

## Roughening and ripple instabilities on ion-bombarded Si

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Experimental studies of 10–40-keV  $\text{Xe}^+$  ion bombardment of Si at polar incidence angles between  $0^\circ$  and  $45^\circ$  to the surface normal at temperatures between 100 and 300 K show little roughening for near normal incidence but ripple production for  $45^\circ$  incidence. It is shown that inclusion of the directed flux of atoms parallel to the surface and generated by ion bombardment in a stochastic differential equation description of the dynamics of surface evolution during sputtering erosion can induce smoothing for near-normal ion incidence. For oblique incidence, roughening and ripple production occurs with a late stage dynamics dictated by the competition between curvature-dependent sputtering processes and surface relaxation (which is also, probably, irradiation motivated), gradient-dependent sputtering, and other higher-order effects.

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### INTRODUCTION

Ion-bombardment sputtering of solids leads to their erosion and, since ion arrival is a stochastic process and sputtering events are spatially distributed and of variable magnitude, surfaces are generally roughened during bombardment. In the case of semiconductors and other materials which are amorphized by bombardment, this roughening has been observed to be spatially periodic<sup>1,2</sup> and of ripple form, with a wave vector parallel to the projection of the ion flux on to the sample. A number of earlier studies<sup>3,4</sup> demonstrated that this ripple structure apparently only develops for ion flux polar incidence angles  $\theta$  to a surface normal greater than  $30^\circ$ – $40^\circ$ . The present studies, with 10–40-keV  $\text{Xe}^+$  bombardment at normal incidence or a polar incidence angle of  $45^\circ$ , on to Si were undertaken in order to explore this phenomenon further. Additionally, ion fluence was varied over several orders of magnitude ( $10^{17}$ – $10^{20}$   $\text{cm}^{-2}$ ) and the substrate temperature was varied from 100 to 300 K to examine the factors which induce ripple formation and evolution. It will be demonstrated that the absence of ripple formation for near-normal incidence was confirmed, and that for  $45^\circ$  incidence the results do not correspond to current theory on the competition between curvature-dependent sputtering and thermal surface diffusion in initiating and developing ripples.

Recently a number of theoretical studies<sup>5–10</sup> have examined the dynamics of the roughening instability and ripple production based upon the above or a similar process competition, but these cannot explain the lack of ripple production for small values of  $\theta$ , nor, indeed, the absence of general roughening with a preferred correlation length. The present investigations show how inclusion of a previously ignored surface smoothing mechanism can resolve this dilemma. The majority of earlier treatments<sup>5–9</sup> are based upon the competition between roughening processes which result from the surface curvature dependence of the sputtering process<sup>10–12</sup> and the random nature of sputtering and smoothing processes resulting from surface<sup>2,10,13</sup> or bulk diffusion,<sup>14</sup> and the surface gradient dependence of the sputtering process.<sup>5,6,10,15</sup> Carter, Nobes, and Katardjiev<sup>16</sup> also suggested that ion and atom-atom collision processes could provide a ballistic

atomic drift parallel to the surface in addition to the generally accepted isotropic ballistic diffusivity,<sup>17</sup> and included this in a generalized stochastic differential equation description of surface evolution, but did not examine its detailed form nor its influence on the surface dynamics. The present study examines this effect in more detail, and shows how its inclusion can aid an explanation of the experimental results outlined earlier.

### EXPERIMENT

$\text{Xe}^+$ -ion bombardments of Si at energies of 10, 20, and 40 KeV were undertaken with an isotope separator with ion flux densities of  $6 \times 10^{15}$  ions  $\text{cm}^{-2} \text{s}^{-1}$ , ion fluences in the range  $10^{17}$ – $10^{20}$   $\text{cm}^{-2}$ , and for target temperatures maintained between 100 and 300 K. The majority of the studies were undertaken for a polar incidence ion angle of  $45^\circ$  to the surface normal, but comparative studies were also made for normal and near-normal ion incidence. Following irradiation the surface topography was examined by atomic force microscopy (AFM) including two-dimensional Fourier-transform analysis.

Before irradiation the surface exhibited a random roughness of mean amplitude 2 nm. After irradiation the first signs of a correlated periodic structure resulting from 40-KeV irradiation at 300 K were clearly observed for an ion fluence of about  $10^{17}$  ion  $\text{s cm}^{-2}$ . This structure and fluence seem to be independent of the initial surface orientation which is entirely expected since the irradiation would create amorphization of the near-surface region of the Si at a fluence 2–3 orders of magnitude lower than this value. Figure 1 shows typical AFM images, cross sections, and two-dimensional Fourier transforms for irradiations at 100 K. It is clear that a quasiperiodic ripple structure of approximate wavelength  $\lambda = 0.4 \mu\text{m}$  is generated, with wave vector  $q_c$  parallel to the ion flux projections on the surface. When bombardment was conducted at normal incidence, even to much higher fluences, not only was the surface roughness much less than for  $45^\circ$  incidence, there was no sign of any surface correlation length similar to the  $0.4\text{-}\mu\text{m}$  periodic structures. Further studies with other incidence angles between  $0^\circ$  and  $40^\circ$  re-

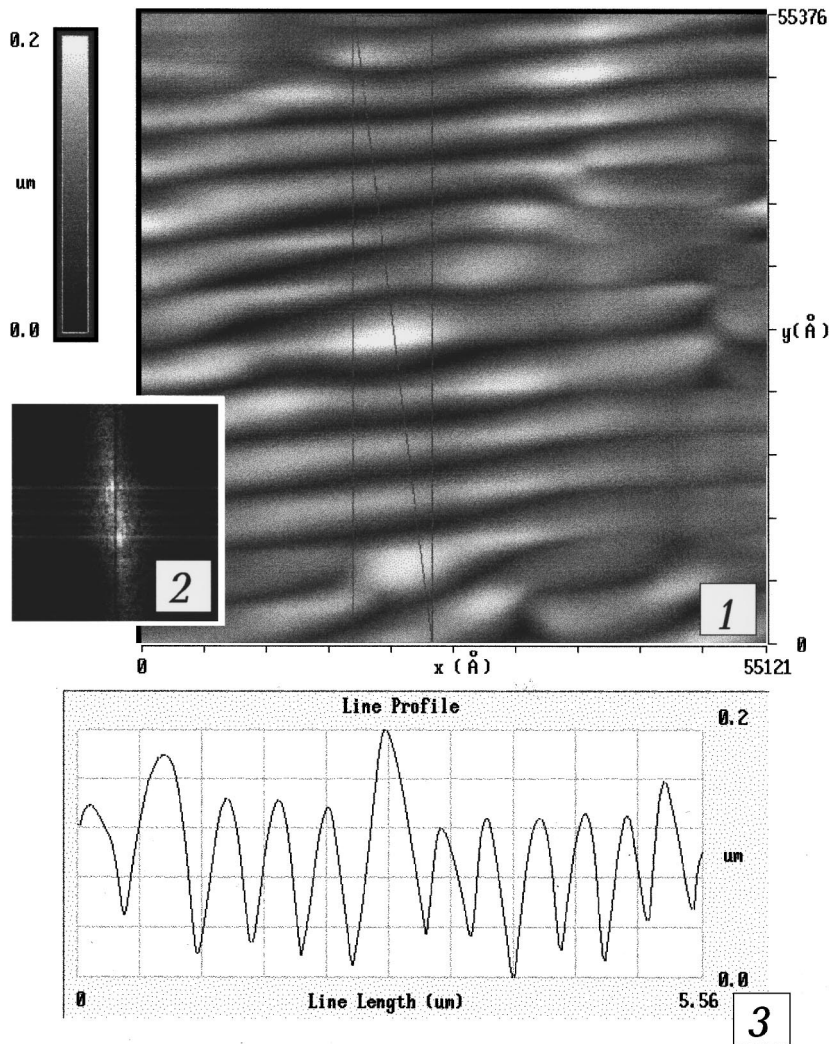


FIG. 1. Silicon implanted with  $\text{Xe}^+$ , 40 keV at  $45^\circ$ , ion influence  $1 \times 10^{18}$  ions  $\text{cm}^{-2}$  at 100 K. (1) Grayscale image. (2) Fourier transformation. (3) Cross section of the grayscale image along the line.

vealed no evidence of ripple formation for any of the experimental conditions described earlier. Figure 2 shows a typical AFM image for normal ion incidence irradiation. The behavior of the surface roughness amplitude and the wavelength  $\lambda_c$  is shown as a function of ion fluence  $\Phi$  for 40 and 20-keV irradiation at 300 K in Fig. 3. For 40-keV irradiation the mean wavelength (determined from two-dimensional Fourier transforms) remains constant over a wide fluence range, while the amplitude increases approximately linearly with fluence, until a fluence was reached at which the mean wavelength began to increase. At this stage, where the angular deviation from the mean surface is becoming larger, the quasisinusoidal structure began to transform to a corrugated and faceted structure. For 20- and 10-keV bombardment, the wavelength observed was virtually identical to that reported above, but the rates of increase of amplitude were reduced. The 10-keV data required much higher fluences (outside the scale of Fig. 3) to develop similar amplitude waves. Finally it may be noted that AFM observations of irradiated surfaces for all bombardment temperatures showed no changes in the surface morphology as a function of time after cessation of bombardment.

## DISCUSSION

In order to explain these results, we first attempted a comparison with the low-amplitude linear stochastic differential

equation description of surface evolution generalized by Carter, Nobes, and Katardjiev<sup>16</sup> following earlier deterministic description by Bradley and Harper<sup>11</sup> and more limited stochastic description by Eklund and co-workers<sup>18,19</sup> and Chason and co-workers.<sup>2,20</sup> In this approach the random arrival of ion causes local sputtering, with a yield dependent upon surface gradient and curvature which generates roughening, but this can be compensated for by surface atomic diffusion. For ion incidence at angle  $\theta$  to the surface normal on to an initially plane surface, the time dependence during bombardment of the height correlation function  $|h(q,t)|^2$  in the wave vector  $q$  domain is given by

$$|h(q,t)|^2 = \eta(q,t) R_q^{-1} [\exp(R_q t) - 1], \quad (1)$$

where

$$R_q = -\frac{\gamma}{\rho} |q| + \nu(z) a \Gamma_1(\theta) q^2 - B q^4, \quad (2)$$

$\gamma$  and  $\rho$  are the surface free energy density and viscosity, respectively,  $a$  is the mean depth of energy deposition,  $\nu(z)$  is the sputtering erosion velocity,  $\Gamma_1(\theta)$  is a calculable function of  $\theta$ ,  $B$  is proportional to the surface diffusivity, and  $\eta(q,t)$  represents the ion beam random arrival. At the end of irradiation, when the bombardment motivated roughening process ceases, any thermal-diffusion process, which also

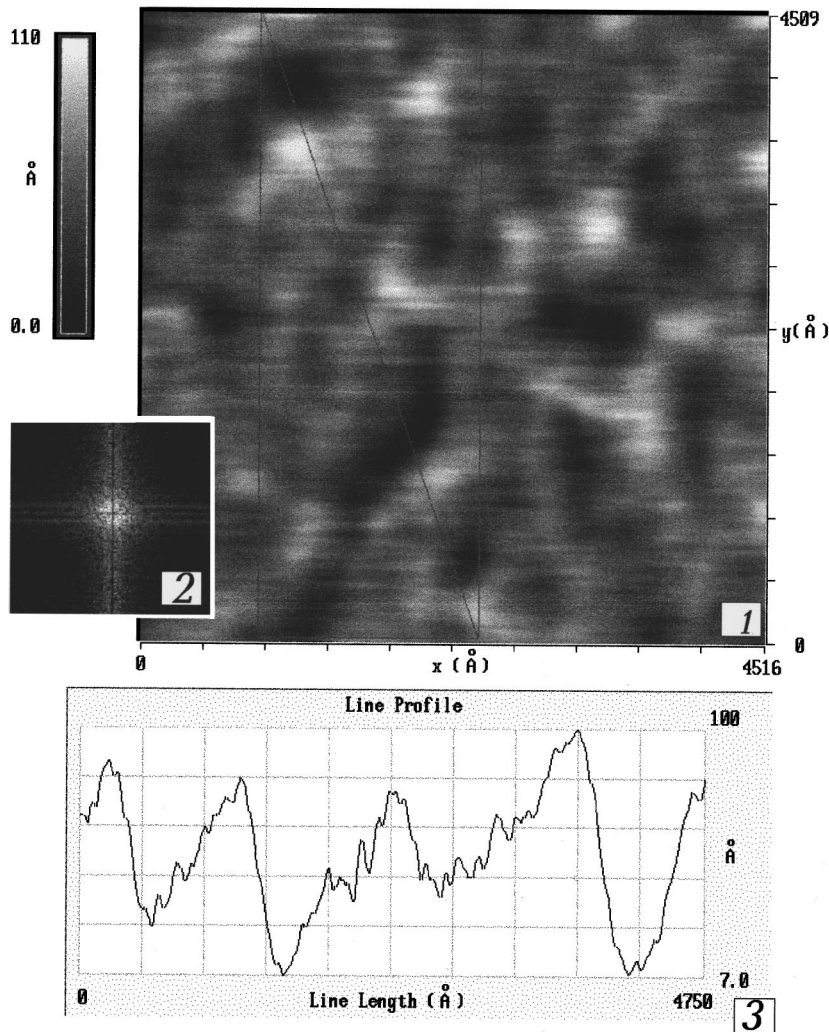


FIG. 2. Silicon implanted with  $\text{Xe}^+$ , 40 keV at  $0^\circ$ , ion fluence at 100 K. (1) Grayscale image. (2) Fourier transformation. (3) Cross section of the grayscale image along the line.

acted during irradiation, should continue, and the surface roughness should, at constant temperature, decay exponentially with time. No such relaxation was observed.

If the dominant mode of surface smoothing is via thermal surface diffusion, then Eq. (2) predicts that waves of vectors  $q_c = [\nu(z)a\Gamma_1(\theta)/2B]^{1/2}$  should develop preferably and grow exponentially in amplitude. Although  $\Gamma_1(\theta)$  is not too sensitive to ion energy,  $a$  will increase with ion energy, while the surface diffusion term should change rather rapidly with temperature. Neither of these behaviors were observed in the  $\gamma_c$  behavior in the present study. Further, parameter  $\Gamma_1(\theta)$  is predicted [Eq. (10)] to be relatively insensitive to  $\theta$  and, although periodic structure should not develop for normal incidence, features with a correlation length of the order of  $\gamma_c (= 2\pi q_c^{-1})$  should grow exponentially, which was not observed. As already indicated, it is certain that the near surface of the Si would be completely amorphized in the present studies and, in view of the low temperatures employed, although it is unlikely that surface diffusion would dominate, radiation-induced viscous flow as discussed by Volkert and Polman<sup>21</sup> may very well be important. If this is the case, then Eq. (2) reveals that only a saturation roughening (or indeed a smoothing of initial roughness) should occur with no dominant wave vector. Again this behavior is not observed in the present study. It is therefore concluded that

curvature-dependent sputtering, competing with a thermally driven surface relaxation process, cannot explain the results described here.

Gutzmann, Klaumunzer, and Schumacher<sup>22</sup> observed ripple growth and motion on 200–300-MeV  $\text{I}^+$ - and  $\text{Xe}^+$ -irradiated metallic glass, and attributed this, mainly, to radiation-induced deformation and creep processes which are known to occur in amorphous materials,<sup>23,24</sup> and in which the irradiated target suffers continuous and irreversible compression parallel to the ion flux direction, and expansion transverse to this direction. This process is related to the effects of the, mainly, inelastic energy-loss process suffered by such energetic ions, and no evidence has been found<sup>24</sup> for similar deformation when elastic energy-loss processes dominate as in the present studies. It is therefore unlikely that the same creep process can be responsible for the current observation but the following variant is plausible.

In addition to depositing energy to target atoms ion bombardment transfers momentum. Very little literature has concentrated on this topic, but Littmark and Sigmund<sup>25</sup> showed that, as an ion penetrates a target, a resolved component of the momentum gained by target atoms is, close to the surface, antiparallel to the direction of ion penetration (this correlates with the sputtering process), but for deeper penetration becomes parallel to the ion direction, increases to a

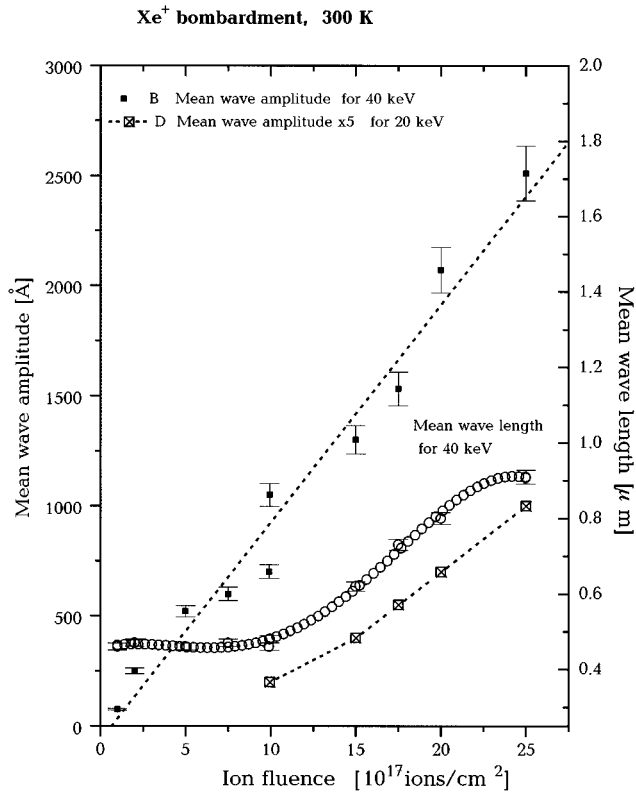


FIG. 3. Silicon implanted with Xe<sup>+</sup> at 45° and at 300 K. Mean peak-to-peak wave amplitude and mean wavelength dependencies upon ion fluence.

maximum, and falls to zero. For ions incident obliquely on a surface, this implies a component of momentum resolved parallel to the surface and a component of momentum resolved normal to the surface. For normal incidence the *mean* transverse momentum will be zero, but atoms will be displaced both parallel and transverse to the incident ion direction, constituting an effective ballistic diffusivity.<sup>17</sup> The transverse momentum for non-normal incidence will constitute an effective atomic drift parallel to the local surface. Both these processes were suggested in a qualitative manner, in a generalized stochastic description of surface evolution in Ref. 16.

Considering, first, the effective ballistic diffusivity resulting from energy deposition and atomic displacement, simplified estimates by Andersen<sup>17</sup> suggest that the effective diffusivity, proportional to  $B_{\text{eff}}$ , should scale approximately linearly with ion energy, and be insensitive to target temperature. In fact the overall parameter  $a\Gamma_1(\theta)/B_{\text{eff}}$  will be relatively insensitive to ion energy and so, as described earlier, the observed ripple wavelength should be relatively independent of incident ion energy and target temperature. Moreover, the rate of increase of ripple amplitude,  $R_q$ , at the selected wave vector is dependent not only upon  $a$ ,  $\Gamma_1(\theta)$ , and  $B_{\text{eff}}$ , but also upon  $\nu^2(z)$ . Even in the combination of the former parameters is only weakly dependent upon ion energy (and will in fact increase with ion energy), the surface erosion velocity  $\nu(z)$  certainly increases with ion energy in the range investigated. Consequently a ripple growth rate is expected, as observed, to increase with ion energy. It is

therefore suggested that, in the parameter range investigated here, collisionally motivated atomic displacement and effective diffusivity can be the dominant surface relaxation mechanism, although, at higher temperatures, thermal diffusion may assume an increasing importance. Further studies over a wider temperature range, and with different ion flux densities, are in progress to investigate any transitional behavior. Ion flux density should be an important parameter since, from Eq. (2), if the diffusion process is of ballistic origin, the ratio  $\nu(z): B_{\text{eff}}$  should be ion flux density independent and, consequently, so should  $q_c$  and  $\lambda_c$  whereas the ripple growth rate should increase linearly with ion flux density rather than the square law dependence found for 1-keV Xe<sup>+</sup>-bombarded Ge at higher temperatures, where thermal surface diffusion was proposed to be the dominant relaxation mechanism.<sup>2</sup>

Although ballistic diffusivity can account for the temperature insensitivity of ripple production it cannot, alone, explain the lack of ripple or correlated feature roughening for nearer-normal-incidence ion irradiation. However, the second ballistic displacement effect outlined earlier, atomic drift, can provide an explanation. In order to demonstrate this effect, the stochastic differential equation which leads to Eqs. (1) and (2) is reexamined and extended.

For analytic simplicity the 1+1 dimension systems only is considered here with an ion flux of mean value  $J$  incident at angle  $\theta$  in the  $xOz$  plane to the normal to the mean ( $Ox$ ) direction of a gently undulating surface (i.e.,  $\partial h/\partial x$  is everywhere small, where  $h$  is the local height). The sputtering yield (number of atoms sputtered per incident ion)  $Y(\phi, \kappa)$ , is taken to be a function of the angle of incidence,  $\phi$ , to the local surface normal<sup>26</sup> and to the local curvature  $\kappa$ ,<sup>10-12</sup> and the solid atomic density is  $N$ . Bradley and Harper<sup>10</sup> showed that, due to sputtering erosion alone, the deterministic defining equation for  $h(x, t)$  can be written

$$-\frac{\partial h}{\partial t} = \frac{J}{N} Y_0(\theta) - \frac{J}{N} \frac{\partial}{\partial \theta} [Y_0(\theta) \cos \theta] \frac{\partial h}{\partial x} + \frac{Ja}{N} Y_0(\theta) \Gamma_1(\theta) \frac{\partial^2 h}{\partial x^2}, \quad (3)$$

where  $Y_0(\theta)$  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  $\theta$ , and the standard deviations  $\alpha$  and  $\beta$  of the bi-Gaussian ellipsoidal ion energy spatial deposition density function. For an order-of-magnitude estimation the ellipsoidal distribution may be approximated by a spherical distribution with  $a = \alpha = \beta$ , in which case

$$\Gamma_1(\theta) = \sin^2 \theta - (\cos^2 \theta / 2)(1 + \sin^2 \theta). \quad (4)$$

If, now, the stochastic nature of bombardment and sputtering, surface relaxation processes and any atomic drift parallel to the surface are considered, then Eq. (3) becomes

$$\begin{aligned}
-\frac{\partial h}{\partial t} = & \frac{J}{N} Y_0(\theta) - \frac{J}{N} \frac{\partial}{\partial \theta} [Y_0(\theta) \cos \theta] \frac{\partial h}{\partial x} + \frac{\gamma}{\eta} \left| \frac{\partial h}{\partial x} \right| \\
& - \frac{Ja}{N} Y_0(\theta) \Gamma_1(\theta) \frac{\partial^2 h}{\partial x^2} + \frac{1}{N} \frac{\partial}{\partial x} F(s) + \frac{B \partial^4 h}{\partial x^4} \\
& + \eta(x, t). \tag{5}
\end{aligned}$$

The term  $\gamma/\eta |\partial h/\partial x|$  has been suggested by Chason *et al.*<sup>2</sup> to account for viscous relaxation effects ( $\gamma$  is identically equal to the surface free energy density, and  $\eta$  to the viscosity). As discussed earlier,  $B$  has been associated with thermal surface diffusion<sup>2,5-10,13,14</sup> or, as in the preceding analysis, with radiation assisted or ballistic effective diffusion.<sup>8</sup> Although the bombardment-sputtering system is essentially one of multiplicative noise, Lauritsen, Cuerno, and Makse<sup>8</sup> have shown that this can be accurately approximated by the summative noise term  $\eta(x, t)$  with zero mean and mean square equal to the mean ion flux. The term  $(1/N)[\partial F(s)/\partial x]$  describes the effect of the gradient of any atomic flux  $F(s)$  parallel to the local surface on erosion, whereas the other sputtering-related terms are consequences of atomic fluxes normal to the local surface.

Equation (5) can be further supplemented<sup>5-9</sup> by addition of a nonlinear term  $(\lambda_x/2)(\partial h/\partial x)^2$  to account for the higher-order variation of  $Y_0(\theta)$  and somewhat arbitrarily,<sup>6</sup> but in analogy with deposition and growth models,<sup>27</sup> by addition of a smoothing factor  $\nu_x(\partial^2 h/\partial x^2)$ . It will be shown shortly that this is, in fact, the form of the surface parallel atomic drift gradient component. From the solution of a generalized three-dimensional deterministic Eq. (3), as indicated in Eq. (2), which also ignored the surface parallel flux  $F(s)$ , Bradley and Harper<sup>10</sup> demonstrated, by evaluating  $\Gamma(\theta)$ , that ripple production with wave vector  $q_c$  parallel to the projection of the ion flux on to the surface would occur for small  $\theta$ , but, for larger  $\theta$ , the wave vector would be transverse to this projection. These coherent patterns result from the competition between curvature-dependent sputtering, which roughens the surface and smoothing terms. The value of  $q_c$  was predicted<sup>10</sup> to be almost independent of  $\theta$  for  $0 > \theta > 3\pi/8$ . For normal (and near normal) incidence no preferred ripple direction is expected, but the roughness should still increase with a dominant correlation length similar to  $q_c$ . Cuerno and co-workers<sup>5-8</sup> have examined the dynamics and scaling properties of Eq. (5), including the nonlinear term, assuming  $\nu_x$  to be independent of  $\theta$ , and excluding both the first-order relaxation term and the surface parallel flux term, and indeed showed that ripple formation can be predicted with the wave vector described earlier. For early times the smoothing term reduces the rate of random roughening while, for late times, the ripple structure dissociates, and nonlinear effects determine the evolution; however, these are not initially influential. Consequently, even if, for some unclear reason, the estimations of  $\Gamma_1(\theta)$  and  $\Gamma_2(\theta)$  by Bradley and Harper,<sup>10</sup> assumed to be valid by other investigators,<sup>5-8</sup> are erroneous, these will only change the observed habit of any coherent surface pattern, but not eliminate the evolution of correlated features. It therefore seems necessary to invoke a further surface smoothing mechanism which dominates for near-normal ion incidence conditions.

During the penetration and slowing down, generally by

elastic collision processes, of a relatively low-energy ion into a solid, a cascade of moving atoms is generated, and these slow down to rest. Those atoms with outwardly directed momentum and sufficient energy to surmount a surface energy barrier are sputtered, but those with forward momentum may be permanently displaced, particularly in an amorphous solid. Additionally, those atoms ejected from the surface with too low an energy to escape the energy barrier will translate parallel to the surface, and be recaptured. For ions incident obliquely ( $\theta > 0$ ) onto a surface, this is equivalent to generating a component of atomic momentum parallel to the surface in the direction of the forward projection of the ion flux on to the surface, as demonstrated by Littmark and Sigmund.<sup>25</sup> Although an exact calculation of the net effective surface parallel atomic flux is possible using the calculation scheme of Littmark and Sigmund,<sup>25</sup> an estimate can be obtained by simplistic arguments.

Each incident ion generates  $f(E) = k(E)/2E_d$  atomic recoils,<sup>28</sup> where  $k(E)$  is the fraction of energy deposited in elastic collisions, and  $E_d$  is a displacement energy. The recoils will be anisotropically distributed, with favored motion parallel to the initial ion direction, and travel, on average, a small distance  $d$  of the order of a few interatomic distances,<sup>17</sup> either across the surface plane or below the surface plane. A fraction (order  $\frac{1}{6}$ ) will travel such a distance parallel to the ion flux direction or a distance  $d \sin(\theta - \gamma)$  parallel to the surface. The total atomic flux parallel to the local surface is thus given by  $F(s) = J \cos(\theta - \gamma) f(E) d \sin(\theta - \gamma)$ , where  $\gamma$  is the local angle between the surface normal and the mean surface normal, and is equal to the local gradient [ $\gamma = \tan^{-1}(\partial h/\partial x)$ ] of the surface. In the same, first-order, expansion as in Eq. (3), with  $\gamma \approx \partial h/\partial x$ ,  $\partial F(s)/\partial s \approx \partial F(s)/\partial x$ , and  $\partial h/\partial t \equiv (1/N)[\partial F(s)/\partial x](1/\cos \gamma)$ , the gradient of this flux gives rise to a smoothing term  $(1/N)[\partial F(s)/\partial x] = (J/N)f(E)d(\partial/\partial \theta)(\sin \theta \cos \theta)(\partial^2 h/\partial x^2)$ . Combining this smoothing term with the curvature-dependent sputtering term of Eq. (3) and the approximation of Eq. (4) leads to a net destabilizing term

$$\begin{aligned}
-\frac{J}{N} \left\{ f(E)d \cos 2\theta - Y_0(\theta)a \left[ \sin^2 - \frac{\cos^2}{2}(1 + \sin^2 \theta) \right] \right\} \frac{\partial^2 h}{\partial x^2}. \tag{6}
\end{aligned}$$

In this identity the number of recoils generated,  $f(E)$ , will generally exceed the sputtering yield  $Y(\theta)$  by two orders of magnitude or more, whereas the cascade dimension  $a$  will similarly exceed the mean recoil displacement distance  $d$ . It is therefore probable that the coefficients of the angular-dependent terms will be similar. Consequently, for normal ( $\theta = 0$ ) and near-normal incidence, the destabilizing term is negative, and smoothing dominates roughening at all wave vectors. As  $\theta$  increases, the roughening term will assume an increasing importance, and for some critical  $\theta_c$  roughening will dominate at all wave vectors. The exact value of  $\theta_c$  can be determined from more exact calculations of  $F(s)$ , which are currently in progress. This analysis clearly reveals the angle,  $\theta$ , dependence of the net smoothing (or roughening) coefficient  $\nu_x$  introduced by Cuerno *et al.*<sup>6</sup>

The above scenario explains why roughening appears to be suppressed for near-normal ion incidence on to Si, as demonstrated in the present and earlier works,<sup>4</sup> although ex-

perimental data are not yet available to compare the dynamics of the interface width evolution in this case with the theoretical predictions of Eq. (5) and the added smoothing term as evaluated by Cuerno *et al.*<sup>6</sup> The approach also explains how, when  $\theta$  is increased, curvature-dependent roughening can compensate for the surface parallel atomic drift smoothing process, and enable ripple evolution. The selected wave vector will be given, in the linear form of Eq. (5) by the square root of the ratio of the net coefficient of the roughening term to the coefficient of the relaxation term. This will depend upon the mechanism assumed for surface relaxation which will dictate the dependence of the coefficient  $B$ , and hence the selected ripple wave vector and amplification rate of the ripples,<sup>2</sup> upon ion flux and temperature, as already discussed. It should be noted that if only the first-order relaxation term in Eq. (5) is operative, then no ripple evolution can occur.

An interesting consequence of Eqs. (3) and (5) is that ripples are predicted to translate in the  $Ox$  direction with a velocity  $-(J/N)(\partial/\partial\theta)[Y_0(\theta)\cos\theta]$  as discussed by some authors.<sup>10,11</sup> Recent attempts<sup>29</sup> to observe such a translation have demonstrated their speed to be vanishingly small. However, it is known<sup>26</sup> that  $Y_0(\theta)$  behaves approximately as  $Y_0(\theta)\sec\theta$  for  $\theta$  as large as  $45^\circ$ , and so the predicted velocity should, as observed, approach zero.

Finally, as shown by Cuerno *et al.*<sup>6</sup> for slowly undulating surfaces, the first-order nonlinear additive term related to the gradient dependence of  $Y(\theta)$  can mitigate the roughening process but not fully compensate for it. In the linearized equations (3) and (5), the surface gradients remain continuous, whereas, when the detailed evolution of a surface is considered using Huygens's principle deterministic approaches,<sup>30</sup> gradient discontinuities are generated by the inward collapse of peaks on the surface. In this case, following reasoning similar to that of Tang, Alexander, and Bruinsma<sup>31</sup> for surface growth, Carter, Nobes, and Katardjiev<sup>32</sup> showed that any initial sinusoidal surface rapidly transformed to a cusp and trough structure (the cusps corresponding to peaks) with the same wavelength as the initial sinusoid. The cusp-trough base height scales approximately as  $[\lambda^2 N/8JY_0(0)]t^{-1}$ , where  $\lambda$  is the initial sinusoid wavelength. Identifying this height with the interface width of correlated features of wave vector  $q=(2\pi/\lambda)$  indicates that features of wave vector  $q$  smooth at a rate proportional to  $q^{-2}t^{-2}$ , i.e., long-wavelength features smooth more rapidly than do short-wavelength features. This, large gradient, nonlinear effect acts in a similar manner to the other relaxation processes discussed earlier, except that smoothing is dominant at larger, rather than shorter, wavelengths. Moreover, the other roughening and smoothing processes discussed earlier give rise to an exponentially increasing interface width<sup>2,18</sup> with time as compared to this  $t^{-1}$  smoothing, and so, as concluded by Cuerno *et al.*,<sup>6</sup> no steady-state rough morphology might be expected as a result of this process. For non-normal ion incidence, however, shadowing processes may assume importance, and the surface could become faceted<sup>4</sup> rather than increasingly quasirandomly rough as suggested by Cuerno *et al.*<sup>6</sup> Both the present work and recent studies<sup>33</sup> with a range of different ion species and energies bombarding Si targets have demonstrated that such a faceting transition does indeed occur as ripple amplitude

increases. Unlike the processes discussed previously, which are local, ion flux shadowing is a more distant process, and is related to extensive topography. Similar distant effects include redeposition of sputtered atomic flux from points of origin to other surface regions and scattered ion flux, which can enhance the incident external flux locally.<sup>34</sup> These processes will be relevant for normal as well as oblique ion incidence, and will become increasingly important as the ion incidence angle  $\alpha$  to the local surface normal increases. For normal incidence,  $\alpha \geq 60^\circ - 70^\circ$  might be anticipated to be critical, which, in real space, determines the maximum amplitude: wavelength ratio [i.e., in Fourier space the  $h(q,t)q$  product] of features above which distance effects become important. For oblique incidence ( $\theta > 0$ ) this amplitude: wavelength ratio will be smaller.

Consequently, although the first-order nonlinear correction to Eq. (5) of the form suggested by Cuerno *et al.*<sup>6</sup> may well describe the ripple to roughening transition, it should be expected that higher-order nonlinear processes, which represent the distant effects described above, will dominate the late stages of dynamic evolution of the surface. For initial evolution stages, their influence will be negligible.

## CONCLUSIONS

In conclusion, it has been demonstrated that the gradient of the atomic flux parallel to the surface generated by ion bombardment can compensate for the curvature-dependent sputtering process, and lead to net smoothing particularly for normal incidence conditions, whereas, for larger incidence angles, roughening and ripple formation can occur. Although the authors are not aware of atomic scale experimental studies that support this conjecture, several related observations confirm its reality. First, longer flight path atomic redeposition is well confirmed experimentally,<sup>34</sup> and the shorter path events across the surface plane under the influence of the surface binding forces should also occur. Second, at higher energies than discussed here, directional macroscopic mass transport of InP parallel to the surface under the oblique ion incidence condition has been observed experimentally.<sup>35</sup> Third, present studies have fully confirmed that no ripple production occurs for  $0 < \theta < 40^\circ$  for  $\text{Xe}^+$  ion-irradiated Si with energies from 10 to 40 keV and, indeed no correlated features are observed. Finally present studies of  $\text{Xe}^+$ -implanted Si have revealed that the ripple wave vector  $q_c$  is relatively insensitive to both ion energy and target temperature, and that following the end of irradiation no relaxation of the surface occurs. The only plausible explanation of such observations is that the diffusive smoothing term in Eq. (5) is of ballistic rather than thermal origin and, in the absence of irradiation, no smoothing occurs. As indicated in our earlier work,<sup>16</sup> irradiation can generate isotropic ballistic recoil processes which are equivalent to diffusion, in addition to the directed process discussed here. The sources of these additional terms appear to be well founded. It therefore appears that the present experimental observations can be explained, qualitatively, on the basis of ballistic atomic transport processes, and the authors are currently engaged in extending the ion energy range and species together with a computation of surface atomic fluxes in order to test the model proposals more quantitatively.

- <sup>1</sup>M. J. Nobes, G. W. Lewis, G. Carter, and J. L. Whitton, *Nucl. Instrum. Methods* **170**, 363 (1980).
- <sup>2</sup>E. Chason, T. M. Mayer, B. K. Kellerman, D. T. McIlroy, and A. J. Howard, *Phys. Rev. Lett.* **72**, 3040 (1994).
- <sup>3</sup>G. Carter, M. J. Nobes, C. Cave, and N. Al-Quadi, *Vacuum* **45**, 71 (1994).
- <sup>4</sup>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.
- <sup>5</sup>R. Cuerno and A.-L. Barabasi, *Phys. Rev. Lett.* **74**, 4746 (1995).
- <sup>6</sup>R. Cuerno, H. A. Makse, S. Tomassone, S. T. Harrington, and H. E. Stanley, *Phys. Rev. Lett.* **75**, 4464 (1995).
- <sup>7</sup>R. Cuerno and K. B. Lauritsen, *Phys. Rev. E* **52**, 4853 (1995).
- <sup>8</sup>K. B. Lauritsen, R. Cuerno, and H. A. Makse, private communication (unpublished).
- <sup>9</sup>M. Rost and J. Krug, *Phys. Rev. Lett.* **75**, 3894 (1995).
- <sup>10</sup>R. M. Bradley and J. M. E. Harper, *J. Vac. Sci. Technol. A* **6**, 2390 (1988).
- <sup>11</sup>G. Carter, M. J. Nobes, and R. P. Webb, *J. Mater. Sci.* **16**, 2091 (1981).
- <sup>12</sup>P. Sigmund, *J. Mater. Sci.* **8**, 1545 (1973).
- <sup>13</sup>G. Carter, *J. Mater. Sci.* **11**, 1091 (1978).
- <sup>14</sup>G. Carter, J. S. Colligon, and M. J. Nobes, *Contributed Papers of the Proceedings of the VIIIth International Summer School of the Physics of Ionized Gases, Dubrovnik, Yugoslavia, 1976*, edited by B. Navinsk (J. Srefan Insr, Ljubljana, Yugoslavia, 1976), Vol. 239, p. 281.
- <sup>15</sup>G. Carter, M. J. Nobes, H. Stoere, and I. V. Katardjiev, *Surf. Interf. Anal.* **20**, 90 (1993).
- <sup>16</sup>G. Carter, M. J. Nobes, and I. V. Katardjiev, *Philos. Mag. B* **68**, 231 (1993).
- <sup>17</sup>H. H. Andersen, *Appl. Phys.* **18**, 131 (1979).
- <sup>18</sup>E. A. Eklund, R. Bruinsma, and J. Rudnick, *Phys. Rev. Lett.* **67**, 1759 (1991).
- <sup>19</sup>E. A. Eklund, E. J. Snyder, and R. S. Williams, *Surf. Sci.* **258**, 157 (1993).
- <sup>20</sup>T. M. Mayer, E. Chason, and A. J. Howard, *J. Appl. Phys.* **76**, 1333 (1994).
- <sup>21</sup>C. A. Volkert and A. Polman, in *Phase Formation and Modification by Beam-Solid Interactions*, edited by G. S. Was, L. E. Rehn, and D. Follstaedt, *MRS Symposia Proceedings No. 235* (Materials Research Society, Pittsburgh, 1992), p. 3.
- <sup>22</sup>A. Gutzmann, S. Klaumunzer, and P. Meier, *Phys. Rev. Lett.* **74**, 2256 (1995).
- <sup>23</sup>M. D. Hou, S. Klaumunzer, and G. Schumacher, *Phys. Rev. B* **41**, 1144 (1990).
- <sup>24</sup>A. Audouard, E. Balanzat, J. C. Jousset, D. Leseur, and L. Thome, *J. Phys. Condens. Matter.* **5**, 995 (1993).
- <sup>25</sup>U. Littmark and P. Sigmund, *J. Phys. D* **8**, 241 (1975); M. W. Sckerl, P. Sigmund, and M. Vicaneek, *Mat. Fys. Medd. Kgl. Dan. Vid. Selsk.* **44**, 3 (1996).
- <sup>26</sup>P. Sigmund, *Phys. Rev.* **184**, 383 (1969).
- <sup>27</sup>M. Kardar, G. Parisi, and Y. C. Zhang, *Phys. Rev. Lett.* **56**, 889 (1986).
- <sup>28</sup>P. Sigmund, *Appl. Phys. Lett.* **14**, 114 (1969).
- <sup>29</sup>G. Carter, V. Vishnyakov, and M. J. Nobes, *Nucl. Instrum. Methods Phys. Sect. B* **106**, 174 (1996).
- <sup>30</sup>I. V. Katardjiev, G. Carter, and M. J. Nobes, *J. Phys. D* **22**, 1813 (1989).
- <sup>31</sup>C. Tang, S. Alexander, and R. Bruinsma, *Phys. Rev. Lett.* **64**, 772 (1990).
- <sup>32</sup>G. Carter, M. J. Nobes, and I. V. Katardjiev, *Philos. Mag. B* **66**, 419 (1992).
- <sup>33</sup>G. Carter, V. Vishnyakov, Yu. V. Martynenko, and M. J. Nobes, *J. Appl. Phys.* **78**, 3559 (1995).
- <sup>34</sup>For a general discussion of redeposition and ion scattering processes and unevaluation of their contributions to specific isolated, nonrepetitive, surface structures, see I. H. Wilson, J. Belson, and O. Auciello, in *Ion Bombardment Modification of Surfaces, Fundamentals, and Applications*, edited by O. Auciello and R. Kelly (Elsevier, Amsterdam, 1984), Chap. 6, p. 225.
- <sup>35</sup>L. Cliche, S. Roorda, M. Chicoine, and R. A. Masut, *Phys. Rev. Lett.* **75**, 2348 (1995).