Nanoscale self-affine surface smoothing by ion bombardment

D. K. Goswami and B. N. Dev*

Institute of Physics, Sachivalaya Marg, Bhubaneswar 751005, India

Abstract

Topography of silicon surfaces irradiated by a 2 MeV Si^+ ion beam at normal incidence and ion fluences in the range $10^{15} - 10^{16}$ ions/cm^2 has been investigated using scanning tunneling microscopy. At length scales below $\sim$ 50 nm, surface smoothing is observed; the smoothing is more prominent at smaller length scales. The smoothed surface is self-affine with a scaling exponent $\alpha = 0.53 \pm 0.02$.

PACS no. 61.16.Ch; 61.80.Jh; 68.35.Bs; 68.35.Ct

One of the fundamental problems in materials science is to understand the effects of particle radiation on solid surfaces. The evolution of solid surface topography during ion-beam irradiation 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 [1–4] and self-affine topographies [4–6]. 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 evolved in ion bombardment [4–6]. 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 σ is defined as $\sigma =< [h(x,y) - h]^2 >^{1/2}$, where $h(x,y)$ is the surface height at a point (x,y) on the surface and h is the average height. The surface is termed self-affine if σ changes with the horizontal sampling length L according to $\sigma \propto L^{\alpha}$, where $0 < \alpha < 1$ is the roughness exponent [6]. The roughness exponent quantifies how roughness changes with length scale and its value is indicative of the surface texture.

For graphite bombarded with 5 keV Ar ions at an angle $\theta = 60^{\circ}$ with respect to the surface normal, Eklund et al [5]. reported $\alpha \simeq 0.2 - 0.4$, consistent with the predictions of the Kardar-Parisi-Zhang (KPZ) equation in 2+1 dimensions. Krim et al [6] observed a self-affine surface roughness generated by 5 keV Ar ion bombardment of an Fe thin film sample at $\theta = 25^{\circ}$, with a scaling exponent α=0.53, with no theoretical model predicting this value. In all these cases an increase of surface roughness was observed due to ion bombardment. Since ion arrival on the surface is a stochastic process and sputtering events are spatially distributed and of variable magnitude, surfaces are generally roughened during bombardment. In all the studies mentioned above the conditions are such that the erosion of the surface due to sputtering in ion bombardment is dominant over surface atomic diffusion. However, if the surface atomic diffusion dominates over sputtering, surface smoothing rather than roughening can occur [2]. Carter and Vishnyakov [2] have shown that inclusion of a directed flux of atoms parallel to the surface, generated by ion bombardment, in a stochastic differential equation description of the dynamics of surface evolution during sputter-erosion can induce smoothing for near-normal ($\theta \approx 0$) ion incidence. The flux of atoms parallel to the surface provides an effective diffusion causing surface smoothing which competes with the roughening caused by sputtering. For $\theta \approx 0$, roughening is weak as sputtering yield is small and smoothing dominates. Indeed for an ion incidence angle $\theta \approx 0$, surface smoothing have been observed in ion bombardment over a large range of ion energies [2,7]. Although some observations of surface smoothing have been reported, to our knowledge there has been no scaling studies of ion-beam induced surface smoothing. In scaling studies for nonequilibrium film growth by deposition, a value of $\alpha \approx 0.35$ is expected when surface mobility of deposited particles are not allowed and α=0.66 is expected when surface mobility is allowed [8–10]. For ion-induced roughening the observed value of α=0.2−0.4 is in reasonable agreement with the exponent for growth without surface diffusion. For ion-beam induced smoothing, where surface diffusivity is important, one may expect a different value of the scaling exponent α.

In this Letter, we present scanning tunneling microscopy (STM) characterization of surface smoothing in 2 MeV Si^+ ion irradiation of Si surfaces at normal incidence ($\theta = 0$). At length scales below ~ 50 nm we observe smoothing of the ion-bombarded surface. The observed value of the roughness exponent $\alpha = 0.53 \pm 0.02$ indicates the self-affine nature of the smoothed surface. The ion irradiated surface shows smoother surface texture at smaller length scales. We have chosen MeV ions for which sputtering yield is small. In comparison, the collision-induced atomic displacement and effective surface diffusivity is large. Together with normal incidence, these conditions are expected to cause smoothing. The observation of scale dependent smoothing with increased smoothing at smaller length scales has direct bearing on ion beam processing of nanostructures.

Si(100) substrates were irradiated with 2.0 MeV Si^+ ions in the ion implantation beam line of our 3 MV tandem Pelletron accelerator [11,12]. The ion beam was incident along the surface normal ($\theta \approx 0$) and rastered on the sample in order to obtain a uniformly irradiated area. One half of the sample was masked and hence unirradiated. An ion beam flux of $\approx 1 \times 10^{12}$ cm^{-2} sec^{-1} was used with fluences in the range 10^{15} to 10^{16} ions/cm^2. The samples were kept at room temperature during ion irradiation. The pressure in the chamber was $\sim 10^{-7}$ mbar. The sample was then taken out of the irradiation chamber and inserted into a STM chamber (pressure: 3×10^{-10} mbar) with an Omicron variable temperature STM operating at room temperature. STM height calibration was done by measuring atomic step heights on clean Si(111) and Si(100) surfaces. Roughness measurements were made on the pristine and the irradiated halves of the sample. We did not remove the thin ($\sim$1.5 nm) native oxide from the Si surface because the surface topography may be perturbed by the effect of Ehrlich-Schwoebel barriers in different crystallographic directions on a crystalline surface. In this regard the presence of the thin oxide layer is helpful and the effect of the anisotropic diffusion can be neglected.

In order to determine the roughness exponent from STM images we follow the procedure described in ref. 6. Typical STM images from the pristine and the irradiated (fluence 4×10^{15} ions/cm^2) parts of a sample are shown in Fig.1. A large number of scans, each of size L, were recorded on the surface at random locations. The σ values for the rms roughness given by the instrument for the individual scans were then averaged. This procedure was repeated for many different sizes and a set of average σ vs. L values was obtained (each $\bar{\sigma}$ is the average of six to fifteen measurements). Each σ value was computed after the instrument plane fitting and subtraction procedure had been carried out. $\bar{\sigma}$ vs. L log-log plots for both halves of the sample are shown in Fig.2. For the ion-bombarded area of the sample we observe surface smoothing and by fitting the linear part of the data we obtain $\alpha = 0.53 \pm 0.02$ below a length scale of $\simeq$ 50 nm, indicating the self-affine nature of the irradiated surface. Below this length scale, the pristine half of the sample shows no linear region in the log-log plot of $\bar{\sigma}$ vs. L. Two vertical profiles $h(x)$ along the lines marked in Fig.1. are shown in the inset of Fig.2. It is also clear from these profiles that for the irradiated part of the sample the surface is much smoother at shorter length scales as indicated by the roughness data and the scaling exponent.

In earlier scaling studies [5,6] on ion-bombarded surfaces, the conditions of ion energy and the angle of incidence were favorable for strong sputtering and sputter-erosion of surfaces caused roughening. In order to explain the dominance of smoothing over roughening in our case let us first compare the sputtering yields. From the conditions in refs.[5] and [6], we estimate the sputtering yields of 3.7 atoms/ion and 3.9 atoms/ion, respectively, using the TRIM (transport of ions through matter) code [13]. In our case the higher ion energy and the normal incidence $-$ both contribute to lowering the sputtering yield, which is < 0.2

atom/ion. Thus the sputtering yield is smaller by almost a factor of 20. This indicates why surface erosion, main reason for roughness enhancement, is not significant in our case. In fact at large length scales surface roughness remains unaffected by ion bombardment. On the other hand, 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 − a phenomenon known as 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 cannot leave the surface. They can however drift parallel to the surface. We show the results of a TRIM simulation of sputtering yield for our case in Fig.3. This shows the atoms reaching the surface vs. their energies normal to the surface. Atoms which have energies greater than the surface binding energy (≈ 4.7 eV) will be sputtered. However, we notice that a large number of atoms reach the surface with low energy (< 4.7 eV) with the number of atoms/eV peaking at ~ 1 eV. These atoms will not leave the surface (not be sputtered) [14]. The role of these atoms is important in surface smoothing. These atoms have too low an energy (normal to surface) to escape the energy barrier at the surface and will translate parallel to the surface. This collision-induced atomic displacement and the consequent effective diffusivity parallel to the surface due to ballistic atomic transport can be the dominant surface relaxation mechanism. As a result smoothing may dominate roughening as discussed later in more details. Eklund at al [5] studied submicron-scale surface roughening induced by ion bombardment and obtained an scaling exponent $\alpha \simeq 0.2 - 0.4$. This value of the exponent is reasonably explained by the anisotropic KPZ equation ($\alpha = 0.38$) [15] when the surface diffusion term is expected to contribute negligibly. On the other hand, there are no concrete predictions of the exponents for the case where ion beam induced surface smoothing or diffusivity is dominant. Neither we know any scaling theory which predicts $\alpha \approx 0.5$. 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, we compare the observed exponent 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 [8–10]. In the first case the exponents are in good agreement for deposition and ion bombardment. In our case surface mobility is important and the observed value of $\alpha = 0.53$ is closer to that for the deposition model that includes surface mobility. Incidentally, Krim *et al.* [6] also observed $\alpha = 0.53$ for ion bombardment of an Fe film on a MgO substrate where roughening, rather than smoothing, was dominant.

For ion irradiation, Carter and Vishnyakov [2] derived an expression showing the relative magnitudes of the roughening (sputtering) term and the smoothing term due to recoiled atoms which qualitatively explains the domination of smoothing over roughening at normal and near-normal ($\theta \approx 0$) incidence of the ion beam. However, there is no prediction of scaling exponent. For $\theta \approx 0$ they predict that smoothing dominates roughening at all wave vectors. We find that at larger 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.

In order to show the relative strength of the smoothing and the roughening terms, Carter and Vishnyakov [2] extended the treatment given by Bradley and Harper [16], who showed (in 1+1 dimension) that, due to sputter-erosion alone, the deterministic defining equation for $h(x,t)$ can be written as

$$-\frac{\partial h}{\partial t} = \frac{J}{N}\, Y_0(\theta) - \frac{J}{N}\frac{\partial}{\partial \theta}\ [\ Y_0(\theta)\ cos\ \theta\ ]\ \frac{\partial h}{\partial x} + \frac{Ja}{N}\ Y_0(\theta)\ \Gamma_1(\theta)\ \frac{\partial^2 h}{\partial x^2} \tag{1}$$

where J is the mean ion flux incident at angle θ, N is the solid atomic density, $Y_0(\theta)$ is the sputtering yield of a plane surface, a is the mean depth of energy deposition by an ion, and

$\Gamma_1(\theta)$ is a function of θ, and standard deviations α and β of the bi-Gaussian ellipsoidal ion energy spatial deposition density function. For order of magnitude estimation the ellipsoidal distribution has been approximated by a spherical distribution with $a = \alpha = \beta$, in which case

$$\Gamma_1(\theta) = sin^2\,\theta - \left(cos^2\,\theta/2\right)\left(1 + sin^2\,\theta\right) \tag{2}$$

In order to introduce the effective diffusion parallel to the surface they estimated the atomic flux parallel to the surface to modify the last term in Eq.(1):

$$-\frac{J}{N}\left\{f(E)\ d\ cos2\theta\ - Y_0(\theta)\ a\ \left[sin^2\,\theta - \frac{cos^2\theta}{2}(1 + sin^2\theta\ )\right]\right\}\frac{\partial\,^2 h}{\partial\,x^2} \tag{3}$$

where $f(E)$ is the no. of recoil atoms each ion generates in the solid and d is the average distance traveled by the recoiled atoms. d is of the order of a few interatomic distances. For $\theta = 0$, the expression (3) [i.e, the last term in Eq(1)] is negative and smoothing dominates roughening at all wave vectors. $f(E) = k(E)/2E_d$, where $k(E)$ is the fraction of ion energy deposited in elastic collisions and E_d is a displacement energy [17]. In the simulation results shown in Fig.3 we have used E_d=15 eV. The results shown in Fig.3 only qualitatively shows how a large number of hyperthermal recoil atoms, arriving at the surface but unable to escape the surface, can cause surface smoothing as implied by Eq.(1) along with expression (3). Expression (3) only qualitatively describes the effect of $f(E)$ in surface smoothing. For a quantitative understanding future theoretical work should include the effect of a distribution like that shown in Fig.3. So far theoretical works concentrated only on the low ion energy regime where sputter-erosion is dominant and the approximation ($a = \alpha = \beta$) used in deriving the expression for $\Gamma_1(\theta)$ [Eq.(2)] may be valid. However, for high ion energies, it is not valid. For example in our case, for 2 MeV Si ions in Si, $a = 1.94$ μm, $\alpha = 248$ nm and $\beta = 288$ nm. In the existing theories it is assumed that energy released by the ions at a depth a contributes an amount of energy to surface points that may induce surface atoms to break their bonds and leave the surface [15]. This is true for low ion energies where a is small. However, ion energy release deep inside the sample would hardly have any effect on surface atoms. Future theories must take this aspect into account.

In conclusion, we have observed nanoscale surface smoothing in ion bombardment. The smoothed surface is a self-affine fractal surface with a scaling exponent $\alpha = 0.53 \pm 0.02$. Below a length scale of ~ 50 nm, the smoothing is more dominant at smaller length scales. This phenomenon may be used in reducing surface roughness of nanostructural devices by ion beam processing as ion beams are widely used in device fabrication. Transport in nanostructures is expected to improve when roughness is minimized. For an understanding of the scaling exponent observed in surface smoothing further theoretical studies will be necessary.

REFERENCES

[1] E. Chason, T. M. Mayer, B. K. Kellerman, D. T. McIlroy and A. J. Howard, *Phys. Rev. Lett.*, **72**, 3040 (1994).

[2] G. Carter and V. Vishnayakov, *Phys. Rev.*, **B54**, 17647 (1996).

[3] For a review, see K. Wittmaack, in *Practical Surface Analysis*, Vol.2 *Ion and Neutral Spectroscopy*, edited by D. Briggs and M. P. Seah (Wiley, Chichester, 1992) chap.3, p.122.

[4] S. Habenicht, W. Bolse, K. P. Lieb, K. Reimann, and U. Geyer, *Phys. Rev.*, **B60**, R2200 (1999).

[5] E. A. Eklund, R. Bruinsma, J. Rudnick, and R. S. Williams, *Phys. Rev. Lett*, **67**, 1759 (1991).

[6] J. Krim, I. Heyvart, D. V. Haesendonck, and Y. Bruynseraede, *Phys. Rev. Lett*, **70**, 57 (1993).

[7] A. Gutzmann, S. Klaumunzer, and P. Meier, *Phys. Rev. Lett.*, **74**, 2256 (1995).

[8] M. Kardar, G. Parisi, and Y. Zhang, *Phys. Rev. Lett.*, **56**, 889 (1986).

[9] D. E. Wolf, and J. Villain, *Europhys. Lett.*, **13**, 389 (1990)

[10] Z. W. Lai, and S. Das Sarma, *Phys. Rev. Lett.*, **66**, 2348 (1991).

[11] K. Sekar, P. V. Satyam, G. Kuri, D. P. Mahapatra, and B. N. Dev, *Nucl. Instr. and Meth.*, **B73**, 63 (1993).

[12] G. Kuri, B. Sundaravel, B. Rout, D. P. Mahapatra, and B. N. Dev, *Nucl. Instr. and Meth.*, **B111**, 234 (1996).

[13] SRIM 2000 − a version of the TRIM program: J. F. Ziegler, J. P. Biersack, and U. Littmark, *The Stopping and Range of Ions in Matter* (Pergamon Press, New York,

1995)

[14] These qualitative nature remains the same when the native oxide layer on the silicon surface is explicitly taken in the simulation. Both Si and O atom distributions peak at $\sim$1 eV, O peak appearing at a slightly higher energy.

[15] R. Cuerno, and A. L. Barabasi, *Phys. Rev. Lett.*, **74**, 4746 (1995).

[16] R. M. Bradley, and J. M. E. Harper, *J. Vac. Sci. Technol.*, **A6**, 2390 (1988).

[17] P. Sigmund, *Appl. Phys. Lett.*, **14**, 114 (1969).

*Corresponding author: bhupen@iopb.res.in

FIGURE CAPTIONS:

Fig.1. STM images recorded on a pristine (top) and ion-bombarded (bottom) silicon surface. The scan size is 300 $\times$ 300 nm^2 and the vertical scale (black to white) is 2.2 nm. Height profiles along the lines are shown in Fig.2.

Fig.2. Average root-mean-square roughness vs. scan size on the pristine and the ion irradiated surfaces. Each point represents an average of 6 to 15 scans recorded at random locations on the surface. Surface smoothing is observed at scan sizes below $\sim 50 \times 50$ nm^2. The least-squares fit (solid line) to the linear portion of the data for the irradiated sample gives the scaling exponent $\alpha = 0.53 \pm 0.02$. No linear part is observed for the pristine sample data. Two vertical profiles $h(x)$ measured along the lines marked in Fig.1, are shown in the inset (scales in nm): (a) pristine, (b) irradiated sample.

Fig.3. A Monte-Carlo simulation result showing the energy distribution of ion-beam induced displaced atoms reaching the surface. Atoms with energy $>$ 4.7 eV leave the surface (sputtered). The large number of atoms below 4.7 eV (surface binding energy) cannot leave the surface and contribute to an effective surface diffusion due to ballistic atomic transport leading to smoothing.

FIGURES

Fig.1 (top)

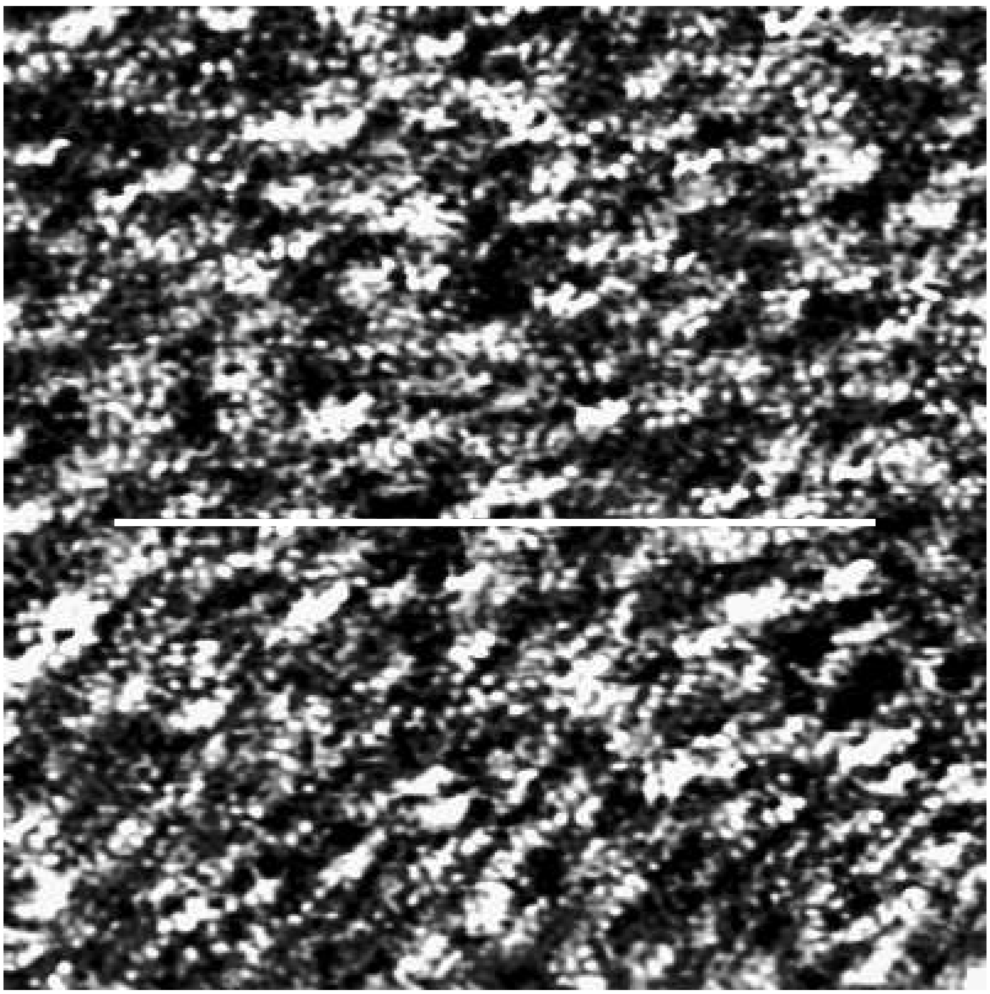


Fig.1 (bottom)

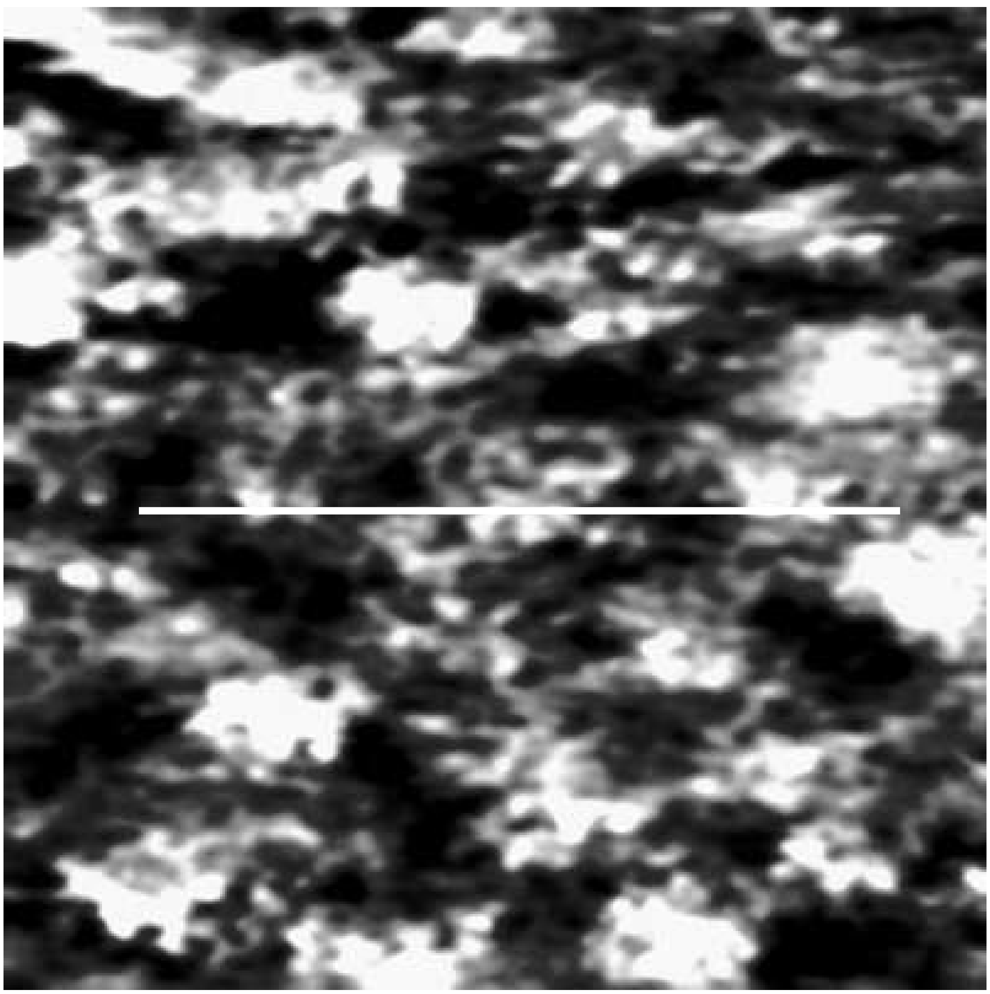
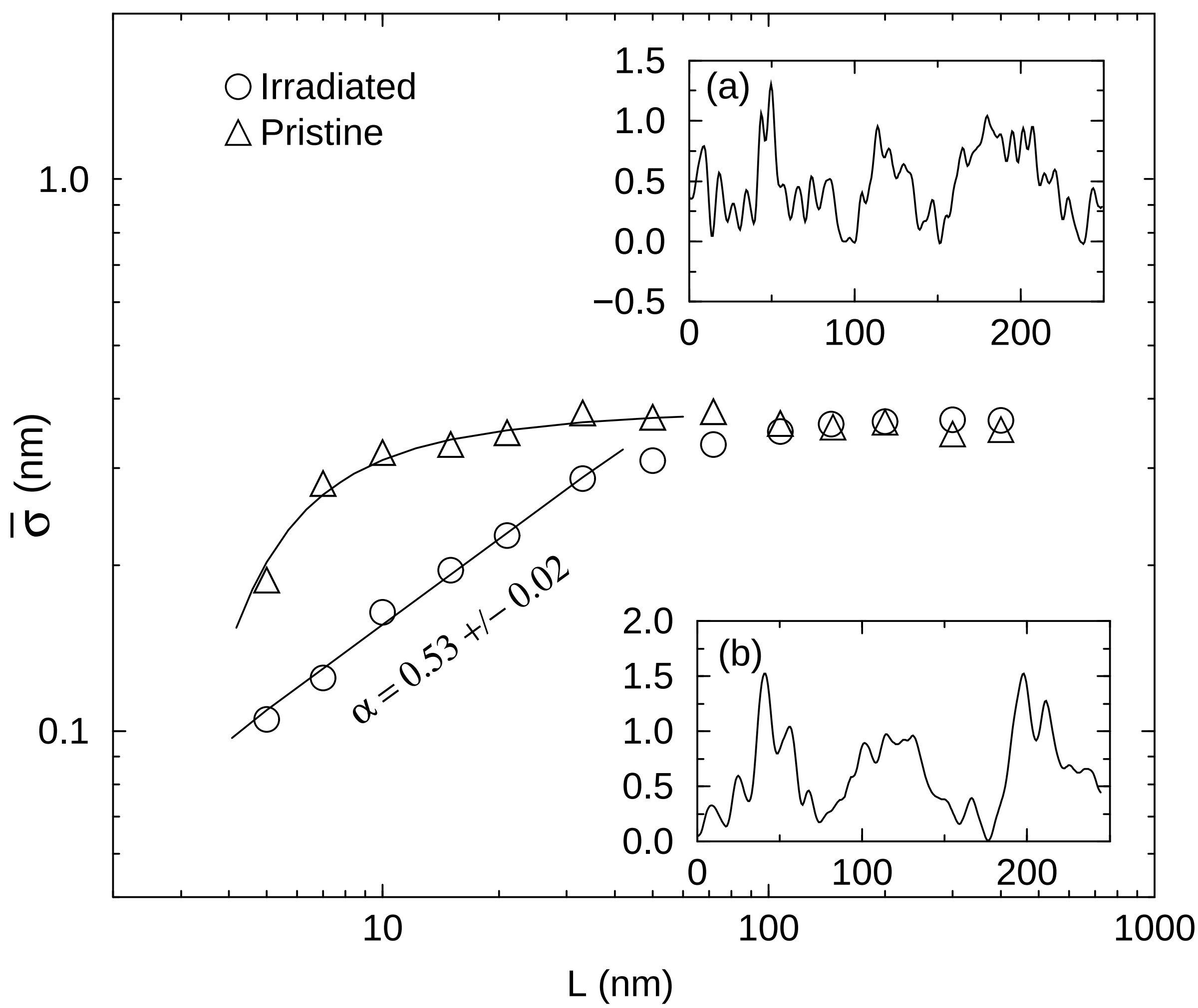
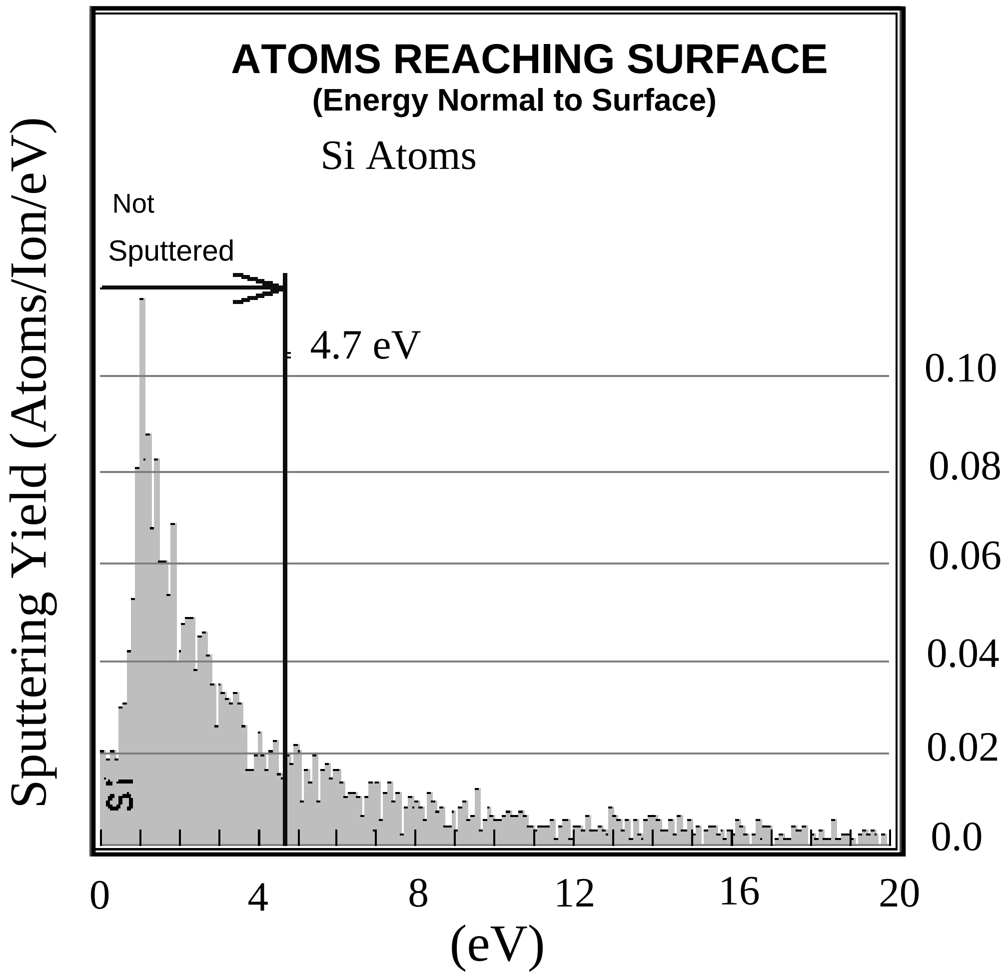


Fig.2.



Irradiated
Pristine
1.0
0.1
10
100
1000
L (nm)
σ̄ (nm)
α = 0.53 +/− 0.02
(a)
1.5
1.0
0.5
0.0
−0.5
0
100
200
(b)
2.0
1.5
1.0
0.5
0.0
0
100
200

Fig.3


ATOMS REACHING SURFACE
(Energy Normal to Surface)
Si Atoms
Not
Sputtered
4.7 eV
Si
Sputtering Yield (Atoms/Ion/eV)
0.10
0.08
0.06
0.04
0.02
0.0
0
4
8
12
16
20
(eV)