CHAPTER 4

4. Evolution of morphology on Si surface under low energy Ar\(^+\)-ion irradiation

4.1 Introduction

It is well-known that ion-beam sputtering (IBS) leads to formation of spontaneously arising patterns of corrugations, holes, and dots [1-2] at the surfaces of different materials viz. metals, semiconductors, and insulators [3-8]. In fact, patterned substrates, mostly in the form of ripples, are being used as templates to grow and tune magnetic and plasmonic properties of thin films by using their morphological anisotropy [9-12]. However, in spite of its potential applications, complete control over tailoring IBS induced self-organized patterns with desired properties is yet to be achieved. One of the major reasons behind this has been the lack of complete understanding of the mechanisms that govern the pattern formation. This is partly due to the huge time-scale separation among different processes that influence the system, which complicates the unambiguous identification of the main underlying physical mechanisms.

Ripple formation is known to result from off-normal ion bombardment of materials. In the linear stability analysis (as mentioned in chapter 3) of Bradley and Harper (BH) theory, patterns originate from destabilizing (roughening) effect in which the erosion rate is enhanced at regions of high concave curvature and the stabilizing (smoothening) effect of surface diffusion [13]. This theory explains many experimental observations (like ripple formation, ripple rotation, growth of ripple amplitude etc.) although some significant contradictions with experimental results also exists. For instance, one of the important predictions of BH theory is that a flat surface is always unstable under ion erosion. In contrast, experimental studies on Si and SiO\(_2\) [14-17] show the existence of flat surfaces, up to incident angles of 50\(^\circ\) or 40\(^\circ\), respectively,
from where ripple formation starts. Such a sharp boundary between a flat (stable) and a rippled (unstable) silicon surface was identified by Madi et al. as bifurcation in control parameter (incident angle, $\theta$ and ion energy, $E$) space [18]. Later on, Madi et al. [18] concluded that stability at low angles and topographical instability at high angles can be explained by taking into account the effects of ion impact-induced prompt atomic redistribution [calculated from Carter-Vishnyakov (CV) theory] [14] and that the erosive component (calculated from BH theory) is essentially irrelevant.

As mentioned in chapter 3, Castro et al. [19] put forward an alternative approach of ion induced solid flow model to explain formation of ripple patterns beyond a threshold angle. This theory is based on the continuum description of the surface flow that is driven by surface confined stress-induced viscous flow of a thin amorphous layer that forms during ion bombardment. It has been pointed out by the same authors that both the mass redistribution and the viscous flow descriptions agree with the experiments with respect to the transition from flat to rippled surface at a particular incidence angle and the dependence of the pattern wavelength on incident angle of ions [20]. However, molecular dynamics (MD) based calculations cannot probe the required timescales associated with pattern evolution whereas the solid flow model offers the advantage of time-dependent (dynamic) evolution of ripple patterns [16,21,22].

The solid flow model is based on the assumption that due to the impact of the ions and the subsequent release of energy within the target, defects are created inside the material. These events occur within a few picoseconds after the impact. Relaxation of some amount of defects leads to sputtering of target atoms which is associated with the generation of a residual stress that is confined within a thin amorphous layer that builds up beneath the surface [23] and reaches a stationary value. This ion-induced (compressive) stress is manifested as a highly viscous flow of the incompressible amorphous layer. Using a hydrodynamic description of the solid flow with a linear approximation in perturbations around a flat target profile, Castro and
Cuerno [24] obtained a real part for the linear dispersion relation (i.e. amplification rate of wave-vector, \( \vec{q} \)) given by:

\[
\omega_q = -\left[f_E d^3 \phi(\theta) q^2 + \sigma d^3 q^4 \right]/3\mu
\]  

(4.1)

where \( \phi(\theta) = \frac{\partial}{\partial \theta} \left( \Psi(\theta) \sin \theta \right) \), \( d \) is the average thickness of the amorphous layer, \( \mu \) is (ion-induced) viscosity, and \( \sigma \) is the interface surface tension. The parameter \( f_E \) can be understood as the gradient of residual stress induced by ions across the amorphous layer, whose angular dependence is described through the function \( \Psi(\theta) \). Since this angular function needs to be prescribed, we take \( \Psi(\theta) = \cos \theta \) as the simplest geometrically motivated choice that shows a good agreement [24]. According to the solid flow model [19], the smallest timescale associated with ripple formation in the linear regime can be written as:

\[
\tau(\theta, E) \sim \frac{\mu}{f_E^2 d^3 \phi^2(\theta)}
\]  

(4.2)

The most general prediction that can be obtained from this theory is that it allows finding the characteristic scale for the exponential growth of the pattern amplitude occurring at short times where linear approximation holds. For a given pair of angle and energy reference values, \((\theta_{\text{ref}}, E_{\text{ref}})\), one can extrapolate the value of the intrinsic timescale, \( \tau \), for any other pair \((\theta, E)\) through the relation:

\[
\tau(\theta, E) = \tau(\theta_{\text{ref}}, E_{\text{ref}}) \frac{J_{\text{exp}}(\theta_{\text{ref}}, E_{\text{ref}}) E_{\text{ref}}^{-7/3+2m} \phi^2(\theta_{\text{ref}})}{J(\theta, E) E^{-7/3+2m} \phi^2(\theta)}
\]  

(4.3)

where \( J_{\text{exp}}(\theta_{\text{ref}}, E_{\text{ref}}) = J_{\text{exp}}(E) \cos \theta \) is the flux used in a particular experiment for energy \( E \) and angle \( \theta \), with \( J_{\text{exp}}(E) \) being the flux at normal incidence and \( m \) can be assumed to be in the range \( 1/3-1/2 \) [19].
In case of solid flow model, based on detailed binary-collision simulations, it is assumed that the generated stress depends on the ion energy (<2 keV) [25,26]. However, there is no direct measurement which quantifies ion induced stress. Thus, it is a good strategy if the parameter, $f_E$, can be avoided while calculating the intrinsic timescale at any angle. In order to do so, we experimentally determined the intrinsic timescale (for a reference ion incidence angle) up to which the growth of roughness follows an exponential behaviour and the ripple wavelengths remained constant. This intrinsic timescale was utilized to verify the validity of solid flow model at other ion incidence angles.

Using this model, we identified the hitherto unexplored angular window in which the solid flow model may be invoked to explain 500 eV Ar-ion induced ripple formation. Since, in general, it is observed that the ion incidence angle (in our case 51°) at which transition from flat to rippled surface takes place remains constant at different energies (in the range of 0.25-1.5 keV) [18,20,22], we have chosen an Ar-ion energy of 0.5 keV as a representative one and tested the applicability of the solid flow model on silicon over an angular window of 51° to 72.5° – wherever parallel-mode ripple patterns evolve. We found out the experimental timescale at an arbitrary angle of incidence – the reference angle (60°) – up to which the system evolved linearly. This time was used to predict the intrinsic timescale (by invoking the solid flow model) for other ion incidence angles and subsequently systematic experiments were carried out to check their proximities at each of those angles. These angles included the transition point of flat to rippled surface and beyond which parallel-mode ripples disappear. We also show that the temporal evolution of ripples at all these angles remains in the linear regime although the nonlinear regime sets in quickly at higher incidence angles.

Identification of the linear regime of pattern formation can be precisely done only with the help of the solid flow model. However, it may be mentioned that on the experimentally front, the major drawbacks have been the lack of adequate experimental data, in particular, at higher
oblique angles of incidence, and the reproducibility of observed patterns. These give rise to the
necessity of performing systematic experiments over a large angular window to develop a
better understanding and to learn how to control and manipulate the surface patterns by ion
irradiation. As mentioned above, usually, parallel-mode ripples (i.e. wave-vector parallel to the
projected ion-beam) are formed and reported beyond an incident angle ($\theta$) of 50° [27,28]. If
one considers the full angular window (i.e. $\theta$= 0° to 85°), reports are available on ripple
formation up to an oblique incidence of ~70° [27,29,30]. Thus, there is hardly any systematic
study on Ar-ion induced pattern formation on Si in the angular window $70^\circ \leq \theta \leq 80^\circ$ which
would have helped unfolding the underlying processes leading to a transition from parallel- to
perpendicular-mode ripples. In the studies presented in the thesis, evolution of surface
topography on Si(100) substrates is investigated under low energy ion bombardment over an
angular window of 63°-83°. This gives rise to evolution of a gamut of unusual patterns which
were not reported earlier (those formed at higher oblique incident angles, viz. 70°-78°).

Through these intermediate patterns parallel-mode ripples are seen to undergo a transition to
perpendicular-mode ripples at an incident angle of 80°. In case of an even higher angle of
incidence surface smoothening is observed. At this point the origin of these unusual structures
was not clear. We have made an attempt to correlate the roughness evolution plot to the
sputtering yield plots as a function of ion incidence angle. However, this does not solve the
problem of explaining unusual patterns observed in the angular window of 70°-72.5°. To
understand the pattern formation mechanism at these two angles, fluence-dependent studies
were carried out at $\theta$=70° and 72.5° by keeping all other parameters same. In doing so, it was
observed that with increasing fluence parallel-mode ripples gets transformed into protruded
mounds/faceted structures. In addressing the mechanism of the observed transition, variation
in the erosion rate of a nonplanar surface is calculated by using the theoretical model of Carter
[31]. It is seen that beyond threshold values of the amplitude to wavelength ratio, inter-peak
shadowing of incident ion-flux can lead to a transition from ripples to faceted structures. The coarsening behaviour of faceted structures with increasing fluence is explained in light of Hauffe’s mechanism based on reflection of primary ions [32]. In addition, Carter’s theory was also employed to calculate fractional change in sputtering yield for ripples and faceted structures (where we have replaced the amplitude to wavelength ratio by amplitude to base-width ratio for the latter ones) to correlate the surface roughness to the sputtering yield.

It may be pointed out that ripples are being utilized as a template for depositing thin films to modify their functional properties [11,33,34], albeit ripple formation is unwanted in various techniques, viz. secondary ion mass spectroscopy (SIMS), Auger electron spectroscopy (AES), and ion milling. The ion incidence angle $\theta \neq 0^\circ$ for a typical SIMS or AES set up results in formation of ripples at the surfaces. This results in rapid degradation in depth resolution. As a way out to this problem Zalar first demonstrated that by rotating the sample with respect to its surface normal during sputtering one can avoid ripple formation [35]. It may be noted that sample rotation does not always become fruitful in suppressing surface roughening. Sometimes sample rotation during ion erosion roughens the surface but at a slower rate [36,37]. Smoothening of the surface under sample rotation is explained by Bradley and Cirlin [38]. According to Bradley’s theory, during sample rotation, smoothing effect due to bulk viscous flow and surface self-diffusion can be predominant over the roughening effect of the curvature-dependent sputter yield and generate a smooth surface [38]. Carter extended this approach to examine the evolution of surface roughness with continuous irradiation under concurrent substrate rotation [39]. In this frame work, the surface height evolution is described by taking into account curvature-dependent roughening [13] and different surface relaxation processes namely, viscous relaxation [40], sputtering relaxation [41], ballistic collisional drift, and diffusion relaxation [42] along with statistical fluctuations. For a semiconducting material, ion irradiation causes defects in the amorphous layer (created during early stage of sputtering)
which subsequently relaxes and leads to a viscous flow. In addition, ion impact generates internal fluxes of moving recoils that transports both parallel and normal with respect to the surface. This movement has a drift component and diffusion-like-component. Furthermore, thermally activated diffusion is also important when experiments are performed at higher temperatures. However, sample can still get roughened during concurrent ion sputtering and sample rotation if the viscosity of the sample is too high. Bradley explained this behaviour by taking into account the sign of a parameter characterizing the curvature dependence of the sputtering yield [41].

In the last part of this chapter, we demonstrate the effect of sample rotation during ion-beam irradiation towards tuning the surface roughness. Si(100) substrates were exposed to argon ions at different incident angles with respect to the surface normal. It is observed that under substrate rotation, flat surface remains flat whereas mound-like structures formed at oblique incidence angles (where ripple morphology prevails without substrate rotation). In addition, a detailed quantitative atomic force microscopy (AFM) measurements are presented for different sputtering times at two incident angles viz. (67° and 60°) under sample rotation with a constant angular speed ($\omega$) during ion-beam erosion to address the temporal evolution of mounds under substrate rotation. Moreover, to check the effect of angular speed variation, the substrates were rotated at different angular velocities for a particular fluence. The results are explained in light of existing models based on ion-matter interaction.

4.2 Experimental

The substrates used in the experiments were sliced into small pieces from a $p$-type Si(100) wafer (B-doped, resistivity 0.01-0.02 $\Omega$ cm). The UHV-compatible experimental chamber was used which is equipped with a 5-axes sample manipulator and an electron cyclotron resonance (ECR) based broad beam, filament-less ion source (more details are available in chapter 2).
The chamber base pressure was below $5 \times 10^{-9}$ mbar and the working pressure was maintained at $2.5 \times 10^{-4}$ mbar by using a differential pumping unit. Silicon samples were fixed on a sample holder which was covered with a sacrificial silicon wafer of the same lot to ensure a low-impurity environment. The beam diameter and the ion flux were measured to be 3 cm and $1.3 \times 10^{14}$ ions cm$^{-2}$ s$^{-1}$, respectively. The experiments to identify the linear regime of pattern formation, were performed at this fixed flux and exposure times in the range of 1 to 120 min for four incidence angles, viz. $51^\circ$, $60^\circ$, $65^\circ$, and $72.5^\circ$. In addition, Argon-ion energy was chosen to be 500 eV since it lacks a systematic study at this energy for higher oblique incident angles under consideration. The current density was measured to be $21 \mu$A cm$^{-2}$ (except for $65^\circ$ where the current density was fixed at $28 \mu$A cm$^{-2}$). The experiments were performed at a fixed fluence of $5 \times 10^{17}$ ions cm$^{-2}$ for the entire angular window of $63^\circ$-$83^\circ$. In addition, measurements were carried out to study flux- and fluence-dependent evolution of surface morphology in case of selective angles.

To observe the ripple to facet transition, experiments were performed for fluences in the range of $1-20 \times 10^{17}$ ions cm$^{-2}$ and at two different ion incidence angles, namely $70^\circ$ and $72.5^\circ$ (with respect to the surface normal). According to TRIDYN simulation [43], although the sputtering yield maxima is close to $70^\circ$, for the sake of completion, we also performed measurements at $72.5^\circ$ which is not far off from the sputtering yield maxima and at this higher angle the shadowing effect is expected to be more prominent.

Following Ar-ion exposure the samples were imaged by *ex-situ* atomic force microscopy (described in chapter 2). Root mean square (rms) surface roughness, $w$, and two-dimensional (2D) autocorrelation function were calculated for all AFM images by using the WSxM software [44]. Wavelength of ripple patterns was calculated from the respective autocorrelation functions. For faceted structures, instead of wavelength, we considered the average base-width value which were calculated from a large number of line profiles drawn on the respective AFM
images. In addition, x-ray photoelectron spectroscopic measurements were performed on Ar-ion bombarded Si samples which did not show the presence of any impurity above their respective detection limits.

For addressing the effect of concurrent substrate rotation during ion-beam irradiation, 500 eV Argon-ion energy was chosen and due to the lack of any systematic study at this energy for higher oblique incident angles under substrate rotation during ion-beam irradiation. A thorough angle-dependent study was performed with a substrate rotational speed of 0.5° s\(^{-1}\) at 67°. Most of the experiments were performed for different angles of incidence by properly choosing the rotational speed and the time of erosion so that the system evolves in the linear regime. For 67°, six different angular speeds (in the range of 8×10\(^{-3}\) - 44×10\(^{-3}\) rad/s) were chosen to realize the effect of rotational speed variation. Moreover, experiments were carried out to study time evolution of patterns for the incident angles of 67° and 60°.

### 4.3 Results and discussion

#### 4.3.1 Temporal evolution of ripple patterns

Figure 4.1 presents a high-resolution cross-sectional TEM image of a silicon sample which was exposed to argon ions at 60° for the duration of 11 min. The presence of ion-induced surface corrugation in the form of ripples (wavelength, \(\lambda\) ~ 30 nm) is clearly seen from Fig. 4.1. In addition, a thin amorphous silicon layer at the top is also evident which forms during ion bombardment and subsequently acts as a highly viscous and incompressible medium. This meets one of the pre-requisites for invoking solid flow model in the present case. To further justify the use of solid flow model, we performed systematic experiments at 60° which was chosen to be the reference angle.

Figure 4.2 shows the roughness evolution plot for silicon following argon ion bombardment (up to 11 min). The insets show selective AFM images for three different exposure times, viz.
1, 5, and 11 min. The exponential fit ensures that the temporal evolution of ripples remains in the linear regime up to 11 min.

Figure 4.1: Cross-sectional TEM image corresponding to ion incidence angle of 60° where the amorphous layer is marked by the red line. The inset shows slightly zoomed out version of the same sample.

Figure 4.2: Roughness evolution plot corresponding to incidence angle of 60°. The inset shows the AFM topographic images corresponding to exposure times of 1, 5.5, and 11 min, respectively.
Figure 4.3: AFM micrographs of $p$-Si(100): Exposed at an incidence angle of 51º for different times starting from 5 to 66 min. The arrow indicates the direction of the incoming ion beam.

Figure 4.4: Variation in roughness with exposure time for the incidence angle of 51º where the exponential fit is well realized. The inset shows the plot for wavelength versus exposure time at 51º.
However, beyond 11 min of erosion time (i.e. the intrinsic timescale for 60°) the rms roughness does not follow the exponential growth behaviour any more (further discussed in the later part of this section). This is in accordance with the solid flow model as described above (Chapter 3) and thus, we identify this threshold time (11 min) as the reference intrinsic timescale (τ) to test the applicability of solid flow model at other incidence angles, viz. 51° (the transition point from flat to parallel-mode rippled surface), 65°, and 72.5° (beyond which the parallel-mode ripples disappear).

In the present study, we performed our experiments at a constant flux and energy which simplify Eq. 4.3 further. This leads to the estimated intrinsic timescale, \( \tau(\theta,E) = 64 \text{ min} \), at \( \theta=51° \). A systematic time evolution study (starting from 1 to 66 min of exposure to argon ion beam) was carried out at this angle and the AFM images are presented in Figs. 4.3(a)-(e). These micrographs involve very small roughness values and hence, lack the typically coherent look of ripple patterns observed at other angles.

Corresponding roughness evolution plot is shown in Fig. 4.4 where a clear exponential growth of the rms roughness is observed up to an irradiation time of 66 min which is very close to the time predicted by the solid flow model (64 min). Beyond this time, an increase in the roughness takes place although the exponential behaviour is not followed any more. The validity of the linear regime is further confirmed from the wavelength versus exposure time plot where the wavelengths remain constant (average value of 43 nm) up to an exposure time of 66 min (inset, Fig. 4.4).

To verify the applicability of this theory far from the stable to rippled surface transitional point, we chose 65° as one of our test angles. For this angle, the intrinsic timescale calculated by using Eq. 4.2 turned out to be 5 min.

Accordingly, early stage evolution of ripple pattern was studied and the results are presented in Fig. 4.5 which shows time-dependent roughness evolution.
The insets in this figure show morphological evolution in the form of selective AFM images. The roughness evolution data get fitted well by an exponential curve up to 4.5 min which can be identified as the point of transition from the linear to the non-linear regime. In addition, the ripple wavelengths remain constant ($\lambda \sim 23$ nm) up to this time. This clarifies the relevance of the solid flow model at an incidence angle greater than the reference angle ($60^\circ$).

Figure 4.5: Temporal evolution of surface roughness at $\theta=65^\circ$. Images corresponding to 2.2, 3.3, and 4.4 min of exposure times are shown as insets for a realization of the corresponding surface morphology.

Likewise, an exponential growth in roughness is also observed for ripples, formed by irradiation at an incidence angle of $72.5^\circ$, till 2.5 min which is little earlier than the intrinsic time scale predicted by the solid flow model (3.8 min). A comparison between the predicted ($\tau_{\text{pre}}$) and the experimentally measured ($\tau_{\text{ob}}$) timescales for transition from the linear (ripple wavelength remains constant) to the nonlinear (ripple coarsening) regime is depicted in Table 4.1.
It is clearly seen that with increasing ion incidence angle, the said transition takes place at a shorter timescale. In addition, $\tau_{ob}$ deviates from $\tau_{pre}$ corresponding to some incidence angles which is not well understood at this point. It may be mentioned that evolution of parallel-mode ripples, under the present experimental conditions and at higher energies do not occur beyond this angle and hence the solid flow model could be successfully tested for $51^{\circ} \leq \theta \leq 72.5^{\circ}$.

**Table 4.1**

Predicted ($\tau_{pre}$) and experimentally measured ($\tau_{ob}$) timescales for transition from the linear to the non-linear regime at different ion incidence angles.

<table>
<thead>
<tr>
<th>Angle of incidence (°)</th>
<th>$\tau_{ob}$ (min)</th>
<th>$\tau_{pre}$ (min)</th>
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<tbody>
<tr>
<td>51</td>
<td>66</td>
<td>64</td>
</tr>
<tr>
<td>60</td>
<td>11</td>
<td>11</td>
</tr>
<tr>
<td>65</td>
<td>4.5</td>
<td>5.0</td>
</tr>
<tr>
<td>72.5</td>
<td>2.5</td>
<td>3.8</td>
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In order to address the temporal evolution of roughness beyond the linear regime, experiments were carried out for long durations over the entire angular window (i.e. $\theta = 51^{\circ}$ to $72.5^{\circ}$) where ripple formation takes place and the results are summarized in Fig. 4.6. These plots clearly demonstrate that the exponential growth behaviour of roughness does not hold true beyond the intrinsic timescales (as discussed above) where ripple formation remains in the linear regime. According to Castro *et al.* [19], nonlinear effects can originate from ion induced stress [24] or from purely erosive effects [45] due to prolonged sputtering although they are not included in the scope of the solid flow model. However, a more generalized nonlinear theory — lubrication theory — which is beyond the scope of the present study, has already been invoked which can access the nonlinear regime of ripple evolution [46].
4.3.2 Role of ion incidence angle on pattern formation

Figures 4.7(a-h) present the AFM topographic images of silicon surface before and after exposure to argon ions at different oblique incident angles. Fig. 4.7(a) depicts AFM image of the pristine sample which has a smooth surface ($w=0.09$ nm). From Fig. 4.7(b) it is observed that ripple morphology evolves at 65°. Estimations using WSxM software show that ripples have wavelength of 38 nm, height of 3 nm, and rms surface roughness of 1.7 nm. Careful measurements reveal that the ripple wavelength is independent of ion flux. Fig. 4.7(c) represents the morphology corresponding to an incident angle of 67° where parallel-mode ripples (wavelength of 60 nm, height of 3 nm, and rms roughness of 2.6 nm) are formed. In
addition, mounds (average dimension of 65 nm) appear on top of the ripples. Formation of such parallel-mode ripples is consistent with literature [23, 29].

Starting from this point (i.e. at 67°), where mounds on ripples are observed, unusual patterns evolve at higher incident angles which were hardly seen earlier. Fig. 4.7(d) presents the AFM image corresponding to an oblique incidence of 70° where mounds are observed instead of parallel-mode ripples. These mounds have an average dimension of 88 nm and the corresponding surface roughness is 5.7 nm. There is a mild signature that these mounds may be protruded in the direction of ion-beam projection on the sample surface. For the incident angle of 72.5°, cone-like structures are observed where their apexes point towards the direction of ion beam [Fig. 4.7(e)]. The calculated surface roughness, mean value of the apex angle, and the base dimension are 7.8 nm, 34.8°, and 110 nm, respectively.

Figure 4.7: AFM images of Si(100): (a) Pristine and exposed to 500 eV Ar⁺-ions to the fluence of 5×10¹⁷ ions cm⁻² at incident angles of (b) 65°, (c) 67°, (d) 70°, (e) 72.5°, (f) 77.5°, (g) 80°, and (h) 82.5°. Corresponding height scales in (a)-(h) are 1 nm, 15 nm, 23 nm, 37 nm, 51 nm, 19 nm, 3 nm, and 2 nm, respectively. Arrows indicate projection of the ion beam on the surface. Insets show the 2D FFT obtained from the corresponding images.
Upon increasing the angle of incidence to 77.5° elongated structures are formed which resemble more like nano-needles. These needle-like structures (having an average length of 200 nm) lie parallel to the projection of ion beam onto the surface [Fig. 4.7(f)].

Further, increasing the ion incidence angle to 80°, perpendicular-mode ripples (of wavelength 65 nm and rms surface roughness of 0.4 nm) are formed [Fig. 4.7(g)]. Thus, it is clear that parallel-mode ripples undergo a transition to perpendicular-mode ripples at this angle. However, the transition from parallel- to perpendicular-mode ripples is not sharp since it undergoes a series of unusual pattern formation at intermediate angles. At 82.5°, a smooth surface is observed [Fig. 4.7(h)] whose roughness is comparable to the pristine surface. Formation of such ultra-smooth surfaces under nearly similar experimental conditions was reported earlier even at 85° [28]. The insets corresponding to AFM images shown in Figs. 4.7(b)-(h) represent the respective 2D FFT. For instance, the FFT corresponding to the incident angle of 65° depicts the obvious presence of a characteristic wavelength, which gets weakened in case of incident angle of 67°. On the other hand, starting from 70° to 80°, FFTs show signature of topographical anisotropy.

From the above discussion it is observed that due to 500 eV Ar-ion bombardment many different patterns evolve over the entire angular window being considered under the present study. However, for the angular window of 0°-50°, surface remains stable under ion bombardment whereas parallel-mode ripples emerge at 51° incidence angle (demonstrated in Fig. 4.8). It may be mentioned that formation of ion-induced self-organized nanostructures is generally caused due to the interplay between roughening caused due to sputter erosion and smoothening due to surface diffusion [13,47]. However, no single theory is adequate to explain all kinds of observed experimental features. This fortifies the need to invoke possible multiple physical effects to explain our observed patterns at different incident angles.
Evolution of morphology on Si surface under low energy Ar-ion irradiation

Let us first examine the role of sputtering on the origin of pattern formation observed in our case. In order to do this, an attempt has been made to correlate the evolution of surface roughness with sputtering. Fig. 4.9(a) shows the evolution of surface roughness [obtained from AFM images shown in Figs. 4.7(b)-(h)] at different incident angles. We also performed TRIDYN simulation [43] to calculate sputtering yield for 500 eV Ar-ions (for the fluence of $5 \times 10^{17}$ ions cm$^{-2}$) corresponding to different angles. Figure 4.9(b) shows the variation in sputtering yield with incident angle. From these figures it is seen that the roughness value peaks at 72.5° whereas the sputtering yield is maximum at ~70°. Thus, it is expected that sputtering will play a role for unusual pattern formation in our case.

The evolution of mounds/faceted structures at 70° and 72.5° angles of incidence will be discussed in the next section. It may be mentioned that the elongated needle-like structures
seen at $77.5^\circ$ can be attributed to shadowing effect which is expected to cause an increase in erosion of surface protrusions compared to depressions [28,47].

![Figure 4.9: (a) Evolution of rms surface roughness corresponding to different angle of incidence. (b) Plot of sputtering yield versus angle of incidence obtained from TRIDYN simulation performed for 500 eV Ar$^+$-ions to the fluence of $5\times10^{17}$ ions cm$^{-2}$.](image)

The same mechanism should be operative at $80^\circ$ where the surface evolves with a relatively low rms roughness. Such a low surface roughness may be attributed to a reduced sputtering yield at $80^\circ$ [Fig. 4.9(b)]. For an even higher glancing angle of incidence (viz. $82.5^\circ$), the observed smoothening of the surface can be explained in terms of extremely low sputtering yield where most of the ions would get reflected rather than contributing to sputtering [28].

### 4.3.3 Role of shadowing: Ripple to facet transition

Figures 4.10(a)–(g) present AFM topographic images obtained from silicon samples before and after exposure to argon-ion incidence angle $70^\circ$ at different fluences. Fig. 4.10(a) presents AFM image of the pristine sample which shows a smooth surface (rms surface roughness $= 0.09$ nm). Figs. 4.10(b) and (c) show signature of corrugated surfaces formed at low fluences, namely $1\times10^{17}$ and $2\times10^{17}$ ions cm$^{-2}$, respectively. However, small mound-like entities also start appearing on the corrugated surface at the latter fluence.
Figure 4.10: AFM images of (a) pristine-Si and those exposed to 500 eV Ar\textsuperscript{+}-ions at an incidence angle of 70° to various fluences: (b) 1×10\textsuperscript{17} ions cm\textsuperscript{-2}, (c) 2×10\textsuperscript{17} ions cm\textsuperscript{-2}, (d) 5×10\textsuperscript{17} ions cm\textsuperscript{-2}, (e) 10×10\textsuperscript{17} ions cm\textsuperscript{-2}, (f) 15×10\textsuperscript{17} ions cm\textsuperscript{-2}, and (g) 20×10\textsuperscript{17} ions cm\textsuperscript{-2}, respectively. The corresponding height scales for (a)-(g) are: 1 nm, 4.3 nm, 9.9 nm, 39.5 nm, 60.9 nm, 85.7 nm, and 182.2 nm. For better clarity (a)-(c) represent images acquired over a scan area of 1µm×1µm whereas (d)-(g) are of scan area 2µm×2µm. Insets show the 2D autocorrelation functions for corresponding images.

Figs. 4.10(d)-(g) depict AFM images where mound formation becomes predominant (at the fluence of 5×10\textsuperscript{17} ions cm\textsuperscript{-2}) which transform into faceted structures corresponding to the fluence of 10×10\textsuperscript{17} ions cm\textsuperscript{-2} and grow further at even higher fluences.

Figures 4.11(a)-(f) show AFM topographic images corresponding to incidence angle of 72.5° where presence of ripple morphology is clear up to a fluence of 1×10\textsuperscript{17} ions cm\textsuperscript{-2}. Beyond this fluence, ripples disappear and small mounds as well as faceted structures evolve (which grow further with increasing fluence) which is evident from Figs. 4.11(b)-(f).
Figure 4.11: AFM images of silicon exposed to 500 eV Ar$^+$-ions at 72.5° incidence angle at fluences of (a) $1 \times 10^{17}$ ions cm$^{-2}$, (b) $2 \times 10^{17}$ ions cm$^{-2}$, (c) $5 \times 10^{17}$ ions cm$^{-2}$, (d) $1 \times 10^{18}$ ions cm$^{-2}$, (e) $1.5 \times 10^{18}$ ions cm$^{-2}$, and (f) $2 \times 10^{18}$ ions cm$^{-2}$, respectively. The corresponding height scales for (a)-(f) are: 4 nm, 4 nm, 74 nm, 86 nm, 154 nm, and 165 nm. For better clarity (a) & (b) have scan size of 1µm×1µm whereas (c)-(f) have scan size of 2µm×2µm. Insets show the 2D autocorrelation functions for corresponding images.

The insets of all the images shown in Figs. 4.10 and 4.11 represent corresponding 2D autocorrelation functions. In Fig. 4.10, ripple anisotropy is clearly observed at the fluence of $1 \times 10^{17}$ ions cm$^{-2}$ whereas the same in Fig. 4.11 is clear up to the fluence of $2 \times 10^{17}$ ions cm$^{-2}$.

The average values (calculated from the AFM images shown in Figs. 4.10 and 4.11) of ripple wavelength, feature height, and base-width of mounds/facets are listed in Table 4.2 for different fluence values. An increasing trend in height and base-width of mounds/facets is observed for both angles of incidence with increasing Ar-ion fluence albeit the effect is more prominent at 72.5°. The aspect ratio of the feature is one of the crucial parameters in understanding the variation of the feature size as a function of ion fluence. In the present thesis, the variation of average structure height has been depicted as a function of ion fluence. We have calculated the
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The aspect ratio of faceted structures formed at 70° and 72.5° incidence angles. Table 4.2 depicts the aspect ratio of facets at different fluences corresponding to the above two incident angles.

**Table 4.2**

Calculated values of ripple wavelength (λ), average feature height (h), and average base-width of the faceted structures formed under 500 eV Ar⁺-ions bombardment at different fluences at different angles of incidence.

<table>
<thead>
<tr>
<th>Angle of incidence</th>
<th>Fluence (ions cm⁻²)</th>
<th>λ (nm)</th>
<th>Average feature height (nm)</th>
<th>Average base-width (nm)</th>
<th>Aspect ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>70°</td>
<td>1×10¹⁷</td>
<td>34</td>
<td>2</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td></td>
<td>2×10¹⁷</td>
<td>57</td>
<td>5</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td></td>
<td>5×10¹⁷</td>
<td>--</td>
<td>16</td>
<td>131</td>
<td>0.122</td>
</tr>
<tr>
<td></td>
<td>1×10¹⁸</td>
<td>--</td>
<td>22</td>
<td>152</td>
<td>0.145</td>
</tr>
<tr>
<td></td>
<td>1.5×10¹⁸</td>
<td>--</td>
<td>30</td>
<td>199</td>
<td>0.151</td>
</tr>
<tr>
<td></td>
<td>2×10¹⁸</td>
<td>--</td>
<td>56</td>
<td>357</td>
<td>0.157</td>
</tr>
<tr>
<td>72.5°</td>
<td>1×10¹⁷</td>
<td>26</td>
<td>1</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td></td>
<td>2×10¹⁷</td>
<td>27</td>
<td>2</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td></td>
<td>5×10¹⁷</td>
<td>--</td>
<td>28</td>
<td>237</td>
<td>0.118</td>
</tr>
<tr>
<td></td>
<td>1×10¹⁸</td>
<td>--</td>
<td>50</td>
<td>363</td>
<td>0.137</td>
</tr>
<tr>
<td></td>
<td>1.5×10¹⁸</td>
<td>--</td>
<td>78</td>
<td>486</td>
<td>0.166</td>
</tr>
<tr>
<td></td>
<td>2×10¹⁸</td>
<td>--</td>
<td>90</td>
<td>525</td>
<td>0.171</td>
</tr>
</tbody>
</table>

To explain the transition from a rippled surface to faceted structures we invoke the shadowing condition stated in Eq. 3.56. Let us first consider the case of 70° and the fluence of 1×10¹⁷ ions cm⁻² where the calculated value of \(2πh₀/λ\) turns out to be 0.369 whereas tan \((π/2-θ)\) is 0.364. Thus, \(2πh₀/λ\) is slightly above the limiting condition which indicates shadowing effect to start playing a role at this fluence itself. In the case of 2×10¹⁷ ions cm⁻², the shadowing effect becomes more prominent since \(2πh₀/λ\) turns out to be 0.551. As a result, crests of the ripples should undergo more erosion compared to troughs and hence there is a likelihood of

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mounds/facets to evolve. This explains the observation of mounds at this fluence. Similar behaviour is observed in case of 72.5°. For instance, in the case of 1×10^{17} ions cm^{-2}, 2\pi h_0/\lambda equals to 0.242 while \tan(\pi/2 - \theta) turns out to be 0.315. Thus, the condition for no shadowing, i.e. \tan(\pi/2 - \theta)\geq2\pi h_0/\lambda gets satisfied here and ripples are expected to be seen. The observation of sinusoidal ripples in Fig. 4.10(a) supports this theoretical prediction. On the other hand, shadowing sets in at the fluence of 2×10^{17} ions cm^{-2} since in this case \tan(\pi/2 - \theta) becomes smaller than 2\pi h_0/\lambda (=0.465). This leads to formation of small mound-like entities (in the form of broken ripples) appearing on the corrugated surface.

For further investigation on the role of shadowing effect in morphological evolution we extracted line profiles of the observed structures along the direction of incident ion-beam onto the surface as shown by the arrow marks on the respective AFM images. Line profiles obtained from Figs. 4.10(b) & (c) and Figs. 4.11(a) & (b) are shown in Figs. 4.12 and 4.13, respectively. It is observed from Figs. 4.12(b) and 4.13(b) that at the beginning of shadowing transition the line profiles are still sinusoidal in nature. As discussed above, beyond shadowing transition one would expect signature of saw-tooth like waveform. The fact that for both incidence angles saw-tooth like waveform is not yet formed may be attributed to early stage of shadowing where h_0/\lambda ratios are very close to the limiting values or little above. To check this, line profiles obtained from Figs. 4.10(d) & 4.11(c) (corresponding to a higher fluence of 5×10^{17} ions cm^{-2}) are shown in Figs. 4.12(c) &4.13(c) which clearly show a transition to saw-tooth like waveform. This is due to the fact that h_0/\lambda ratios (in both cases) are well beyond the respective shadowing limits (0.767 and 0.741, respectively). Thus, we can infer that the effect of ion-beam shadowing plays a dominant role in the transition from rippled surfaces to faceted structures and is expectedly more prominent for the higher incidence angle as is evident from the above discussion. We now go on to explain the coarsening behaviour of faceted structures (as is evident from Table 1) at higher fluences (>5×10^{17} ions cm^{-2}) by using the mechanism
proposed by Hauffe [32]. In this framework, the intensity of reflected ions impinging on an arbitrary area on a facet depends on the dimensions of the reflecting adjoining facets. According to $V_n \sim jY$ where $j$ is the ion density on the surface element (which also contains the reflected ions), $Y$ is the sputtering yield, and $V_n$ is the displacement velocity of a surface element in the direction of its normal, it is clear that the displacement velocity will be higher for the larger facet.

This does not require a particular form of spatial distribution of reflected ions albeit it is necessary that the reflected ions should fall on the neighbouring facets. Accordingly, a smaller facet will disappear into the next bigger one and form an even bigger facet. This corroborates well with the cross-sectional line profiles corresponding to faceted structures shown in Figs. 4.12 and 4.13.

Figure 4.12: Line profiles extracted from the AFM images of ion exposed samples at 70° for various fluences: (a) $1 \times 10^{17}$, (b) $2 \times 10^{17}$, (c) $5 \times 10^{17}$, (d) $10 \times 10^{17}$, (e) $15 \times 10^{17}$, and (f) $20 \times 10^{17}$ ions cm$^{-2}$, respectively. Arrow indicates the direction of ion-beam onto the surface.

Figure 4.13: Line profiles extracted from the AFM images of ion exposed samples at 72.5° for different ion fluences: (a) $1 \times 10^{17}$, (b) $2 \times 10^{17}$, (c) $5 \times 10^{17}$, (d) $10 \times 10^{17}$, (e) $15 \times 10^{17}$, and (f) $20 \times 10^{17}$ ions cm$^{-2}$, respectively. Arrow indicates the direction of ion-beam onto the surface.
4.12(d)-(f) and Figs. 4.13(d)-(f) which reveal clear enhancements in lateral dimension and height of the faceted structures with increasing ion fluence.

Figure 4.14: Schematic diagram depicting the mechanism responsible for coarsening of faceted structures at 72.5° (as a representative one). Arrows indicate the incident ion-beam.

Formation of faceted structures and their coarsening behaviour discussed above are beyond the scope of linear stability analysis of BH theory because of the presence of ion-beam shadowing and possible slope-dependent nonlinear effect. In the linear regime, based on Sigmund’s theory of sputtering [47], BH theory takes into account a competition between curvature dependent sputtering and surface diffusion. Sputtering is treated as a surface roughening mechanism in this theory and hence, it is always useful to study the temporal evolution of surface roughness under ion-beam erosion to address pattern formation. Fig. 4.15 presents the roughness spectrum (i.e. variation in surface roughness with ripple wavelength/facet base-width) for both the angles under consideration. In both cases we observe an increase in roughness with increasing feature (ripple/facet) dimension. Under this frame work, we examine the role of sputtering by using Eq. 3.63 which is solved by assuming the dependence: \( Y(\theta) = Y(0) \sec \theta \) [48]. Although this form is known to be reasonable for not too large values of \( \theta \), in our case this approximation simplifies the sputtering yield calculation and explains our results qualitatively.
Figure 4.15: Variation in rms surface roughness, ‘w’ with lateral feature dimension corresponding to both angles of incidence.

The variation in fractional change of the sputter erosion rate, \( F \), with ripple wavelength/facet base-width is shown in Fig. 4.16 for both the angles under consideration. It is observed from Fig. 4.16 that \( F \) follows nearly the similar trend as observed in the case of surface roughness (although a slight mismatch is observed in case of 70\(^\circ\)). Therefore, results shown in Figs. 4.15 and 4.16 can be considered to be well correlated and confirm our claim that evolution of faceted structures at higher angles of incidence may also be driven by significant contribution from the sputter erosion induced roughening phenomenon.

Figure 4.16: Variation in fractional change of the sputter erosion rate, ‘\( F \)’ with lateral feature dimension corresponding to both angles of incidence.
4.3.4 Evolution of morphology under substrate rotation

An example of the evolution of surface morphology (along with the pristine silicon) at different incidence angles under continuous substrate rotation is depicted in Fig. 4.17(a)-(f). Figure 4.17(a) shows presence of smooth surface (rms roughness=0.1 nm) in case of ion exposed silicon substrate at 40° at a fixed fluence of 5×10^{17} ions cm^{-2} under continuous azimuthal rotation (rotational speed 0.5° s^{-1}). Figs. 4.17(b-d) show nanoscale mound formation at three different oblique angles of incidence, namely 60°, 67°, and 70°, respectively. It may be mentioned that our previous experiments show presence of ripples and mounds at these particular angles without any substrate rotation [15].

Figure 4.17: AFM images of (a) Pristine-Si and and exposed Si to 500 eV argon ions at a fluence of 5×10^{17} ions cm^{-2} at different angles under continuous substrate rotation in the angular range of 40°-85°. Corresponding height scales in (a-f) are 1 nm, 1 nm, 4 nm, 8 nm, 10 nm, and 2 nm, respectively. Insets shows 2D FFT obtained from the corresponding images. (g) Shows angular dependence of surface roughness for both static and rotating samples. The reduction in surface roughness is very clear in case of rotating samples.
Figure 4.17(e) reveals the presence of bigger mounds (diameter~80 nm) at 72.5° incidence angle whereas smooth surface in Fig. 4.17(f) is observed due to ion bombardment at 85° incidence angle with substrate rotation. Thus, in comparison to our earlier results [15], it is observed that flat surface remains flat whereas ripple patterns get transformed into mounds by losing their spatial anisotropy [as confirmed from the inset of Figs. 4.17(a)-(e) where the respective 2D FFT shows the absence of any satellite peak]. As seen in earlier cases, ripple formation can be avoided using a rotating substrate albeit our results end up in mound formation instead of a smooth surface. Corresponding rms roughness evolution plots are shown in the Fig. 4.18 as a function of incidence angle of the ion-beam with and without substrate rotation. It is clear from this image that there is a significant reduction in the roughness values using the concurrent substrate rotation. The maximum decrease occurred in case of 72.5° where \( w \) goes down below 2 nm from 5.7 nm under substrate rotation.

![Roughness evolution plot](image)

Figure 4.18: Roughness evolution plot as a function of ion incidence angle (shown later). Solid lines are guide to the eyes.

The mound size is observed to increase with angle of incidence (e.g. in case of 60°, 67°, and 70° mound dimensions are 28, 32, and 38 nm, respectively). At a much higher oblique angle of incidence, effect of substrate rotation again becomes trivial which is evident from Fig.
4.17(f) where a flat surface emerges at 80° incidence angle. Thus, following the roughness evolution plot, one can infer that under concurrent substrate rotation, with increasing angle of incidence, silicon surface evolves from flat surface to intermediate mounded surface (rougher surface) to flat surface.

In Fig. 4.19, we compile our observations into a phase diagram of patterns formed (or lacked thereof) versus control parameter like angle of incidence under two experimental conditions, viz. with and without concurrent substrate rotation. To the best of our knowledge, this would be the first study of this kind which addresses temporal evolution of silicon surface under both static and rotating substrate condition. From this phase diagram, it is clear that the surface remains stable up to $\theta = 51^\circ$ whereas parallel-mode ripples undergo a transition to mounds under substrate rotation. At high $\theta (=80^\circ)$, again stable surface merges.

![Phase diagram](image)

Figure 4.19: Phase diagram for different patterns versus control parameters like ion incidence angle ($\theta$) and ion-beam energy ($E$). Both phase diagram are in the linear regime corresponding to the respective angle of incidence.
To understand the observations mentioned above, we first consider the situation without any substrate rotation. Let the unperturbed steady-state solution be \( h = h_0 - v_0 t \), where \( h_0 \) and \( v_0 > 0 \) are constants. For a surface that is not flat, \( h = h_0 - v_0 t + u \), where \( u = u(x,y,t) \) and including only the lowest order nonlinearities, the equation of motion becomes

\[
u = v_0 u_x + A_0 u_{xx} + A_1 u_{yy} - B \nabla^2 \nabla^2 u + \frac{1}{2} \lambda u_x^2 + \frac{1}{2} \lambda u_y^2,
\]

where the subscripts denote the partial derivatives like \( u_t = \partial u / \partial t \) etc.

We know from BH theory:

1. For \( A_1, A_2 > 0 \), the surface remains flat,
2. For \( A_1 < 0 \) and \( A_1 < A_2 \), parallel-mode ripples develop, and
3. For \( A_2 < 0 \) and \( A_2 < A_1 \), perpendicular-mode ripples emerge.

Now, under concurrent substrate rotation, if the angular speed is sufficiently large, the equation of motion becomes [41]:

\[
u = A \nabla^2 u - B \nabla^2 \nabla^2 u + \frac{1}{2} \lambda (\nabla u)^2
\]

where \( A = \frac{1}{2} (A_1 + A_2) \) and \( \lambda = \frac{1}{2} (\lambda_1 + \lambda_2) \).

If \( A > 0 \), the surface remains flat. If \( A < 0 \), on the other hand, Eq. 3.65 is the Kuramoto-Sivashinsky equation and the surface becomes unstable. At long times, the surface consists of an irregular array of mounds and it exhibits a spatio-temporal chaos. These theoretical predictions match well (qualitatively) with our experimental results shown in Fig. 4.16 where the presence of irregular mounds are observed within the angular range of 51°-72.5°. The plot shown in Fig. 4.16(g) does not clearly identify the linear and nonlinear regimes. Thus, it would be difficult to draw any conclusion out of this plot. However, this problem can be overcome by performing the experiments in the linear regime. According to Bradley’s theory, at early
times when the system evolves in the linear regime, the nonlinear terms in Eqs. (1) and (2) can be neglected and the rms roughness should be

\[ \sigma_{nr} \geq \sigma_r \]  \hspace{1cm} (3.66)

where, \( \sigma_{nr} \) and \( \sigma_r \) are roughness values for both without and with substrate rotation.

Physically, this means that the amplitude of the mounds formed with sample rotation grows more slowly than the amplitude of the ripples formed without sample rotation.

To verify the prediction mentioned in Eq. (3.66), we performed similar angle-dependent experiments by selecting a proper timescale where the pattern formation can be described by taking into account linear approximation only (the nonlinear term in Eq. 3.65 can be ignored). To find out the timescale, we took help of the solid flow model and calculated the upper limit of intrinsic time scale of pattern evolution. In this process, we observed that 5 min would be a safe time scale to remain in the linear regime for the angular window of 55°-67° whereas 2 min should the appropriate one for the angular window of 70°-72.5°. Thus, we carried out experiments within the angular range of 55°-67° with and without substrate rotation. It may be mentioned that particularly for this case, we chose the rotational speed to be 6°/s or 0.104 rad/s so that we can safely use the linear extension of Eq. 3.65. From Fig. 4.20, it is obvious that rms roughness decreases as we switch on the substrate rotation. The above mentioned experiments clearly demonstrate the validity of prediction mentioned in Eq. 3.66.
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Figure 4.20: Roughness evolution plot in linear regime as a function of ion incidence angle.

**Temporal evolution of mounds:**

Temporal evolution of the mounds under continuous substrate rotation is summarized in Figs. 4.21(a-d) at 67\(^\circ\) incidence angle. Fig. 4.21(a) shows the morphology corresponding to an exposure time of 11 min where smaller dimensional mounds are visible. With increasing the sputtering time up to 110 min, mounds grow in dimension.

Figure 4.21: AFM images showing temporal evolution of mounds corresponding to different times (fluences) namely (a) 11 min, (b) 22 min, (c) 54 min, and (d) 108 min at an incidence angle of 67\(^\circ\). Corresponding height scales in (a-d) are 6 nm, 8 nm, 8 nm, and 13 nm, respectively. Insets show 2D FFT obtained from the corresponding images.
The inset of the respective images does not show any anisotropy from the 2D FFT (no satellite peak is observed). This behaviour can be well realized from Figs. 4.22 (a) and (b) where one can see that the growth of mounds is proportional to the square root of the sputtering time. This is followed by an increase in rms roughness in a linear fashion as a function of sputtering time. It can be mentioned here that Frost et al. observed coarsening of mounds (followed by an increase in rms roughness) with increasing sputtering time in case rotating InP substrates under argon-ion bombardment [49]. This behaviour of mound coarsening is not consistent with the equations (e.g. KS equation or Eq. 3.65) that describe topographical evolution due to sputtering. Frost et al. attributed their results to the preferential sputtering of phosphorous which leads to indium enrichment and agglomeration at the surface. On the contrary, for a silicon surface the possibility of preferential sputtering is totally ruled out.

Figure 4.22: (a) Variation in the lateral dimension of mounds as a function of square root of the sputtering time. (b) rms roughness plot as a function of sputtering time at 67° angle of incidence. Dashed lines are for guide to the eyes.

Hence, to account for the results shown in Fig. 4.22, we must go beyond Eq. 3.65. In particular, the next-to-lowest order nonlinearity must be appended to Eq. 3.65. This nonlinearity is
discussed by García et al. [50]. According to them, the more refined equation of motion should be

\[ u_r = A\nabla^2 u - B\nabla^2\nabla^2 u + \frac{1}{2}(\nabla u)^2 - \frac{1}{2}\mu\nabla^2(\nabla u)^2 \]  

(3.72)

Simulation of this equation reveals that once the mounds form, they get coarsened for a certain amount of time but then stop growing. This is called “interrupted coarsening”. In the present experiments, we have observed the coarsening, but the arrest of coarsening is not. To observe this arrest, one should go to even higher fluences.

In order to do so, we chose 60° incidence angle and carried out similar temporal evolution study. The results are presented in Figs 4.23 (a) and (b).

Figure 4.23: (a) Variation of the lateral dimension of the mounds as a function of square root of the sputtering time. (b) rms roughness plot as a function of sputtering time at 60° angle of incidence. Dashed lines are for guide to the eyes.

These two figures clearly show that the arrest of coarsening takes place faster in case of 60° angle of incidence and supports all the prediction by García et al. [50].

**Rotational speed variation**

The effect of rotational speed variation on the formation of nanostructures has not been studied to a great extent [51] in case of sputtering process. Thus, to explore the consequence of rotational speed variation, we exposed silicon wafers to 500 eV argon ion-beam at an oblique incidence of 67° with rotational speed varying in the range of 8×10²-44×10² rad s⁻¹.
constant fluence of $5 \times 10^7$ ions cm$^{-2}$ (selective images are shown in Fig. 4.24). Corresponding roughness evolution plots as a function of rotational speed are shown in Fig. 4.25(a) while Figs. 4.25(b) and (c) depict the variation in the mound size and the mound amplitude as a function of rotational speed ($\omega$), respectively. It is observed that with increasing $\omega$, roughness and mound height show an increasing trend up to a speed of $35 \times 10^2$ rad s$^{-1}$ whereas beyond this it gets saturated (within our present experimental domain). On the other hand, mound dimension remains constant as a function of substrate rotational speed [Fig. 4.25 (b)]. It can be mentioned that recently, Chowdhury et al. showed a non-monotonic increase in the rms roughness as function of $\omega$ over a wide range of variation (0-25 rpm) [51].

Figure 4.24: AFM images showing the effect of rotational speed variation corresponding to different angular speeds, namely (a) 17.4, (b) 26.2, and (c) 43.6 rad s$^{-1}$ at an incidence angle $67^0$. Corresponding height scale in (a-c) is 11 nm. Insets shows 2D FFT obtained from the corresponding images.

Figure 4.25: (a) rms roughness versus rotational speed variation plot for $67^0$ and at a fixed fluence of $5 \times 10^{17}$ ions cm$^{-2}$ incidence angle. (b) and (c) show corresponding mound dimension and height versus rotational speed plots, respectively.
According to Yasseri et al., one should observe non-monotonic dependence of the rms roughness upon the rotational speed ($\omega$) [52]. However, we restricted ourselves within the range of 0.083-1.33 rpm (due to the instrumental limit) and also observe a non-monotonic nature in roughness values.

4.4 Conclusions

In this chapter, experimental results were presented to understand low energy ion induced ripple formation in the framework of ion induced solid flow model over an angular range of (51°-72.5°). The present experimental results show the existence of an intrinsic timescale (where ripple evolution is in the linear regime) and beyond which the nonlinear effects dominate (e.g. ripple coarsening starts). It is observed that the experimentally determined intrinsic timescale for all angles matches quite closely with those predicted by the solid flow model and in general, it decays faster with increasing ion incidence angle. In addition, angle-dependent studies at higher fluences reveal that the transition from parallel-to perpendicular-mode ripples is not a sharp one but undergoes a series of unusual pattern formation (viz. mounds, facet/cone-like, and needle-like structures) at intermediate angles. We find that facet formation is a very special case where parallel-mode ripples are formed at lower fluences and subsequently undergo a transition to mounds/faceted structures. This transition from ripples to mounds and/or faceted structures is explained geometrically which takes into account the inter-peak shadowing effect. With increasing ion fluence, faceted nanostructures undergo coarsening, i.e. they grow bigger in both lateral dimension and height. The coarsening behaviour is explained by invoking Hauffe’s mechanism which is based on reflection of primary ions on facets. In addition, the effect of concurrent substrate rotation during ion bombardment is also investigated. It is found that, in the linear regime, substrate rotation leads to a reduction in surface roughness compared to the static case, which was supported by the theoretical predictions. Angle-dependent study reveals that a flat surface remains flat whereas
ripples transform into mounds under concurrent substrate rotation. Our results qualitatively support a nonlinear theory proposed by Bradley on surface roughening under substrate rotation. Temporal evolution of mounds show arrest of the coarsening behaviour which has been explained by appending next-to-lowest order nonlinearity in the nonlinear equation proposed by García et al. [50].

Bibliography

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