Nanoscale self-affine surface smoothing by ion bombardment

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(Received 26 February 2003; published 1 July 2003)

The 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 $\alpha=0.53\pm0.03$.

DOI: 10.1103/PhysRevB.68.033401 PACS number(s): 61.80.Jh, 68.35.Bs, 68.35.Ct, 68.37.Ef

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 characteristic surface features like quasiperiodic ripples and self-affine topographies. These have been observed in the ion energy regime where sputtering is dominant and ion incidence is tilted to the surface normal. Although there are a large number of observations of ripple formation, there are only a few studies of the scaling of surfaces evolved in ion bombardment. 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 $\sigma=\sqrt{\langle h^2 \rangle - \langle h \rangle^2}$, where $h(x,y)$ is the surface height at a point $(x,y)$ on the surface and $\langle h \rangle$ is the average height. The surface is termed self-affine if $\sigma$ changes with the horizontal sampling length $L$ according to $\sigma \sim L^\alpha$, where $0<\alpha<1$ is the roughness exponent. 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, Ekblund et al. reported $\alpha=0.2-0.4$, consistent with the predictions of the Kardar-Parisi-Zhang (KPZ) equation in 2+1 dimensions. Krim et al. 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 $\alpha=0.53$, 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. Carter and Vishnyakov 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 ($\theta=0$) 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 $\theta=0$, roughening is weak as sputtering yield is small and smoothing dominates. Indeed, for an ion incidence angle $\theta=0$, surface smoothing has been observed in ion bombardment over a large range of ion energies (keV to hundreds of MeV). In a recent study Mayr and Averback reported the observation of surface smoothing in 1.8 MeV Kr$^+$ ion irradiation of amorphous Zr$_{57}$Al$_{17}$Cu$_{27.4$ alloy and attributed the smoothing to irradiation-induced viscous flow. Although many 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 non-equilibrium film growth by deposition, a value of $\alpha=0.35$ is expected when surface mobility of deposited particles is not allowed and $\alpha=0.66$ is expected when surface mobility is allowed. For ion-induced roughening the observed value of $\alpha=0.2-0.4$ 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 $\alpha$. Various other mechanisms that could lead to smoothing may lead to different values of $\alpha$. At present, for ion-beam-induced smoothing, neither any theoretical prediction nor any experimental determination of $\alpha$ is available.

Here we report the determination of the roughness exponent $\alpha$ for a surface smoothed by ion bombardment. We present a 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 $\sim 50$ nm we observe smoothing of the ion-bombarded surface. The observed value of the roughness exponent $\alpha=0.53\pm0.03$ 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 the sputtering yield is small. In comparison, the collision-induced atomic displacement and effective surface diffusivity are 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. The ion beam was incident along the...
surface normal ($\theta=0$) 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 $10^{15} - 10^{16}$ ions/cm$^2$. The samples were kept at room temperature during ion irradiation. The pressure in the chamber was $\approx 10^{-7}$ mbar. Considering the vacuum in our irradiation chamber ($\approx 10^{-7}$ mbar) one may think of the possibility of C deposition on the surface which might affect the results of further measurements on the irradiated samples. It may be noted that we do not use any diffusion pump. The accelerator and beam line have cryopumps and ion pumps, and the chamber is evacuated with a turbomolecular pump. In earlier ion irradiation experiments with this facility for comparable ion flux and even higher fluences we could not detect any C deposition on the surface, using very sensitive techniques like x-ray reflectivity and Rutherford backscattering spectrometry under channeling conditions (RBS/C). It may be noted that the RBS/C technique is very sensitive to detect impurities on surfaces including C. The problem of C deposition was specifically addressed in Ref. 14. In the present case we have not performed a RBS/C analysis to avoid further exposure of the sample to ion beams. Based on our experiments in Ref. 14 we feel that our results here are not affected by any significant C deposition on the surface.

Following ion irradiation, the samples were taken out of the irradiation chamber and inserted into an ultrahigh vacuum (UHV) chamber with an Omicron variable-temperature (VT) STM equipment. The UHV system ($3 \times 10^{-10}$ mbar) with VTSTM, coupled with a molecular-beam epitaxy system, has been described elsewhere. STM height calibration was done by measuring atomic step heights on clean Si(111) and Si(100) surfaces. Roughness measurements were made at room temperature on the pristine and irradiated halves of the sample. We did not remove the thin ($\approx 1.5$ 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 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 $4 \times 10^{15}$ 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 $\sigma$ 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 $\sigma$ vs $L$ values was obtained (each $\sigma$ is the average of 6–15 measurements). Each $\sigma$ value was computed after the instrument plane fitting and subtraction procedure had been carried out. $\sigma$ 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 $\alpha=0.53\pm0.03$ below a length scale of $\approx 50$ nm, indicating the self-affine nature of the ion-bombardment-induced smoothed surface. Below this length scale, the pristine half of the sample shows no linear region in the log-log plot of $\sigma$ 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.

Earlier scaling studies involved ion-bombardment-induced surface roughening rather than smoothing. In these studies of 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, respec-
In our case the higher ion energy and the normal atomic recoils generated by each incident ion. They have contribution can be estimated from energy barrier but can translate parallel to the surface. This is due to those atoms which are ejected from the surface with too low an energy to escape from the solid— 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. Carter and Vishnyakov predict that smoothing dominates roughening at all wave vectors. We find that at larger length scales initial surface roughness remains practically unaffected by ion bombardment while smoothing becomes increasingly dominant at lower length scales below 50 nm. They do not predict the scaling exponent associated with this smoothing process.

Eklund et al. studied submicron-scale surface roughening induced by ion bombardment and obtained a scaling exponent \( \alpha = 0.2 - 0.4 \). This value of the exponent is reasonably explained by the anisotropic KPZ equation \( \alpha = 0.38 \) (Ref. 21), when the surface diffusion term is expected to contribute negligibly. On the other hand, there are no concrete predic-
tions of the exponents for the case where ion-beam-induced surface smoothing or diffusivity is dominant. Assuming the possibility that the scaling theories applicable to nonequilibrium film growth may also be applicable to ion bombardment, as long as no eroded material is redeposited onto the surface, we compare the observed exponent with those expected for the deposition process, which are $\alpha = 0.35$ when surface mobility of the deposited particles is ignored and $\alpha = 0.66$ when surface mobility is allowed.\(^9\) 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.)

Mayr and Averback\(^8\) also observed surface smoothing in ion irradiation. They identified the smoothing process using stochastic rate equations for the evolution of the surface $h(\mathbf{x},t)$ in Fourier space $[h(\mathbf{q},t)]$. The spectral power density is $C(q) = D(q)/\Sigma a_i q_i$, where dominant surface relaxation mechanism can be identified from the log-log plot of $C(q)$. The dominant power of $q$ identifies the smoothing mechanism. In their example, they attributed the smoothing predominantly to irradiation-induced viscous flow. However, they have not derived any scaling exponent. It would be interesting to know the predicted values of $\alpha$ from different surface relaxation mechanisms.

In conclusion, we have observed nanoscale surface smoothing in ion bombardment and determined the roughness exponent of the smoothed surface. The smoothed surface is a self-affine fractal surface with a roughness exponent $\alpha = 0.53 \pm 0.03$. Below a length scale of $\sim 50$ 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 aimed at deriving scaling exponents for various surface relaxation mechanisms will be necessary.

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20 This 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, the O peak appearing at a slightly higher energy.