Chapter 7

Nanoscale self-affine surface smoothing by ion bombardment: Ion-fluence dependence

A scientist in his laboratory is not a mere technician: he is also a child confronting natural phenomena that impress him as though they were fairy tales.

– Marie Curie

7.1 Introduction:

In the molecular beam epitaxy (MBE) growth process, which is usually a high temperature growth, the diffusion process enables the growth of good quality layers with smooth interfaces. When the temperature is lowered, the interface begins to grow in a three-dimensional growth mode and acquires a non-zero roughness. In this regime continuum theories and scaling laws become applicable [1].

There are three main effects determining the final interface morphology of a growing interface: deposition, desorption and surface diffusion. Relative importance of these
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Effects depend on the microscopic properties of the interface, such as the magnitude of the bonding energies and diffusion barriers. These parameters can be modified only by changing the substrate. On a given substrate the experimentally controllable parameters are the deposition rate and temperature. By tuning these parameters a rich variety of morphologies can be achieved from smooth interface to rough self-affine surfaces.

Ion bombardment of surfaces, removes material from the substrate by a process called sputtering. As opposed to the process of deposition in MBE, here the inverse process of atom removal results in rough interfaces. Surface diffusion may act as a smoothing mechanism. A brief description of roughening by ion bombardment and the corresponding roughness exponent have been given by Barabasi and Stanley [2].

Ion bombardment of various surfaces have shown to produce a self-affine fractal surface both for the situations where ion bombardment causes surface roughening [3, 4, 5] or surface smoothing [6, 7]. Even for situations where ion bombardment causes smoothing over some length scales and roughening for other length scales in the same system, self-affine fractal surfaces are formed by ion bombardment [6, 7].

Growth on fractal surfaces [8] is an interesting area of research. Self-assembled nanostructures, such as quantum dots, are grown by MBE. These nanostructures are formed via Stranski-Krastanow (layer-plus-island) or Volmer-Weber (island) growth process. These structures are grown on flat surfaces. Shape of these nanostructures may be modified when grown on a fractal surface [8]. Electronic energy levels of such nanostructures may be tuned by controlling the shape.

In a previous study by Goswami and Dev [6] ion beam induced surface smoothing was observed; scaling behaviour of surface roughness has shown this surface to be a self-affine fractal surface with a roughness exponent of 0.53 ± 0.03. 2MeV Si\textsuperscript{+} ions at a fluence of 4\times10\textsuperscript{15} ions/cm\textsuperscript{2} is thought to be above the threshold fluence. An
investigation on the determination of threshold fluence [9] is presented in this chapter.

We present a brief description of how ion beams via ion-solid interactions, modify surface topology. This is followed by a description of ion beam induced surface smoothing, the scaling concept and roughness exponent. Scaling studies as a function of ion fluence, in order to determine the threshold fluence contribute the main aspect of this chapter.

7.2 Interaction of energetic ions with solid surfaces

When a beam of energetic ions enter and travel through a solid (target), energy from the ions is transferred to the solid via ion-solid interaction. This energy transfer causes various kinds of surface or subsurface modifications in the target material. These modifications depend on the type of ions, their energy and the nature of the solid. The main processes which undergo during the fast moving, unidirectional charged particles penetrating a solid target, are (i) penetration of ions into the solid, (ii) rearrangement of the surface and near-surface target atoms, (iii) emission of particles/atoms from the solid-surface. Ion-solid interaction can cause completely opposite effects like amorphization and crystallization. The energy transferred from the ions to the surface or near-surface atoms of the solid can cause atomic motions on the surface and even ejection (sputtering) of atoms. This leads to a modification of the surface topography via opposing effects like surface roughening and surface smoothing.

There are two distinct processes by which an energetic ion loses energy during its passage through a solid material. The first one corresponds to electronic energy loss where an energetic ion excites or ejects electrons of target atoms. In this process ion loses energy through inelastic interactions. The other process involves elastic collisions between the nuclei of the ions and the atoms in the solid. In this process an ion imparts a part of its energy to the atoms in the solid. This is known as nuclear energy loss and it is dominant mainly at lower ion energy regime. Here, energy is
transferred from the projectile to the target via Coulomb interaction between the screened charges of the two nuclei. Since the energy of an ion decreases through its path of penetration into the solid, for a high energy ion initially inelastic loss is dominant and towards the end of its path elastic energy loss is dominant. The energy loss of the projectile ions with penetration depth ‘x’ in the target material can be written as,

\[
\frac{dE}{dx} = \left(\frac{dE}{dx}\right)_{\text{nuclear}} + \left(\frac{dE}{dx}\right)_{\text{electronic}}
\]

and the ‘range’ (depth) of the ions before coming to rest is given by,

\[
R = \int_0^{E_0} \left(\frac{dE}{dx}\right)^{-1} dE
\]

where, \(E_0\) is the incident ion energy at the solid surface, \(x\) is the distance measured along the ion path, \(\frac{dE}{dx}\) is the total (nuclear + electronic) energy loss of the ion with energy \(E\). The depth distribution of the ions follows a Gaussian distribution function given by,

\[
N(x) = \frac{N_{\text{total}}}{\sigma \sqrt{2\pi}} \exp \left(-\frac{(x - \mu)^2}{2\sigma^2}\right)
\]

where \(N(x)\) is the number of ions at any depth \(x\), \(N_{\text{total}}\) is the total number of ions the solid surface has been exposed to, \(\mu\) and \(\sigma\) are the mean and the standard deviation respectively (\(\sigma^2\) is the variance). These ions are incorporated into the solid and are known as implanted ions. However, the energy imparted by ions to the atoms in the solid can displace these atoms, which may have sufficient energy to displace neighbouring atoms. Thus a cascade of displaced atoms is formed. Some of these displaced atoms have a momentum component outward from the surface. If they have an energy larger than the surface binding energy, these atoms are ejected from the surface. These atoms are sputtered atoms and the process is known as sputtering. Various process happening in ion-solid interactions are shown in Fig.7.1.
The evolution of surface topography during ion bombardment is governed by the interplay between the dynamics of surface roughening due to sputtering and smoothing due to material transport during surface diffusion. These competing processes are responsible for the creation of characteristic surface features like quasiperiodic ripples [3, 10, 11, 12] and self-affine topographies [3, 4, 5]. These have been observed in the ion energy regime where sputtering is dominant and ion incidence is tilted to the surface normal. Although there is a large number of observations of ripple formation there are only a few studies on the scaling of the surfaces evolving under ion bombardment [3, 4, 5, 6, 7]. Among these refs. [3, 4, 5] deal with surface roughening and ref. [6] and ref. [7] involve surface smoothing. Interestingly, both roughening and smoothing produce self-affine fractal surfaces. A common feature of most rough surfaces observed experimentally or in discrete models is that their roughness follows simple scaling laws. Surface root-mean-square roughness $\sigma$ is defined as

$$\sigma = \left< (h(x, y) - \bar{h})^2 \right>^{1/2},$$  

(7.4)

where $h(x, y)$ is the surface height at a point $(x, y)$ on the surface and $\bar{h}$ is the average height. The surface is termed self-affine if $\sigma$ changes with the horizontal sampling length $L$ according to $\sigma \propto L^\alpha$, where $0 < \alpha < 1$ is the roughness exponent [5].
roughness exponent quantifies how roughness changes with length scale and its value is indicative of the surface texture.

This work is mainly based on our previous observations of surface smoothing in the nanometer scale due to ion-irradiation effect. In this chapter, our primary aim is to discuss the ion-fluence dependence to achieve this nano-scale surface smoothing and the threshold ion-fluence value which is responsible to start the surface smoothing effect and transform the surface into a self-affine topography. These results have not been discussed in our previous reports. Therefore, here, our discussion and new results will mainly evolve based on ref. [6] and ref. [7].

7.3 Surface smoothing by ion bombardment

The first scaling studies involving surface smoothing by ion bombardment was reported by Goswami and Dev [6]. In this study Si(100) crystals were irradiated with 2.0 MeV Si⁺ ions at near-normal incidence. Only one half of the sample was irradiated and the surface roughness measurements were made on both the pristine and the ion-bombarded halves of the sample.

Typical STM images from the pristine and the ion-irradiated (fluence $4 \times 10^{15}$ ions/cm²) parts of a sample are shown in Fig.7.2(a) and Fig.7.2(b). The qualitative topographical difference between the two images is obvious. A quantitative difference can be revealed by a roughness scaling study. For each scan size ($L$) an average roughness ($\bar{\sigma}$) can be obtained. This is done as follows. A large number of scans, each of size $L$, are recorded on the surface at random locations. The $\bar{\sigma}$ values for the rms roughness given by the instrument for the individual scans are then averaged. This procedure is repeated for many different sizes and a set of average $\sigma$ vs $L$ values is obtained (each $\bar{\sigma}$ is usually the average of many measurements). Each $\sigma$ value is computed after instrument plane fitting and subtraction procedure has been carried out. $\bar{\sigma}$ vs $L$ log-log plots for the pristine and the irradiated surfaces are shown in Fig.7.3. A comparison shows that at length scales below $\sim 50$ nm the irradiated surface is smoother.
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Figure 7.2: STM images obtained from (a) pristine and (b) ion-irradiated Si(100) surfaces. The image size for both the cases is 300×300 nm² and the vertical scale from black to white is 2.2 nm. The height profiles obtained along the white lines marked in the images are shown below the images. Surface smoothing of the irradiated sample in smaller length scales is obvious from the height profiles. [From ref.[6]]

than the pristine surface. In addition, the linear dependance in the log-log plot for the ion-beam-smoothed surface and the value of $\alpha$ ($0 < \alpha < 1$), given by the slope, indicates that the surface is a self-affine fractal surface with a local fractal dimension $D = 3 - 0.53$ or 2.47. At length scales > 50 nm, ion irradiation appears to have no significant effect as the roughness remains practically unaltered. Two height profiles $h(x)$ measured along the lines marked in Fig.7.2(a) and Fig.7.2(b) are shown respectively below the images. At smaller length scales the pristine surface looks more rough. That is, ion bombardment has made the surface smoother at smaller length scales.

Earlier scaling studies [4, 5] involved ion-bombardment-induced surface roughening rather than smoothing. In these studies on ion-bombarded surfaces, the conditions of ion energy and the angle of incidence were favourable for strong sputtering and
Figure 7.3: Average root-mean-square roughness $\bar{\sigma}$ vs scan size $L$ on pristine (Δ) and ion-bombarded (○) Si(100) surfaces. [From ref.[6]]

sputter-erosion of surfaces caused surface roughening. In order to explain the dominance of smoothing over roughening a comparison of the sputtering yields has been made in ref.[6]. From the conditions in ref.[4] and ref.[5], where surface roughening was observed, the sputtering yields of 3.7 atoms/ion and 3.9 atoms/ion, respectively were estimated by TRIM (transport of ions through matter) calculation [6]. In ref.[6], where smoothing was observed, the higher ion energy and the normal incidence — both contributed to lowering the sputtering yield, which was < 0.2 atom/ion. Thus the sputtering yield is smaller by almost a factor of 20. This indicates why surface erosion, main reason for surface roughness enhancement, was not significant in ref.[6]. In fact at large length scales surface roughness remains unaffected by ion bombardment. On the other hand, the number of surface atoms that would contribute to effective surface mobility is large as discussed below. In ion-atom collisions in solids and at the surface, the elastic energy lost by an ion is transferred to a recoil atom, which itself
collides with other atoms in the solid and so forth. In this way the ion creates what is called a collision cascade. The displaced atoms in this collision cascade may acquire a kinetic energy enough to escape from the solid surface by sputtering. However, if the energy (component normal to surface) of the displaced atoms is smaller than the surface binding energy, the atoms may reach the surface but can not leave the surface. They can however drift parallel to the surface. Carter and Vishnyakov [11] discussed various surface relaxation mechanisms proposed earlier (such as, viscous relaxation effects, thermal surface diffusion or radiation assisted effective diffusion etc.) and found it necessary to invoke a further surface smoothing mechanism which dominates for normal and near-normal ion incidence conditions. This smoothing is due to those atoms which are ejected from the surface with too low an energy to escape the energy barrier but can translate parallel to the surface. This contribution can be estimated from $f(E)$, the number of atomic recoils generated by each incident ion. They have incorporated this $f(E)$-dependent smoothing term in the Bradley-Harper equation [13] and reached the qualitative conclusion that for $\theta = 0$, smoothing dominates roughening at all wave vectors.

A surface smoothing contribution coming from $f(E)$, as discussed above, could be dominant. This is conceivable from the presence of a large number of hyperthermal atoms on the surface. In order to illustrate this point a TRIM simulation has been presented in ref.[6], for 2 MeV Si$^+$ ions incident on Si at $\theta = 0^\circ$, to show the energy distribution of these atoms. $f(E) = k(E)/2E_d$, where $k(E)$ is the fraction of ion energy deposited in elastic collisions and $E_d$ is a displacement energy. In the simulation result $E_d = 15$ eV was used. This shows the atoms reaching the surface vs their energies normal to the surface. Atoms which have energies greater than the surface binding energy ($\approx 4.7$ eV) will be sputtered. However, a large number of atoms reach the surface with low energy ($< 4.7$ eV) with the number of atoms/eV peaking at $\sim 1$ eV. These atoms will not leave the surface (not be sputtered). The role of these atoms is important in surface smoothing. For $\theta \approx 0$ Carter and Visnyakov predict that smoothing dominates roughening at all wave vectors. We find that at larger

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length scales (> 50 nm) initial surface roughness remains practically unaffected by ion bombardment while smoothing becomes increasingly dominant at lower length scales below 50 nm. Carter and Vishnyakov did not predict the scaling exponent associated with this smoothing process.

Eklund et al [4] studied submicron-scale surface roughening induced by ion bombardment. They obtained a scaling exponent $\alpha \approx 0.2 - 0.4$, which is reasonably explained by the anisotropic Kardar-Parisi-Zhang (KPZ) equation ($\alpha = 0.38$) [14] when the surface diffusion term is negligible. On the other hand, there are no concrete predictions of the exponents for the case where ion-beam-induced surface smoothing or when diffusivity is dominant. Assuming the possibility that the scaling theories applicable to nonequilibrium film growth may also be applicable to ion bombardment, so long as no eroded material is redeposited onto the surface, the observed exponent was compared with those expected for the deposition process, which are $\alpha \approx 0.35$ when surface mobility of the deposited particles is ignored and $\alpha = 0.66$ when surface mobility is allowed [15, 16, 17]. In the first case the exponents are in good agreement for deposition and ion bombardment. In the second case surface mobility is important and the observed value of $\alpha = 0.53$ [6] is closer to that for the deposition model that includes surface mobility. (Incidentally, Krim et al. [5] also observed $\alpha = 0.53$ for ion bombardment of an Fe film on a MgO substrate where roughening, rather than smoothing, was dominant).

7.4 Surface smoothing: dependence on ion fluence

In ref.[6] surface smoothing and a concomitant formation of self-affine fractal surface was observed for an ion fluence of $4\times10^{15}$ ions/cm$^2$. Surface smoothing leading to the development of a linear region in $\log \sigma$ vs $\log L$ plot is expected to have a threshold fluence. Here we investigate the fluence dependence of the surface smoothing process. We use the same conditions as in ref.[6], namely bombardment of Si(100) crystals by 2.0 MeV Si$^+$ ions at near-normal incidence and the same kind of analysis as presented
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Figure 7.4: Surface roughness scaling results at different ion fluences. (a) $\phi = 1 \times 10^{14}$ ions/cm$^2$: pristine surface ($\square$), ion-irradiated surface (O) – marginal surface smoothing, but no formation of self-affine surface. (b) $\phi = 1 \times 10^{15}$ ions/cm$^2$: ion-irradiated surface. (c) $\phi = 5 \times 10^{15}$ ions/cm$^2$: ion-irradiated surface. At $5 \times 10^{13}$ ions/cm$^2$ there was practically no effect of ion-irradiation.

in the previous section.

Ion irradiation was carried out in the fluence ($\phi$) range $5 \times 10^{13} - 5 \times 10^{15}$ ions/cm$^2$. The results, log$\bar{\sigma}$ vs log$L$ plots, are shown in Fig.7.4. For the lowest fluence ($\phi = 5 \times 10^{13}$ ions/cm$^2$) there is hardly any observable change in surface roughness. For a fluence of $1 \times 10^{14}$ ions/cm$^2$, some smoothing has occurred at shorter length scales, however, no linear region has developed in the log$\bar{\sigma}$-log$L$ plot. At $\phi \geq 1 \times 10^{15}$ ions/cm$^2$ we notice surface smoothing along with a linear region in the log$\bar{\sigma}$ vs log$L$ plot. The value of the roughness exponent $\alpha$ between 0 and 1, indicates the formation of a self-affine
fractal surface. Thus, for the conditions used here the threshold fluence ($\phi_{th}$) appears to be close to $1 \times 10^{15}$ ions/cm$^2$ ($1 \times 10^{14} < \phi_{th} \leq 1 \times 10^{15}$ ions/cm$^2$). The roughness exponent observed here ($\alpha \approx 0.35$) contradicts that in ref.[6]. The exact reason for this deviation is not clear. One difference is the ion flux. In ref.[6], the ion beam flux was $1.3 \times 10^{12}$ ions/cm$^2$/sec. In the present study it is $\sim 1.0 \times 10^{12}$ ions/cm$^2$/sec. In the present set of samples, the roughness values were also found to be inhomogeneous, on the same sample the rms roughness varying in the range 2–4 Å for large $L$ values at different regions [9]. In order to study the effect of ion energy, a lower energy ion beam was used where the ion flux was maintained as in ref.[6] and a roughness exponent value of $\sim 0.5$ was obtained [18, 19]. These results are not included in this thesis. This is a likely indication on the importance of ion flux in determining the roughness exponent.

7.5 Summary, Discussions and Conclusions

When Si(100) surfaces are bombarded with 2 MeV Si$^+$ ions at a fluence of $4 \times 10^{15}$ ions/cm$^2$, smoothing is observed at smaller length scales while surface roughness remains practically unchanged at larger length scales. In the smoothed region, typically < 50 nm, the surface roughness has a scaling behaviour in conformity with a self-affine fractal surface. The observed roughness exponent $\alpha = 0.53$ corresponds to a local fractal dimension of 2.47. In order to study the evolution of surface smoothing as a function of ion fluence, studies were conducted in the ion fluence ($\phi$) range $5 \times 10^{13} - 5 \times 10^{15}$ ions/cm$^2$. At the lowest fluence, no significant change in surface roughness was observed. At $\phi = 1 \times 10^{14}$ ions/cm$^2$ some smoothing was observed, however, no self-affine surface was formed. At $\phi = 1 \times 10^{15}$ and $\phi = 5 \times 10^{15}$ ions/cm$^2$ surface smoothing led to the formation of a self-affine surface. The threshold fluence for this process appears to be $\sim 1 \times 10^{15}$ ions/cm$^2$ or more precisely $1 \times 10^{14} < \phi_{th} \leq 1 \times 10^{15}$ ions/cm$^2$. However, the roughness exponent observed in this study is $\alpha \approx 0.35$ in contrast to what was observed earlier ($\alpha = 0.53$). The source of this disagreement is not immediately clear. In this study of fluence dependence ion flux was intended
to be kept the same as that in the earlier study in ref. [6]. However, ion current fluctuation during irradiation produced an average flux of $1.0 \times 10^{12}$ ions/cm$^2$/sec, while the flux in ref. [6] was $1.3 \times 10^{12}$ ions/cm$^2$/sec. Although a higher ion flux can cause dynamic annealing aiding the surface smoothing process, the fluxes involved in these experiments are reasonably low and a strong thermal contribution to surface smoothing is not very likely. In the studies presented here, it turned out that the surface roughness was inhomogeneous. Samples cut from the same wafer showed different values of roughness. Also on the same sample the roughness varied over a factor of 2 on different regions. Whether this had any influence on the value of the roughness exponent is also not clear from our present understanding. However, some studies, carried out at lower ion energies but maintaining the same flux as in ref. [6], yielded $\alpha \approx 0.5$ pointing towards the importance of ion flux in determining the roughness exponent.
Bibliography


