

Long-time effects in a simulation model of sputter erosion

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A simple $(2+1)$ -dimensional discrete model is introduced to study the evolution of solid surface morphologies during ion beam sputtering. The model is based on the same assumptions about the erosion process as the existing analytic theories. Due to its simple structure, simulations of the model can be performed on time scales where effects beyond the linearized theory become important. Whereas for short times we observe the formation of ripple structures in accordance with the linearized theory, we find a roughening surface for intermediate times. The long-time behavior of the model strongly depends on the surface relaxation mechanism.

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INTRODUCTION

During the last years, two features of surface morphologies created by ion beam sputtering have attracted particular attention: ripple structures on submicrometer-length scales and self-affine, rough surfaces.¹

The formation of periodic ripple structures has been observed experimentally in amorphous materials,² metallic crystals,^{3,4} and semiconductors amorphized by the ion beam.^{5,6} Ripples are typically oriented perpendicular to the projection of the ion beam in the surface plane for small angles of incidence, Θ (relative to the surface normal), whereas for larger angles Θ , the observed ripple pattern is rotated by 90° . Surfaces eroded by ion bombardment may also exhibit self-affine properties.⁷ With increasing ion fluence, a crossover from ripple structures to self-affine, rough surfaces has recently been observed experimentally.⁴

Our present understanding of these features is based upon the work of Bradley and Harper (BH),⁸ who found that Sigmund's sputtering theory⁹ implies a curvature dependence of the sputtering yield. Based on BH a continuum theory of surface evolution by sputter erosion was formulated as an anisotropic Kuramoto-Sivashinsky (KS) equation¹⁰ with additive noise.¹¹⁻¹³ However, there are strong indications from experiment^{3,14} that surface relaxation processes are also important during pattern formation. Such processes have not yet been adequately included in existing theories. Our results will show that the long-time behavior of patterns depends crucially on details of surface relaxation.

To investigate the analytic theory beyond the linearized regime, numerical integrations of the KS equation have been performed,^{15,16} which uncovered two markedly different long-time regimes, depending on the signs of the nonlinear couplings.

Computer simulations may be helpful in clarifying both the role of surface relaxation and of nonlinear effects. Two types of simulations have been performed up to now. Koponen *et al.*^{17,18} calculated collision cascades emerging from single-ion impact within the binary collision approximation. They find ripples in accordance with linearized BH theory,¹⁸ which appear both with and without additional surface relax-

ation processes. This indicates the presence of an ion-induced surface diffusion mechanism, which has also been predicted from BH.¹³ The simulations did not yet reach time scales where the nonlinear effects of the continuum theories could be analyzed. On the other hand, scaling properties of the roughness of ion-irradiated surfaces have also been investigated within this approach.¹⁷ In a different approach, Cuerno *et al.*¹⁹ proposed a simple, discrete stochastic model with an update rule, which incorporates the Θ dependence of the sputtering yield and a simple curvature dependence of the erosion probability *ad hoc*. Within this model, it is possible to study the crossover from ripples to rough surfaces during the evolution of an irradiated one-dimensional system.

In the present paper, we introduce a simple, yet atomistic Monte Carlo (MC) model on a lattice, which includes the same assumptions on energy deposition from ion impact as BH and we study the evolution of surface morphologies beyond the linearized regime for two different types of surface relaxation mechanisms.

MODEL

We model energy deposition by directly implementing the result of Sigmund's sputtering theory,⁹ upon which BH and the existing analytical theories are based. The surface is described as a two-dimensional field of discrete time-dependent height variables $h(x,y,t)$ on a square lattice of size $L \times L$. Here t denotes the time, measured in terms of ion fluence. In our results, we also indicate the corresponding amount of eroded material in terms of eroded monolayers (ML). Periodic boundary conditions are assumed, i.e., $h(x+L,y,t) = h(x,y,t) = h(x,y+L,t)$. Initially the surface is flat, i.e., $h(x,y,0) = h_0$. Two processes take place: erosion and surface relaxation. For each erosion step an ion is started at a random position in $[0,L] \times [0,L]$ above the surface and moved towards the surface with incidence angle Θ and an angle ϕ with respect to the x axis. After the ion has penetrated a distance a under the surface it stops and distributes its energy. A particle at the surface obtains the energy given by Sigmund's theory⁹:

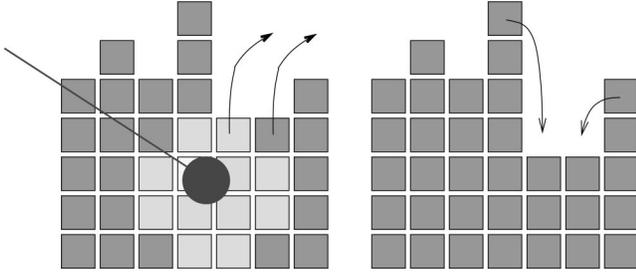


FIG. 1. The model consists of a square field of discrete height variables $h(x,y)$, corresponding to piles of $h(x,y)$ particles at position (x,y) . *Left*: each ion impact is modeled by a distribution describing the energy deposited by the ion. Atoms on the surface are removed with a probability proportional to the energy. *Right*: surface diffusion, by decreasing height differences the energy is decreased.

$$E(x',y',z') = \frac{\epsilon}{(2\pi)^{3/2}\sigma\mu^2} \exp\left(-\frac{(z'+a)^2}{2\sigma^2} - \frac{x'^2+y'^2}{2\mu^2}\right), \quad (1)$$

where (x',y',z') is the position of a particle in a local Cartesian coordinate system of the ion, where the z' axis coincides with the ion trajectory. A particle at point $\mathbf{r}=(x,y,h)$ on the surface is removed, i.e., the height variable decreased by 1, with a probability proportional to $E(\mathbf{r})$ (see Fig. 1).

For surface relaxation, we use either activated surface diffusion (ASD) or Wolf-Villain (WV) type irreversible relaxation as simplified models of high-temperature and low-temperature relaxation mechanisms. ASD is implemented as a nearest-neighbor MC hopping process with an energy $E=(J/2)\sum_{\langle i,j \rangle}[h(i)-h(j)]^2$ as in Ref. 20 (see also right part of Fig. 1) and WV relaxation is implemented according to Ref. 21. For ASD a MC step is performed every $0.001L^2$ erosion steps with temperature $k_b T/J=0.2$. The irreversible WV process is completed for all surface atoms after every ion impact. System sizes up to $L=256$ are considered. To avoid anisotropies, which may be induced by the square lattice, we have chosen the azimuthal angle $\phi=22^\circ$. The angle Θ was varied between 0° and 80° .

We use parameters $a=5.4$, $\sigma=3.3$, and $\mu=1.7$ and $\epsilon=(2\pi)^{3/2}\sigma\mu^2$ [which makes the prefactor in Eq. (1) equal to 1]. One should keep in mind that our choice of ϵ leads to rather high sputtering yields $Y(Y\approx 7.0)$ compared to experiments like Ref. 4, where $Y=0.3, \dots, 0.5$. According to BH, the ripple wavelength λ scales like $\lambda\propto Y^{-1/2}$ so that in our simulations we expect patterns with correspondingly smaller length scales.

RESULTS

The model perfectly reproduces the Θ dependence of the sputtering yield of BH (Ref. 22) and thus should not be used for too large Θ . Wavelength ratios obtained from simulations [$\lambda(45^\circ)/\lambda(30^\circ)=0.89(10)$, $\lambda(60^\circ)/\lambda(30^\circ)=0.84(10)$, $\lambda(70^\circ)/\lambda(30^\circ)=0.89(10)$] agree reasonably well with linear BH theory and with experiment.⁴

Next, we present results on the evolution of the morphol-

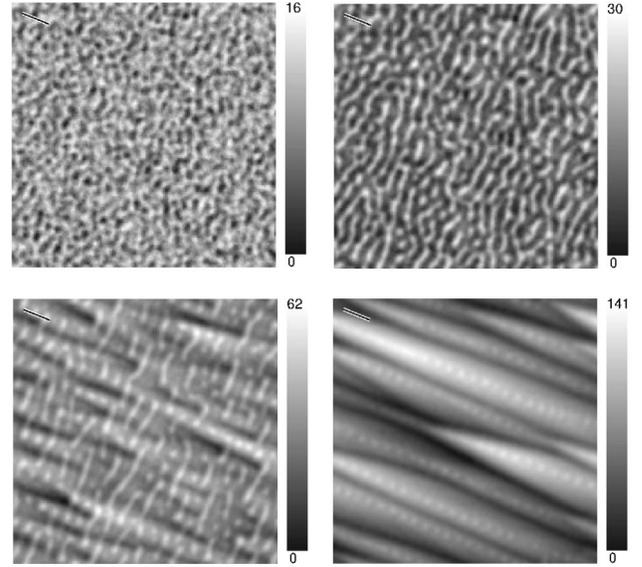


FIG. 2. Surface of a 256×256 system after t ions have hit the system. $\Theta=50^\circ$. Upper row left: $t=2.7\times 10^4$ (corresponding to 3 ML of particles been eroded). Upper row right: $t=2.5\times 10^5$ (30 ML). Lower row left: $t=2.6\times 10^6$ (300 ML). Lower row right: $t=9.3\times 10^7$ (10^4 ML). The bars indicate the direction of the ion beam.

ogy of a surface relaxed by ASD to discuss the crossover between different time regimes. Afterwards we will show that the morphologies reached with WV relaxation are completely different.

For ASD, real-space pictures of the height profiles of a 256×256 surface irradiated with ions under $\Theta=50^\circ$ are shown in Fig. 2 for increasing ion fluences. Figure 3 shows structure factors $S(\mathbf{k})=|\hat{h}(\mathbf{k})|^2$ for wave vectors k_{\parallel} parallel and k_{\perp} perpendicular to the projection of the direction of the ion beam onto the flat surface. Here $\hat{h}(\mathbf{k})$ is the Fourier transform of height fluctuations $[h(x,y)-\bar{h}]$ [$\bar{h}=\sum_{x,y}h(x,y)/L^2$ being the average height]. First signs of a structure appear after 1 ML of eroded material. For short times, after 3 ML have been sputtered ($t=2.7\times 10^4$ ions), a pattern with a typical length scale of 7 lattice spacings can be seen in the real space pictures. After 30 ML have been eroded ($t=2.5\times 10^5$) ripples oriented perpendicular to the direction of the ion beam are visible and maxima in the corresponding structure factor appear. For intermediate times ($t=2.6\times 10^6$ corresponding to 300 ML), the ripples start to disappear again. After long times ($t=9.3\times 10^7$ or 10^4 ML) the morphology approaches a rough surface with anisotropic statistics of height fluctuations. In the structure factor, the peak indicating the ripples has strongly decreased and $S(k_{\parallel})$ approaches a scaling behavior, $S(k_{\parallel})\sim k_{\parallel}^{-2.83}$. A similar crossover from ripples to a rough surface is also seen in experimentally obtained structure factors.⁴ Note that we also have tested that when switching of the sputtering mechanism, the structure factor obtained from pure surface diffusion follows at long times the usual $S(k)\sim k^{-2}$ behavior.¹

The different time regimes are seen most clearly in the time evolution of the roughness $W^2(t)=1/L^2\sum_{x,y}[h(x,y,t)$

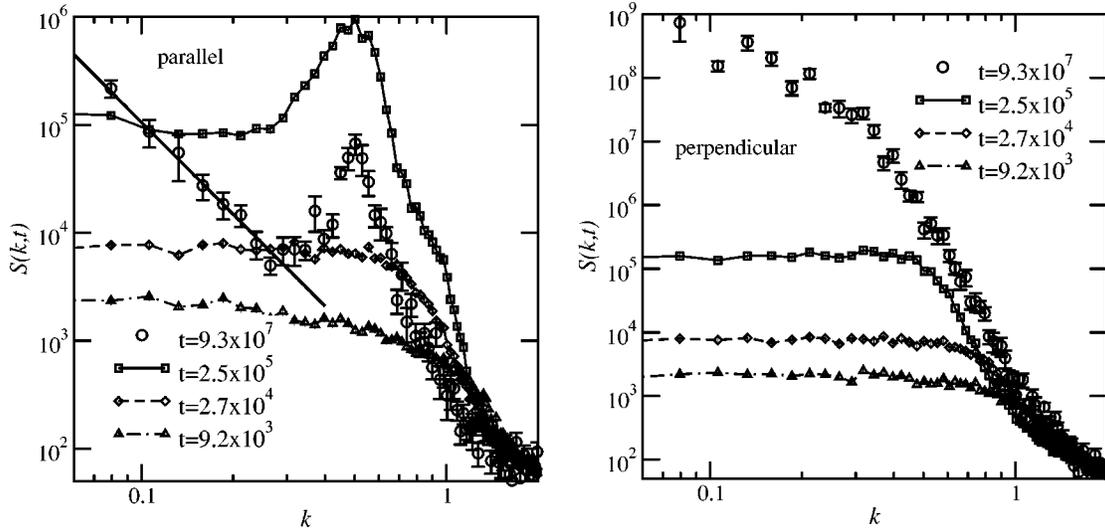


FIG. 3. Structure factor $S(k_{\parallel})$ (upper panel) and $S(k_{\perp})$ (lower panel) for different ion fluences and for $\Theta = 50^\circ$. The solid line in the upper panel represents a $k^{-2.83}$ behavior. The data for the largest ion fluence are an average over ten independent runs, while the other data points are from 300 runs.

$-\bar{h}(t)]^2$ (see Fig. 4). For short times the data exhibit a positive curvature. During that period the ripple structure is formed. The crossover to intermediate times appears rather abruptly in accordance with numerical solutions of the noisy KS equation.¹⁶ A fit with an algebraic growth law $W \sim t^\beta$ for intermediate times reveals $\beta = 0.168(2)$. For longest times, a strong increase in W is observed. We assume that the end of the algebraic regime is marked by finite size effects, which remain to be studied in detail.

For WV relaxation and for short times, we again find ripples which behave in accordance with linear BH theory. The long-time morphologies, however, are completely different from ASD relaxed surfaces as can be seen in Fig. 5. Generally, WV produces stable patterns of pronounced ripples up to the longest times simulated. For $\Theta = 50^\circ$ the ripples are tilted by $\approx 17^\circ$ against the direction perpendicular

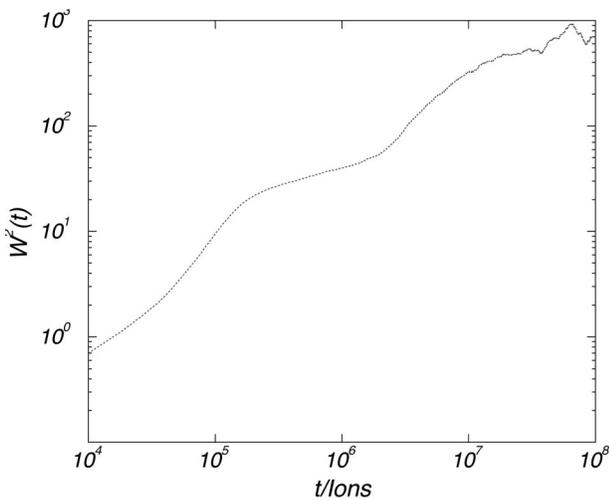


FIG. 4. Surface roughness W^2 as a function of number t of ions for $\Theta = 50^\circ$.

to incidence, whereas for $\Theta = 70^\circ$ striplike domains oriented approximately parallel to incidence appear, which contain ripples with the same tilt angle of $\approx 17^\circ$.

In conclusion, we have presented a simple discrete model for (2 + 1)-dimensional surfaces subjected to ion beam sputtering. The model is based on assumptions, which also underly BH theory and continuum theories. The simplicity of the model allows us to simulate rather large systems over time scales, where nonlinear effects become apparent. We have used the model to study distinct time regimes of surface evolution under the influence of two different surface relaxation mechanisms (ASD and WV). For short times, we always find ripple formation in accordance with linear BH theory.

For longer times, the surface morphologies emerging from ASD and from WV become qualitatively different. For ASD, the ripple wavelength increases with increasing ion fluence, and scaling behavior in the structure factor appears. Ripples get blurred and the surface morphology becomes rough, but stays anisotropic. Replacing ASD by WV surface relaxation, we find pronounced ripples in the long-time regime. Note that in the earlier detailed simulations of the sputtering process,¹⁸ basically the same result was obtained even when no surface diffusion was present. It was assumed that

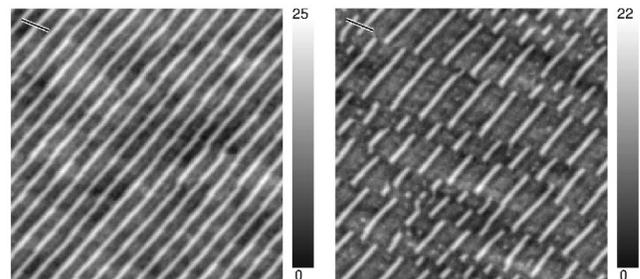


FIG. 5. Surfaces after long-time sputtering with Wolf-Villain type surface diffusion. Left: $\Theta = 50^\circ$. Right: $\Theta = 70^\circ$.

this is due to the smoothing mechanism introduced by the detailed modeling of the sputtering itself. In our simple model, we need an additional surface relaxation process to observe ripples, but we have found for short times no significant differences between the WV and ASD cases. Hence, the nonlinear long-time regime allows a much better comparison of different surface diffusion mechanisms.

WV relaxation and ASD may be considered as the simplest type of relaxation mechanisms for low temperatures and for high temperatures, respectively. Note that the aim of this work is not to evaluate the relaxation mechanisms itself, but to introduce a simple model which allows to study long-time effects and their dependence of the surface diffu-

sion mechanism. Other, more specific surface relaxation mechanisms^{23–30} involving diffusion barriers may be studied within our model using appropriate kinetic MC steps for surface relaxation, especially the model can be easily extended to describe different crystal structures and include surface anisotropies.

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