Minimization of topological defects in ion-induced ripple patterns on silicon

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Abstract. The evolution of self-organized nanoscale ripple patterns induced by low-energy ion sputtering of silicon is investigated. The quality of the patterns is monitored by calculating a normalized density of topological defects from atomic force microscopy images. A strong dependence of the normalized defect density on the applied ion fluence is observed with a well-pronounced minimum at intermediate fluences. Simulations using the damped Kuramoto–Sivashinsky equation yield good agreement with the experiments and are further used to study the dynamics of single pattern defects.

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1. Introduction

The application of nanopatterned substrates to thin film deposition has recently become a popular tool for creating novel nanostructures and controlling the properties of the films. Prepatterened templates were used to produce dichroic thin films [1], ferroelectric nanostructures [2], ordered quantum dot arrays [3] and aligned nanotubes [4]. The

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nanopatterned substrates were mainly fabricated by conventional top-down techniques such as electron beam [2] and atomic force microscope (AFM) lithography [4], or focused ion-beam direct writing [3]. Recently, nanoscale ripple patterns which form by self-organization during oblique low-energy ion sputtering have been used as templates for the deposition of metallic thin films. It was shown that these patterns can induce magnetic [5, 6] and optical [7] anisotropies in the deposited films. The major advantages of this patterning technique are its universality and the easy control of the feature size. Ion-induced ripple patterns have been found on all kinds of materials like metals, insulators and semiconductors, and their lateral dimensions can be tuned in the range from 10 to 1000 nm by altering the process parameters [8, 9]. Ripple patterns similar to those obtained by ion sputtering also arise in semiconductor epitaxy with the ripple wavelength depending on the substrate miscut [10].

However, so far it appears to be difficult to control the regularity of the ion-generated surface patterns. Throughout literature, ripple patterns of very different quality are reported even for identical materials [11]–[14]. This spread in pattern quality can be partly attributed to different experimental conditions. On the other hand, however, it is associated with the stochastic nature of the formation process. For long sputter times, current theoretical models predict a transition from ordered ripple patterns to a random rough surface where lateral order is lost [15, 16]. Such a transition has already been observed experimentally on graphite [17]. In this work, we study the influence of ion fluence on the quality of ripple patterns induced by oblique low-energy ion erosion. For this reason, the dynamics of topological pattern defects was studied experimentally and in simulations. The density of pattern defects is found to depend strongly on the applied fluence. In contrast to epitaxy-induced patterns, where the defect density was found to decrease with increasing film thickness [18], an intermediate fluence regime is identified in both experiments and simulation in which the defect density exhibits a distinct minimum. The existence of such an intrinsic minimum of the defect density of ion-induced ripple patterns offers the possibility to tailor and optimize the quality of nanorippled templates just by varying the sputtering time.

2. Experiment

The experiments were carried out in a vacuum chamber (base pressure \(\sim 10^{-8}\) mbar) equipped with a standard Kaufman type ion source. Because of their technological relevance, commercially available epi-polished Si(100) wafers were chosen as substrates and sputtered with Ar\(^+\) ions of an energy of 300 and 500 eV, respectively. The angle of incidence was fixed at 67° with respect to the surface normal and the flux ranged from \(3.5 \times 10^{14}\) to \(3.5 \times 10^{15} \text{ cm}^{-2}\) s\(^{-1}\). In this range, no influence of the ion flux on the ripple patterns was observed. The applied fluence \(\Phi\) was varied from \(5 \times 10^{16}\) to \(1 \times 10^{20} \text{ cm}^{-2}\) and from \(1 \times 10^{17}\) to \(5 \times 10^{19} \text{ cm}^{-2}\) for 500 and 300 eV, respectively. During the irradiations, the sample temperatures remained below 180°C. Under these experimental conditions, the periodicity of the resulting ripple patterns ranges from 20 to 45 nm depending on ion fluence and energy. After sputtering, the samples were analyzed \textit{ex situ} by AFM.

3. Results and discussion

Figures 1(a)–(c) show three stages of the evolution of the surface under 500 eV Ar\(^+\) bombardment: at low fluences (figure 1(a), \(\Phi = 1 \times 10^{17} \text{ cm}^{-2}\)), short (\(\sim 100–200 \text{ nm on}\)
Figure 1. Experimental (a–c) and corresponding simulated (d–f) surface morphologies at the fluence $\Phi = 1 \times 10^{17}$ (a,d), $1.7 \times 10^{18}$ (b,e) and $1 \times 10^{19}$ cm$^{-2}$ (c,f). The sputtering was done with 500 eV ions. Height scales are 7 nm (a), 10 nm (b,c), 0.07 nm (d), 11.5 nm (e), and 10 nm (f); lateral scales are 1 $\mu$m for each image. The white arrows indicate the direction of the ion beam.

average) ripples with a periodicity of $\lambda \sim 24$ nm are obtained, which are oriented normal to the projection of the ion beam. The ripples become longer with increasing fluence until their average length exceeds 1 $\mu$m and also their periodicity increases. At an intermediate stage (figure 1(b), $\Phi = 1.7 \times 10^{18}$ cm$^{-2}$), larger corrugations overlay the ripple pattern and get more pronounced with increasing fluence. At a fluence of $\Phi = 1 \times 10^{19}$ cm$^{-2}$ (figure 1(c)), the ripple wavelength saturates and the surface enters a steady state. The ripple pattern appears more inhomogeneous now and the ripple wavelength is $\lambda \sim 40$ nm.

In order to quantify the quality of the ripple pattern, we followed the approach of Cuenat et al [19] and calculated a normalized density of pattern defects from the AFM images. After subtracting a quadratic background including the corrugations from each image, the remaining ripple pattern was converted to a binary image (see figure 2(a)). The height threshold was determined using Otsu’s algorithm [20]. The black ripples in the binary image were then thinned to lines with a width of one pixel (figure 2(b)), and every black pixel with more or less than two black neighboring pixels was counted as a defect. Finally, the normalized defect density is given by the total number of defects $N$ multiplied by the square of the ripple wavelength $\lambda$ divided by the scan area $A$, $N_D = N \lambda^2 / A$. Then, a perfect pattern without any defects yields $N_D = 0$, whereas $N_D = 1$ corresponds to a pattern in which each ripple contains one defect per length $\lambda$.

The normalized defect density $N_D$ determined in this way is plotted versus fluence in figure 3. The $N_D$ values are comparable for both energies, although on average $N_D$ appears to be slightly lower for 300 eV than for 500 eV. At the lowest fluence $\Phi = 5 \times 10^{16}$ cm$^{-2}$, the

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Figure 2. AFM image shown in figure 1(b) after (a) conversion into a binary image and (b) thinning the ripples to single pixel lines. Lateral scales are 1 µm for each image.

Figure 3. Normalized defect density for experimental (open symbols, left scale) and simulated (full triangles, right scale) morphologies versus fluence. The error bars represent statistical averages over several samples.

The normalized defect density is around 0.3. With increasing fluence, \( N_D \) decreases until it reaches a minimum value of \( N_D \sim 0.1 \) (500 eV) and 0.07 (300 eV) at a fluence of \( \Phi \sim 2 \times 10^{18} \text{ cm}^{-2} \). Then \( N_D \) increases again until it saturates at \( \Phi \sim 10^{19} \text{ cm}^{-2} \) at a value of \( N_D \sim 0.25 \).

The experiments demonstrate that the ion fluence is a crucial parameter for the optimization of the quality of the ripple patterns. In order to understand the unexpected evolution of the pattern quality with its intermediate optimum, however, one has to study the evolution of single pattern defects. For experimental morphologies, this is almost impossible because one would have to identify and monitor one certain defect during the sputtering process. In simulations, however, one certain surface region can be followed in time and the dynamics of single pattern defects can be studied conveniently.

The formation of ion-induced nanopatterns is described by continuum models like the Bradley–Harper (BH) model [21], which attributes the formation of regular patterns as resulting from the interplay between roughening due to sputter erosion and smoothing by surface diffusion. For the former, a surface instability is induced by the curvature dependence of the
sputter yield, as the local erosion rate is higher in depressions than on elevations. This leads to an amplification of initial surface modulations and, therefore, to roughening of the surface. The competing diffusion process, which tends to smoothen the surface, can be thermally activated or ion-induced [22]. A linear continuum equation was derived which is able to reproduce the formation and early evolution of regular surface patterns [21]. However, at longer times nonlinear terms have to be taken into account, leading to nonlinear models based on the Kuramoto–Sivashinsky (KS) equation [23]. In particular, the long time evolution of the surface patterns could be successfully described by the so-called damped Kuramoto–Sivashinsky (dKS) equation [24]

\[
\frac{\partial h}{\partial t} = \gamma \frac{\partial h}{\partial x} - \alpha h + v_x \frac{\partial^2 h}{\partial x^2} + v_y \frac{\partial^2 h}{\partial y^2} + \xi_x \left( \frac{\partial h}{\partial x} \right)^2 + \xi_y \left( \frac{\partial h}{\partial y} \right)^2 - D \nabla^4 h,
\]

where \( h(x, y, t) \) is the surface height as a function of the lateral coordinates \( x \) and \( y \) and time \( t \), \( \gamma \) is the lateral velocity of the ripples, \( \alpha \) the damping coefficient and \( v_{x,y} \) are the negative surface tensions in the \( x \)- and \( y \)-directions, respectively. In this framework, the projection of the ion-beam points in the positive \( x \)-direction. \( \xi_{x,y} \) are the nonlinear coefficients and \( D \) is the coefficient of the effective (thermal and ion-induced) diffusion, which, for simplicity, is assumed to be isotropic.

Numerical integrations of equation (1) were performed by using a difference scheme on a grid of 500 \( \times \) 500 lateral nodes with \( \Delta x = \Delta y = 2 \) nm and \( \Delta t \) corresponding to \( 1 \times 10^{14} \) cm\(^2\). The parameters \( v_x \) and \( D \) were calculated following the approach of Bobek et al [25] by using the relations [21] for the initial ripple wavelength \( \lambda_c = 2\pi(2D/|v_x|)^{1/2} \) and the growth rate of the ripple amplitude \( R_k = v_x^2/4D - \alpha \). From the experimentally observed patterns at 500 eV, \( \lambda_c \) and \( R_k \) were determined to be 22 nm and 1.37 \( \times \) 10\(^{17} \) cm\(^2\), respectively, yielding \( v_x = -5.32 \) and \( D = 32.6 \). The other parameters used were \( \gamma = 5 \), \( v_y = -1 \), \( \xi_x = 6 \), \( \xi_y = 0.001 \) and \( \alpha = 0.08 \).

Here, \( v_y < 0 \) and \( \xi_x, \xi_y > 0 \) have been assumed and the value of the additional damping has been chosen to yield the best agreement with the temporal evolution of the experimental pattern.

Figures 1(d)–(f) show the simulated surface morphology at three different times. At the initial stage (figure 1(d)), the ripple structure is less pronounced than for the experiment (figure 1(a); note the widely different height scales). This can be attributed to the roughness of the computer generated starting surface which was found to influence the ripple amplitude at low fluences and was significantly lower than the experimental one in this simulation run.

With increasing fluence, the observed ripples become longer and more pronounced until they form a fully developed ripple pattern. Then, larger corrugations appear and superpose the ripple pattern (figure 1(e)). These corrugations grow with time until the surface reaches a steady state with reduced homogeneity of the ripples (figure 1(f)).

Although both the KS and the dKS equations are known to exhibit no coarsening of the ripple wavelength [26, 27], the qualitative evolution of the simulated morphologies shown in figure 1 agrees remarkably well with the experimental one. Moreover, we consider the dynamics of pattern defects and the coarsening of the ripples as decoupled phenomena. The mobility of the pattern defects is restricted to the direction along the ripples, i.e. there is no hopping of defects from one ripple to a neighboring one. On the contrary, wavelength coarsening occurs in the direction across the ripples. Therefore, the dynamics of the pattern defects should not depend on the ripple wavelength. This assumption is supported by our experimental observations that did not provide any indication for a relation between the coarsening of the ripples and the
The evolution of pattern defects. In addition, since the defect density of a given image is normalized to the ripple wavelength and the scan size, the temporal evolution of $N_D$ is completely decoupled from the time dependence of the wavelength.

The normalized defect density of the simulated patterns was determined as described above for the AFM images. The results are shown in figure 3 (full triangles). The agreement with the experimental defect densities is astonishingly good. Again, an initial decrease of the defect density is followed by an increase and a saturation at intermediate and high fluences, respectively. Quantitatively, however, the defect density is generally higher in the simulations. This discrepancy is probably caused by the estimated $\nu_y$ and $\zeta_{x,y}$ values. In our simulations, we observed that $N_D$ depends significantly on the ratios of the linear and the nonlinear coefficients in the $x$- and $y$-directions, respectively. In particular, the nonlinear anisotropy was found to strongly influence the pattern quality and might even cause a complete decay of the ripples into dot-like features at high fluences. Since the ratios of these coefficients depend on the microscopic parameters of the collision cascade, different absolute values of the defect density can be expected for different experimental conditions like different energies (cf figure 3). However, these dependencies can be rather complex. Therefore, a quantitative comparison with experimental defect densities also demands a precise determination of the $\nu_y$ and $\zeta_{x,y}$ parameters. Nevertheless, the good qualitative agreement with the experiments shows that the applied model is suitable for describing the dynamics of pattern defects.

In order to analyze the dynamics of single pattern defects, a simulated surface region was followed during its evolution. In the very beginning, separated ‘islands’ of short ripples form simultaneously on the surface (cf figure 1(d)). The ripples get longer with increasing time until they touch the borders of the neighboring islands where the ripples of the different islands start to coalesce. This can be clearly seen in figure 4(a). Within these coalescence regions, an enrichment of pattern defects is found which are induced by stacking faults, analog to the situation in crystal growth. Two types of defects can be identified in these regions as indicated in figure 4(a): interstitials (I) and bifurcations (B). The resulting morphology of the surface consists of domains (the former islands) of well-ordered ripples separated by defect rich boundaries. Therefore, these defect rich boundaries seem to be the origin of the observed corrugations. This assumption is also supported by AFM observations. In the AFM image shown in figure 4(b), one can clearly see that the corrugations are separated by defect rich regions. Again, bifurcations (B) and interstitials (I) can be observed within these regions.

With increasing time, the normalized defect density is found to decrease. This implies that pattern defects vanish during the evolution of the pattern. Figures 4(c) and (d) show one simulated surface region at two different fluences. Two mechanisms of defect ‘annealing’ can be identified. First, ripples that end in a defect pinch off. This can be seen at the defect D1, where a ripple ends in a bifurcation. At $\Phi = 6 \times 10^{17}$ cm$^{-2}$ (figure 4(c)), the ripple is already constricted and about to pinch off. This leads to two pinched off ripples and a short interstitial (figure 4(d)). These interstitials and also the pinched off ripples shrink with time. This is evident from the defect D2. At $\Phi = 6 \times 10^{17}$ cm$^{-2}$, D2 consists of a double-bifurcation with two equivalent branches. The upper branch, however, pinches off and shrinks until it has completely vanished at $\Phi = 8 \times 10^{17}$ cm$^{-2}$. The resulting hole then gets closed by a rearrangement of the surrounding ripples. As a second process, pinched off ripples can coalesce again. The defect D3 in figure 4(c) consists of three pinched off ripples, a former bifurcation. In figure 4(d), however, one finds that two of the pinched off ripples have coalesced to one single ripple and only one pinched off ripple remains.

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Although these repair mechanisms are quite efficient, they provide only a partial annealing of the pattern defects. In defect-rich regions, the remaining defects may segregate and form ‘defect clusters’ as can be seen in figure 1(e). These defect clusters are unstable and grow with fluence. Figure 5(a) shows a defect cluster consisting of several pinched off ripples at $\Phi = 3.4 \times 10^{18}$ cm$^{-2}$. With increasing time, the pinched off ripples shrink, leading to the formation of rather large holes (see figure 5(b)). The few ripples in-between rearrange in order to close these holes, but this only causes them to constrict and also pinch off as can be seen in figure 5(c). Figure 5(d) shows the same cluster at a later stage ($\Phi = 1 \times 10^{19}$ cm$^{-2}$). Within the cluster, one can observe now a kind of chaotic bubbling with short new ripples forming, coalescing, pinching off and vanishing again.

Based on these observations, the unexpected evolution of the defect density with its intermediate minimum can be explained in the following way. When the pattern forms at low fluences, it exhibits a rather large number of defects. The identified repair mechanisms anneal defects and reduce the defect density with increasing fluence. Since annealing is achieved solely by annihilation of two neighboring defects, i.e. coalescence of pinched off ripples or vanishing of interstitials, the annealing rate decreases with decreasing defect density. In regions with an unusually high defect density like the coalescence regions of the former ripple islands, defects might segregate and form rather large defect clusters. Because of the further shrinking of interstitials and pinched off ripples, large holes are created that cannot be closed by coalescence or a rearrangement of the surrounding ripples. The defect clusters induce a secondary instability.
which produces new defects due to a pinching off of neighboring ripples and the clusters grow in time. Hence, the observed minimum of the defect density results from the growth of defect clusters which at a certain fluence overcomes the fluence-dependent process of defect annealing.

4. Conclusion

In summary, we have shown that the quality of nanoscale ripple patterns created by low-energy ion sputtering strongly depends on the applied ion fluence. An intermediate regime with minimum defect density was found for sputtering at 300 and 500 eV at a fluence of $\Phi \sim 2 \times 10^{18} \text{ cm}^{-2}$. Simulations of the dKS equation yield good qualitative agreement with the experiments. From the simulations, it can be seen that this minimum of the defect density mainly results from pinch off processes which anneal single defects and cause the growth of larger defect clusters.

Even though the present experiments and simulations represent only certain experimental conditions, we have observed the same qualitative behavior in various simulations with different sets of parameters. Therefore, we propose that the observed evolution of the pattern defects and the intermediate region of highest pattern quality are inherent features of the pattern formation process. However, further investigations are necessary to determine the dependence of the high-quality regime on the experimental conditions. This result offers the possibility to tailor the degree of order (or disorder) in the pattern by simply adjusting the irradiation time. With the pattern quality as an additional parameter, one can now start to explore ordering and anisotropy.
effects in thin films deposited on rippled substrates in more detail. Especially for magnetic thin films, a strong influence of the degree of the morphological anisotropy of the substrate on the magnetic properties of the film can be expected [6].

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