattern forms. The situation mimics the sputter co-deposition within region (II). If we keep the direction of incidence of the Fe flux fixed and move the ion beam close to it, such that $\alpha = 30^\circ$, no pattern forms as shown by the STM topograph of figure 10(e). This situation corresponds to the sputter co-deposition in region (I), but without the direct Kr$^+$ flux. Alternatively, if the ion beam is kept in its position but the Fe flux is moved close to it, such that $\alpha = 30^\circ$, again no ion beam pattern forms as shown in figure 10(f). It appears that the angle $\alpha$ between the ion beam and Fe flux is a decisive quantity in pattern formation and that a large $\alpha$ fosters pattern formation.

Zhang et al [17] state in their work on sputter co-deposition of Fe on Si(001): ‘Therefore the initial steps of pattern formation are caused by none of the following: directional effects of the Fe-deposition, directional effects of the ion beam, and . . .’. While it would be a non-trivial task to demonstrate that pattern formation is determined solely by the directionality of the impinging particle fluxes, from our experiments it is obvious that the directional effects of Fe-deposition and ion beam are a decisive factor for pattern formation also in the initial stages of pattern formation. It turns out that co-evaporation experiments with unique and variable directions of the particle fluxes are key to a deeper understanding of impurity-induced pattern formation.

Our finding that the angle $\alpha$ between the ion beam and impurity flux is relevant and should be large for efficient pattern formation is consistent with the observation of dot formation at normal incidence ion erosion. If the impurities are largely supplied from the sample mount, $\alpha$ may be close to 90°. Similarly, for grazing incidence ion bombardment impurities supplied to a given location may be supplied from the ion beam-illuminated parts of the sample mount, thereby resulting in $\alpha$ well above 90°.
Table 1. Partial silicon sputter yield $Y_{\text{Si}}$, total sputter yield $Y$, density $\rho$ and erosion rate $Y/\rho$ as a function of the sample composition.

| Composition | $Y_{\text{Si}}$ (Si atoms per ion) | $Y$ (atoms per ion) | $\rho$ (atoms nm$^{-3}$) | $Y/\rho$ (nm$^3$ ion$^{-1}$) |
|-------------|-----------------------------------|---------------------|--------------------------|----------------------------|
| Fe          | 0                                 | 3.54                | 84.8                     | 0.042                      |
| Fe$_3$Si    | 0.75                              | 2.77                | 88.8                     | 0.031                      |
| FeSi        | 1.28                              | 2.40                | 89.0                     | 0.027                      |
| FeSi$_2$    | 1.56                              | 2.23                | 79.8                     | 0.028                      |
| Si          | 1.77                              | 1.77                | 50.0                     | 0.035                      |

Our observations also do not necessarily rule out pattern formation if the ion beam and co-deposition flux come from similar directions (i.e. $\alpha$ is small). If the flux ratio $\Phi_{\text{impurity}}/\Phi_{\text{ion}}$ is high enough or the ion fluence is large enough also in such a situation a pattern may arise. But our observations indicate that using a large angle $\alpha$ is much more efficient for co-deposition-induced pattern formation and allows one to minimize flux ratio and ion fluence. This finding may be of relevance in view of the application of sputter co-deposition for patterning with the goal of large corrugations, as achieved in the present study with only moderate ion fluences. It also implies that morphologies resulting from ion beam experiments are more tolerant to impurities originating from the ion source itself than they are to impurities sputtered from the sample environment.

Finally, we note that the absence of shallow holes in figures 9(b) and (c) seems to indicate that an influx of energetic particles from different orientations is necessary for hole formation, as in region (I) during sputter co-deposition.

The next relevant observation based on the EELS analysis shown in figure 8 is that the sputter co-deposited material is non-uniformly distributed over the patterned surface. In fact, the material forms a chemical pattern in phase with the topographic pattern. The same conclusion was obtained already several times [17, 29] and was obvious also much earlier from bright-field TEM images that displayed dark shadows on the ripple ridges in registry with the pattern (compare [27, 28]). These dark shadows are due to enhanced electron scattering at the high-atomic-number impurities.

We conducted additional GI-XRD investigations on the 440 K sample and found no trace of a signal for any of the crystalline iron silicide phases. Iron silicides have substantial negative enthalpies of formation in the range from $-20$ to $-30$ kJ mol$^{-1}$ [30] with a maximum of 32.5 J mol$^{-1}$ for FeSi [30]. We therefore assume that although no long-range order exists, depending on the Fe concentrations the local atomic arrangements are close to those of the corresponding iron silicides. This assumption is consistent with the findings of Sánchez-García et al, who conclude, on the basis of a chemical shift in XPS, that, upon Fe sputter co-deposition on Si, iron silicide forms [26]. Using TRIM.SP, and the structure data compiled by Moroni et al [31], we calculated for 2 keV Kr$^+$ incident with $\vartheta = 30^\circ$ the erosion rate for the different iron silicides and the pure elements. As surface binding energy for the different compounds in TRIM.SP we used the cohesive energies calculated in the generalized gradient approximation of density functional theory by Moroni et al [31]. As compiled in table 1, the erosion rate is minimal for FeSi and is for all silicides lower than that for elemental Si. Assuming a steady state situation, the sputtered Fe is replenished through co-deposition. The partial Si yield is in all cases lowered through Fe co-deposition. Our inferences do not depend critically on the local...
angle of incidence. Similar differences in sputtering yield and erosion rate are present in a broad angular range between 0°–60°. Therefore, a height modulation with iron silicide accumulation close to the elevations of the pattern will be amplified by the composition dependence of the erosion rate. Our detailed analysis agrees in this respect with that of Zhang et al [17].

According to the picture developed by Zhang et al [17], phase separation initiates pattern formation. Phase separation enabled by ion-induced mobility is assumed to create a chemically modulated surface which is transformed into a height pattern by a composition-dependent sputter yield. Only then, if a coupled pattern of composition and height is established, directed deposition of Fe enhances the pattern contrast or pattern corrugation. This plausible picture is in agreement with our considerations on the composition-dependent erosion rate and with numerous observations of ion-induced phase separation. As an example, radiofrequency sputter deposition of silicides on silicon substrates results in well-ordered and -aligned rod patterns [32].

However, this picture is hard to reconcile with three of our observations. (i) In region (I) for \(x \leq 1\) mm the Fe concentration is 0.08–0.09 and only a faint irregular pattern with a marginal roughness \(\sigma \leq 0.35\) nm develops (compare figure 1(b) and figure 2). In contrast, in region (II) at \(x = 3\) mm the Fe concentration is only 0.01–0.02, but a pronounced dot pattern with \(\sigma \approx 1\) nm is present. If phase separation were decisive, one would expect that the additional mobility supplied by the scattered Kr atoms and the much higher Fe influx and steady state concentration in region (I) would lead to a much faster phase separation than at \(x = 3\) mm in region (II). Consequently, one would expect a well-developed and well-ordered pattern in region (I) and a pattern of inferior quality for the area at \(x = 3\) mm, contrary to observation. (ii) The phase boundary between regions (I) and (II) visualized in figures 4–6 is abrupt and inhomogeneous, showing pill bugs in flat areas. It is not obvious how an initial phase separation could be reconciled with such a heterogeneous pattern landscape composed of large flat areas with localized pattern patches. (iii) For identical deposited amounts of Fe and identical ion beam exposure conditions, patterns arise or not, depending only on the angle between the ion beam and deposition flux (compare figures 10(a) and (c)). It is not clear how the direction of the impinging Fe on an initially flat surface could determine the occurrence of a phase separation as such.

We propose therefore a scenario for pattern evolution which involves also height fluctuations as a necessary ingredient of pattern formation. In the spirit of a linear stability analysis, we assume a sinusoidal surface profile resulting from a height fluctuation as sketched in figure 11. For pure Si, the profile amplitude will decrease in time until it vanishes. This holds as long as \(\theta \leq 55°\) [15] and thus specifically in our case with \(\theta = 30°\). If the ion flux and the Fe deposition flux come from the same direction, locations that receive the highest Fe flux receive also the highest ion flux, a fact that tends to diminish buildup of concentration variations. However, if (as indicated) the ion flux and the Fe flux enclose a large angle \(\alpha\), locations that receive the highest Fe flux receive the lowest ion flux. Thereby not only concentration differences, but also erosion rate differences build up and the pattern is amplified. Whether this simple view captures all essentials of co-deposition-induced pattern formation on Si(001) might be tested by energetic Fe⁺ ion bombardment. Exposure of Si(001) to Fe⁺ with sufficient energy to cause net erosion, i.e. with energies of a few keV, and for not too large angles \(\theta\), should not cause pattern formation.

We note that, in agreement with an analysis by Zhang et al [17], our observations and the derived pattern formation scenario provide no trace of indication for the relevance of curvature-dependent sputtering and a stress-induced instability as proposed by Zhou and Lu [21]. We find
Figure 11. (a) Sketch of co-deposition with a large angle between the Kr\(^+\) ion beam and Fe deposition flux for a sinusoidal surface profile. In this situation, the highest Fe flux arrives in profile locations, where the erosion flux is minimal. Consequently, the surface profile destabilizes with time \(t\) (see text).

Figure 12. Sketch of a pill bug formation scenario. Steps (i)–(vi) are discussed in the text.

A height fluctuation to induce a modulation of the chemical composition, which in turn amplifies the amplitude of the height fluctuation and thereby initiates pattern formation. Silicide formation itself is a relevant feature of this pattern formation scenario. Its relevance is implicit in table 1. The negative enthalpy of formation of the silicides results in an enhanced cohesive energy and thus an enlarged surface barrier for sputtering, which tends to diminish the Si yield \(Y_{\text{Si}}\). Also, in comparison to pure Si, the silicides have a much higher density, which guarantees their reduced erosion rate.

The phase boundary between region (I) and region (II) cannot be analyzed from the viewpoint of linear stability analysis. The patches of highly corrugated area, i.e. the pill bugs, within a sea of flat phase point to a morphological bifurcation. Inside region (I), all accessible height fluctuations are driven back through the bidirectional ion bombardment. At the edge of region (I), the stabilizing flux of scattered Kr atoms ceases. Still, most fluctuations are driven back, but single, rare and exceptionally large fluctuations bring the system into a strong self-amplification circle of shadowing, silicide formation and corrugation growth around a single height fluctuation. Key to the rapidity of localized pattern formation must be the high Fe flux available in this area. A possible scenario is sketched in figure 12. Starting with a single pointed height fluctuation (i) silicide formation at the exposed flank as well as iron shadowing at the
flank pointing to the ion beam take place (ii). The Fe-exposed flank is eroded more slowly, rises and quickly becomes so steep that the local angle $\theta$ of ion incidence falls beyond the maximum of the erosion rate at $\theta \approx 70^\circ$ (iii). Thereby erosion diminishes, and grazing incidence reflection of ions becomes relevant. Focusing of ions to the foot of the protrusion creates a trench in its immediate surroundings, primarily at its front (iv). While emission of scattered energetic ions is close to the reflection direction, sputtered Fe particles have a broader distribution, but pointed into the forward direction (compare figure 6 of [15]). Thereby Fe accumulates in front of the trench, an exposed flank forms (v) and the process repeats (vi). Although highly speculative, our model accounts properly for a number of prominent features of the pill bugs. (a) The differing numbers of exoskeleton segments represent simply different stages of their growth which takes place segment by segment. (b) The pointed head of the pill bug and the steep ridges with their flanks nearly parallel to the incoming ion beam (compare figures 5(d) and (e)) are consistent with the ion reflection assumption. The decreasing height of the pill bugs towards their tail (i.e. towards the Fe flux) as visible in figure 6(c) is a natural consequence of their growth process as outlined above. (d) The ridge curvature enclosing the initial height fluctuation is a consequence of the two-dimensional distribution of particles scattered from the initial pointed elevation. (e) The key importance of exceptionally large height fluctuations for pill bug formation is underlined by their preferential formation at point (compare figure 5(a)) and line defects (e.g. scratches). We note that similar localized rippled objects have already been observed in the past on Si after 20–40 keV Ar$^+$ ion bombardment with $\vartheta = 45^\circ$ [33, 34].

Instead of a height fluctuation, one might assume that pill bugs nucleate through the formation of silicide seeds. However, given the absence of crystalline iron silicide on our sample as evidenced by x-ray diffraction and given the high Fe concentration, it is not easy to imagine why the formation of an amorphous silicide seed should be a rare event and why its formation should be so efficiently suppressed through additional scattered Kr atoms in region (I).

Our experiments do not provide a clear fingerprint for the nature of the smoothing mechanism, which is necessary for the selection of a characteristic wavelength. As noted above, the appearance of the pill bug surroundings remind us of the pattern caused by the drag of an object through a highly viscous material into the ion beam direction. Therefore it appears plausible that the viscosity of the amorphous layer in conjunction with the surface tension provides the necessary smoothing, in agreement with a recent proposition of Castro and Cuerno [35].

5. Conclusions

We investigated sputter co-deposition and co-evaporation of Fe on Si(001) with the aim to rule out and to identify processes relevant for pattern formation under these conditions. We find thermally activated processes to be irrelevant for 300 K pattern formation. Details of the energy and angular distribution of the co-deposited particles are also irrelevant, as sputter co-deposition and co-evaporation with the Fe flux in a similar direction with respect to the ion beam yield similar patterns. Our observations provide no evidence for the relevance of curvature-dependent sputtering and stress-induced instability for destabilization.

For a given set of ion beam and deposition parameters, it was found that the flux ratio $\Phi_{Fe}/\Phi_{Kr}$ selects the pattern realized. Moreover, the angle $\alpha$ between the ion beam and the Fe deposition was found to be a parameter of significant relevance. A large $\alpha$ fosters pattern formation, whereas for small $\alpha$, pattern formation does not take place for a given flux ratio.
\( \Phi_{Fe}/\Phi_{Kr} \approx 0.04 \). We find a chemical modulation in phase with the height modulation and assume that silicide formation is relevant for pattern formation as it affects the sputtering yields. Pattern formation is assumed to be initiated by height fluctuations, which themselves are amplified by an induced modulation of the chemical composition.

Upon sputter co-deposition, a pattern bifurcation is observed at the location where the flux of scattered Kr atoms ceases. The abruptness and coexistence of fundamentally different patterns side by side indicate that there are conditions where pattern formation does not proceed from a linear to a nonlinear regime, but is a rapid self-accelerated growth process which may be described in terms of pattern nucleation and lateral pattern growth.

Acknowledgments

We acknowledge useful discussions with Hans Hofsäss and Karl-Heinz Heinig and support from Deutsche Forschungsgemeinschaft through Forschergruppe 845. TH acknowledges the access to JEOL JEM 4010 at MPI for Microstructure Physics granted by Peter Werner, and thanks Frank Heyroth (University of Halle) for assistance with EELS measurements.

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