

## **PATTERN FORMATION BY LOW-ENERGY ION BEAM EROSION: FROM ULTRA-SMOOTH SURFACES TO HIERARCHICAL NANOSTRUCTURING**

### **FORMIMI I STRUKTURAVE ME EROZION TË TUFAVE TË JONEVE ME ENERGJI TË ULËT: NGA SIPËRFAQET ULTRA TË LËMUARA TEK NANOSTRUKTURAT HIERARKIKE**

B. ZIBERI, F. FROST, R. FECHNER, M. CORNEJO, J. VÖLLNER, A. SCHINDLER, B. RAUSCHENBACH  
Leibniz-Institut für Oberflächenmodifizierung e. V. (IOM), Permoserstr. 15, D-04318 Leipzig, Germany  
e-mail: bashkim.ziberi@iom-leipzig.de

#### **ABSTRACT**

Low-energy ion beam sputtering, is an inherent part of numerous surface processing techniques. Besides the actual removal of material, this surface erosion process often results in a modification of the surface topography and the formation of well ordered patterns. This self-organized pattern formation is related to a surface instability between curvature dependent sputtering that roughens the surface and smoothing by different surface relaxation mechanisms. If the evolution of surface topography is dominated by relaxation mechanisms surface smoothing can occur. In this paper it will be shown that a multitude of patterns as well as ultra smooth surfaces can develop, particularly on Si surfaces. Additionally the most important experimental parameters that control these processes are discussed. Finally, it will be shown that low-energy ion beam erosion can be combined with lithographic patterning leading multi-scale nanostructuring.

#### **PERMBLEDHJE**

Trajtimi i sipërfaqeve me ane të rezatimit jonik me energji të ulët është pjesë e proceseve të shumta teknologjike. Përveç largimit të materialit nga sipërfaqja, ky proces i gërryerjes së sipërfaqeve rezulton në modifikimin e topografisë së sipërfaqes dhe formimin e strukturave të rregullta. Procesin e formimit të strukturave vete-organizuese është i lidhur ngushtë me jostabilitetin në sipërfaqe ndërmjet procesit të largimit të grimcave që varet nga kurba e sipërfaqes, që çon në vrazhdesimin e saj, dhe lehtësimin të sipërfaqes për shkak të proceseve të ndryshme relaksuese. Nëse evoluimi i topografisë së sipërfaqesore dominohet nga proceset relaksuese atëherë ndodh lehtësimi i sipërfaqes. Në këtë kontribut do të tregohet

se është i mundur krijimi i një shumëllojshmërie strukturash dhe sipërfaqesh shumë të lemuara, veçanërisht në sipërfaqet e Si. Gjithashtu, parametrat me të rendesishëm eksperimentale që dirigjojnë këto procese do të diskutohen. Në fund, do të tregojmë se është i mundur kombinimi i procesit të erodimit me ane të rrezatimit jonik me procesin e strukturimit me ane të litografisë optike që çon në krijimin e strukturave me madhësi të ndryshme.

**Key words:** Patterns, hierarchical nanostructuring, ion beam, self-organization, Si, Ge.

#### **INTRODUCTION**

Today, low-energy ion-beam sputtering, i.e. the removal of atoms from a surface due to the impact of energetic ions or atoms, is an inherent part of numerous surface processing techniques. Thus, sputtering is routinely used in depth profiling analytical techniques and, mostly, it is an integral part of many erosion or deposition techniques, e.g. (reactive) ion beam etching or sputtering and ion beam assisted deposition.

Beside the actual removal of material induced by atomic recoils and the sputtering of atoms from the surface this surface erosion process often results in a pronounced topography evolution, generally accomplished by a kinetic roughening of the surface. Typically, during ion sputtering, the surface of the solid is far from equilibrium and a variety of atomistic surface processes and mechanisms become effective. It is the complex interplay of these processes that either tends to roughen or smoothen the surface, which, finally, can result in a rich variety of surface topographies. Under special circumstances ion beam erosion can, despite the statistical nature of the

process, create well ordered nanostructures on surfaces [1-5].

Two prominent examples are the spontaneous formation of well-ordered ripple and dot pattern. Both special cases are of high interest for many potential applications in nanotechnology, e.g., for cost-efficient production of large-area nanostructured surfaces in a one-step process. In this context, it should be noted that ion beam induced self-organized nanostructures can be observed for a variety of materials (semiconductors, metals, dielectrics), therefore it is a universal process not limited to material classes as self-organization in semiconductor heteroepitaxy. Highly ordered surface structures are promising for quantum dot arrays with specific optoelectronic properties, patterned magnetic media for high-density storage or passive optical elements based on sub-wavelength structured surfaces. In addition to this mainly technological driven motivation, fundamental studies of the pattern formation mechanisms can gain insight into the behaviour of non-equilibrium processes at surfaces and, furthermore of complex processes in nature.

In this contribution our recent activities in the field of tailoring the topography of Si and Ge surfaces at the nanometer and micron scales by low-energy ion beams will be summarized. Examples of low-energy ion beams for self-organized patterning, as well as smoothing of surfaces that can be used for the finishing of high-end optical surfaces with topography and roughness control down to the atomic scale will be presented. At the end, a short outlook will be given about the combination of patterning by self-organization and conventional lithographic techniques for producing hierarchical structures at different length and height scales.

#### EXPERIMENTAL DETAILS

Samples used in this work were commercially available epi-polished Si(100) substrates (p-type and 0.01-0.02  $\Omega\text{cm}$ ), with a root-mean-square (rms) roughness of  $\sim 0.2$  nm. The samples were mounted on a water cooled substrate holder. The angle of the ion beam incidence  $\alpha_{\text{ion}}$  can be varied between  $0^\circ$  and  $90^\circ$  with respect to the surface normal. For the ion beam erosion experiments a home built Kaufman-type broad beam ion source equipped with a two-grid ion optical system (beam diameter 200 mm) was employed, details are given elsewhere [3,6,7]. The ion current density  $j_{\text{ion}}$  was kept constant at about  $300 \mu\text{Acm}^{-2}$  corresponding to an ion flux of  $1.87 \times 10^{15} \text{ cm}^{-2} \text{ s}^{-1}$ .

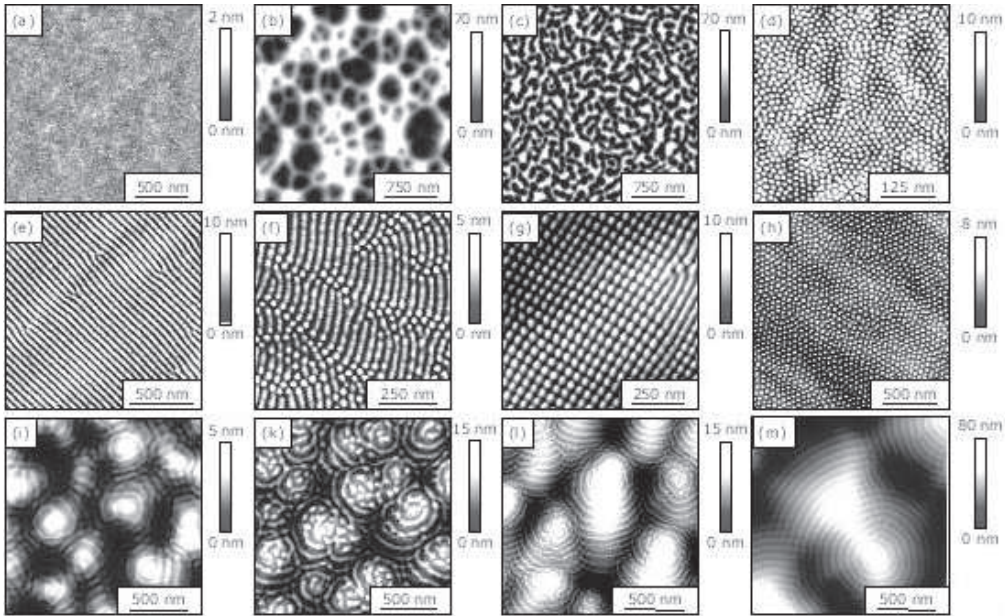
The surface topography was analyzed by Atomic Force Microscope (AFM). The measurements were performed in air using silicon cantilevers with a nominal tip radius less than 10 nm. In order to study the lateral ordering and the characteristic wavelength of ripples the Fast Fourier Transformation (FFT) of the height profile was calculated. From the Fourier transformation the one-dimensional Power Spectral Density (PSD) function was obtained by angular averaging [8].

#### Self-organized patterning of Si and Ge surfaces

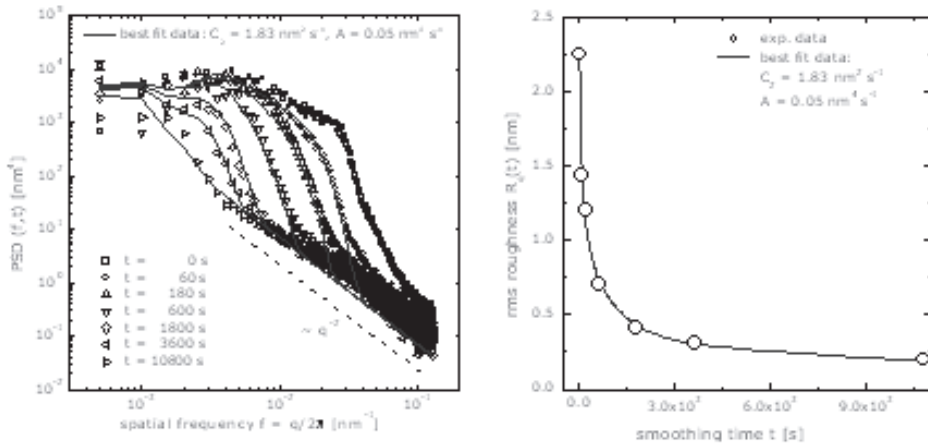
In general, there are many parameters which can play a crucial role for the formation of nanostructures on the surface, especially if broad beam ion sources are used [9-11]. Beginning with the geometrical parameters of the ion-optical system of the ion source, continuing with the extraction voltages applied on the grid systems, and ending with the parameters that influence the primary ion-target interactions. This compilation of process parameters indicates that there are many degrees of freedom for influencing surface topographies arising under ion bombardment. Without going in details Fig. 1 gives a first impression of the diversity of structures and pattern that can result from the ion beam erosion of Si and Ge surfaces. To get a rough idea, the individual patterns are formed under various erosion conditions where different ion energies (between 500 eV and 2000 eV), different ion species ( $\text{Ar}^+$ ,  $\text{Kr}^+$ ,  $\text{Xe}^+$ ), and ion incidence angles (between  $0^\circ$  and  $75^\circ$ ) were used. Partly the ion sputtering was done with simultaneous rotation. On the one hand from Fig. 1 the unique possibilities for tailoring of the surface topography become immediately clear. On the other hand it shows also that there is a big challenge to solve the puzzle of surface evolution under ion beam erosion. In a first step efforts have been made to find the key parameters responsible for the various topographies [11]. At the current state the following key parameters were identified: angle of ion incidence, erosion time, ion beam energy, divergence of the ion beam / angular distribution within the ion beam, ion species used as projectile ( $\text{Ne}^+$ ,  $\text{Ar}^+$ ,  $\text{Kr}^+$ ,  $\text{Xe}^+$ ), temperature, sample manipulation during processing and potential surface contamination.

#### Ion beam smoothing of Si surfaces

In the context of ion beam smoothing, the so-called windows of stability are especially important; this means parameter regimes where surface relaxation dominates the topography evolution (see Fig. 1(a)).



**Figure 1:** Example of diversity of pattern on Si (a, b, d-g, i, m) and Ge (c, h, k, l) surfaces by low-energy ion beam erosion. The following surface topographies measured by AFM are shown: (a) ultra smooth surface, (b, c) meshworks of randomly arranged troughs, (d) domains of hexagonally ordered dots, (e) highly ordered ripple pattern, (f) coexistence of dots and ripples, (g) long range square ordered dots on Si, (h) long range hexagonally ordered dots, (i-m) curved ripples. The individual patterns are formed under various erosion conditions.



**Figure 2:** Ion beam smoothing of a Si surface under  $\text{Ar}^+$  ion beam erosion ( $E_{\text{ion}} = 500 \text{ eV}$ ,  $\alpha_{\text{ion}} = 45^\circ$ ,  $j_{\text{ion}} = 300 \mu\text{A cm}^{-2}$ , simultaneous sample rotation): Power spectral densities (left) and rms surface roughness  $R_q$  (right) for different smoothing times calculated from  $3 \mu\text{m} \times 3 \mu\text{m}$  AFM measurements. The solid line PSD curves were obtained from curve fitting analysis using the model mentioned in the text. The dashed line presents a power law scaling:  $\text{PSD}(q) \propto q^{-2}$ . The initial surface roughness was  $\sim 2.2 \text{ nm}$ . After ion beam smoothing the surface roughness is reduced to value  $< 0.2 \text{ nm}$ .

Applying now ion beams with appropriate values of ion energy and ion incidence angle on initially rough

surfaces, a smoothing is expected [12]. Especially, the time evolution of the rms surface roughness and the

power spectral density (PSD) were analyzed (Fig. 2). From the analysis of the PSD curves, using a model which follows the approach of Bradley and Harper, it has been found that, in the given case, the surface roughening by curvature-dependent sputtering is (over-)compensated by an additional directed flux of surface atoms arising from atomistic drift parallel to the surface caused by momentum transfer. On the basis of this model, the temporal evolution of the surface roughness can be well explained (see 'best-fit data' in Fig. 2). The ballistic drift mechanism was originally proposed by Carter and Vishnyakov to explain the absence of ripple formation at normal or near-normal ion bombardment of Si surfaces with 10–40 keV Xe<sup>+</sup> ions [13].

Finally we will shortly discuss the successful combination of low-energy ion beam induced processes with conventional lithographic patterning techniques to produce new patterns and gain new insight to the process of pattern formation.

### Hierarchical structuring at different length and height scales

Guided self-organization processes are currently in focus regarding their potential for hierarchical micro- and nanoscale structuring, which is of essential importance in various applications of advanced functional surfaces. In this regard, pattern formation due to low-energy ion-beam erosion can offer an alternative approach to nanolithography, especially for the realization of ripple or dot patterns with nanometer scale structure dimensions on the surface of different materials. However, usually this self-organization process lacks long range order and positional control of the pattern. One possibility to improve the ordering and lateral positioning of structures is by using a pre-patterned substrate [14,15]. In this way, due to spatial limitations and guided by the lateral ordering and shape of the pre-patterned templates, the evolving topography often shows an improved ordering. Here we will give some examples that by combining conventional lithographic techniques with ion beam induced self-organization a multi-scale patterning is possible.

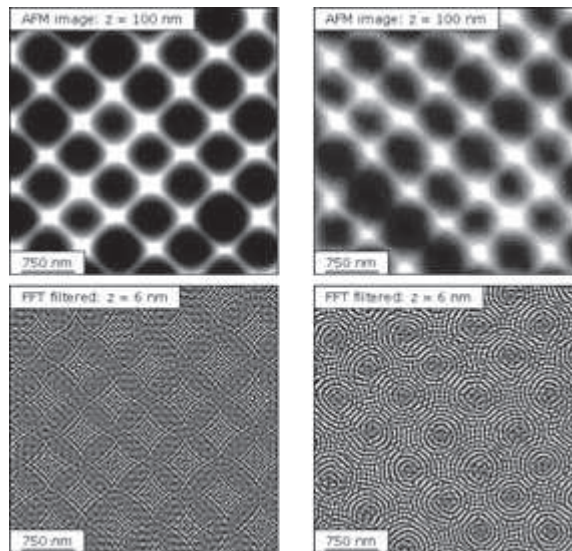


Figure 3: Top: AFM images of Si surfaces with pre-pattern fabricated by Laser ablation and superimposed with self-organized nanostructures induced by ion beam erosion. Bottom: Fourier filtered images (pre-pattern removed) which enlighten the nanopattern formed by self-organization processes during low-energy ion beam erosion. The differences between the left and the right column are only different ion energies and erosion times, respectively.

The pre-patterned substrates are fabricated by different top down techniques with main focus on laser ablation of thin films using phase mask projection for sub-micron pattern formation in combination with reactive ion beam etching techniques for the transfer

of the laser generated patterns into the substrate. In general, it has been observed that the combination of ion beam induced self-organization with conventional lithographic techniques enables, in principle, the formation of new types of patterns, e. g, formation of

curved ripples, circular ripples or nearly perfectly square ordered dots on exact positions on the surface. Additionally, an enhanced ordering of nanostructures and the formation of ripples with different directions depending on the local surface orientation has been found.

The main parameters determining this pattern formation are identified to be the local incidence angle of the ions, the orientation of the local surface with respect to the ion beam direction, and, the local surface curvature. From future exploration on this topic we also expect to gain new insight in the mechanisms of pattern formation itself and, furthermore potential applications, especially in micro- and nanooptics, e. g., in bio-inspired functional nanooptics.

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### REFERENCES

1. Valbusa U, Boragno C, Buatier de Mongeot F, (2002) *J. Phys.: Condens. Matter* 14, 8153.
2. Chan W L, Pavenayotin N, Chason E (2004) *Phys. Rev. B* 69, 245413.
3. Ziberi B, Frost F, Höche Th, Rauschenbach B, (2005) *Phys. Rev. B* 72, 235310.
4. Erlebacher J, Aziz M J, Chason E, Sinclair M B, Floro J A (1999) *Phys. Rev. Lett.* 82, 2330.
5. Chan WL, Chason E, (2001) *J. Appl. Phys.* 101, 121301.
6. Zeuner M, Meichsner J, Neumann H, Scholze F, Bigl F (1996) *J. Appl. Phys.* 80, 611.
7. Tartz M, Hartmann E, Scholze F, Neumann H (1998) *Rev. Sci. Instrum.* 69, 1147.
8. Zhao Y, Wang G-C, Lu T-M (2001) *Characterization of Amorphous and Crystalline Rough Surfaces: Principles and Applications*, Academic Press, San Diego.
9. Carter G (2001) *J. Phys. D* 34, R1.
10. Gago R, Vázquez L, Plantevin O, Metzger T H, Munoz –Garcia J, Cuerno R, Castro M (2006) *Appl. Phys. Lett.* 89, 233101.
11. Ziberi B, Cornejo M, Frost F, Rauschenbach B (2009) *J. Phys.: Condens. Matter* 21, 224003.
12. Frost F, Fechner R, Ziberi B, Völlner J, Flamm D, Schindler A (2009) *J. Phys.: Condens. Matter* 21, 224026.
13. Carter G, Vishnyakov V (1996) *Phys. Rev. B* 54, 17647.
14. Frost F, Ziberi B, Schindler A, Rauschenbach B (2008) *Appl. Phys. A* 91, 551.
15. Cuenat A, George HB, Chang K-C, Blakely JM, Aziz MJ (2005) *Adv. Mater.* 17, 2845.