A MODEL FOR ION-SPUTTERING: FROM PATTERN FORMATION TO ROUGH SURFACES

R. CUERNO[†] H. A. MAKSE[†], S. TOMASSONE[‡], S. T. HARRINGTON[†], AND H. E. STANLEY[†]
[†] Center for Polymer Studies and Dept. of Physics, Boston University, Boston, MA 02215
[‡] Department of Physics, Northeastern University, Boston, MA 02115

ABSTRACT

Many surfaces eroded by ion-sputtering have been observed to develop morphologies which are either periodic, or rough and non-periodic. We have introduced a discrete stochastic model that allows to interpret these experimental observations within a unified framework. A simple periodic pattern characterizes the initial stages of the surface evolution, whereas the later time regime is consistent with self-affine scaling. The continuum equation describing the surface height is a noisy version of the Kuramoto-Sivashinsky equation.

INTRODUCTION

Surface erosion by ion–sputtering is a phenomenon in which a surprisingly rich variety of surface morphologies are generated, depending on experimental conditions [1]. Topography features can be found at all length scales, and from the technological point of view it is very important to understand the mechanisms by which they are formed, and thus gain control on the final morphology of the bombarded samples. On a more fundamental level, the different morphologies observed constitute interesting examples of pattern formation [2] and rough surfaces [3] in non-equilibrium phenomena, where there exists an interesting interplay between mechanisms of deterministic and stochastic natures.

Both types of morphologies (e. g. periodic surfaces and rough surfaces) have been long known to occur in the bombardment of amorphous targets at low energies. Specifically, in many cases a pattern is generated consisting of a ripple structure [4], aligned in directions either parallel to or perpendicular to that of the bombarding beam of ions. On the other hand, one also finds in similar experimental conditions that the surfaces generated are rough and thus display self-affine scaling at long distances and long times [5]. An outstanding question is then how to reconcile these observations with the formation of the periodic ripple structure.

In this work we report on the introduction [6] of a discrete stochastic model that allows to understand the formation of a periodic pattern and the kinetic roughening of the bombarded surface in an unified framework. We argue that the continuum equation describing the evolution of the model interface is a noisy version of the Kuramoto-Sivashinsky (KS) equation. The deterministic KS equation [7] appears very frequently in studies of pattern formation, and is considered as a paradigm of spatiotemporal chaos [2]. In the KS system, an initially flat surface evolves into an almost periodic morphology. At later stages, the surface roughens in a fashion consistent with the Kardar-Parisi-Zhang [8] universality class (for a one dimensional interface) [10]. In our case, the relevant continuum equation differs from the deterministic KS in the presence of a noise term taking into account the stochastic nature processes occurring in the system. Nevertheless, the qualitative behavior is identical with that just described, thus permitting to understand the formation of the ripple structure and of the rough surfaces in the ion-sputtered system as different stages of a same dynamical evolution.

PHYSICAL MECHANISMS IN ION-SPUTTERING

Here we discuss briefly the main physical mechanisms determining the evolution of an amorphous target bombarded with low energy ions (typically with kinetic energies around 1–10 keV).

As established long ago by Sigmund [11], the sputtering is induced by cascades of collisions among the atoms of the solid, triggered by the incoming ions along a finite penetration path inside the target. These collisions may eventually affect an atom living at the surface, which may be ejected leading to the sputtering event proper. The value of the average penetration depth a depends chiefly on the ion/target atom mass ratio and the energy of the ions. A typical value for a is 100 Å. On the other hand, at the energies considered, the cascades of collisions are sufficiently large to involve many atoms (the spatial extent of the cascades is also of the order of 100 Å), but sufficiently small to be accurately described by the linear cascade theory of [11], where binary collisions are supposed to take place between one atom at rest and one atom in motion. This approach describes rather succesfully the phenomenology for rates of erosion, etc. In this description, the velocity of erosion at one point of the surface can be taken to be proportional to the total amount of energy that it gains form all the collisions, thus being proportional to the amount of target solid surrounding that point. As noticed by Sigmund and later ellaborated on by Bradley and Harper [12], this leads to an instability in the system, since the bottom of troughs will be eroded *faster* than the peaks of crests. Phenomenologically, on the other hand, it is known that the velocity at which the surface recedes at a point depends also on the value of the slope of the surface there. This is a surface effect which goes beyond the approximations made in the linear cascade theory, where an infinite medium is assumed. A finite medium has the effect of a lower velocity of erosion, which is usually described by a lower sputtering yield $Y(\varphi)$, defined as the number of eroded atoms divided by the number of bombarding ions. In practice, at any given point on the surface, $Y(\varphi)$ depends on the angle of incidence φ of the ions trajectories to the normal direction at that point. The smaller φ is, the closer is $Y(\varphi)$ to the linear cascade prediction. If φ increases from zero, there's a finite maximum value for the yield, beyond which more and more ions are reflected by the surface itself, and when $\varphi = 90^0$ one has $Y(\varphi = 90^0) = 0$. For a generic point on the interface, the normal direction will not coincide with the normal to the uneroded surface, but will be determined by the value of the slope there.

There exists a physical mechanism that counterbalances the instability existing in the erosion mechanism described above. This is surface diffusion, which tends to smooth out surface features. Surface diffusion is activated by temperature, and in many cases (as typically for surfaces grown by Molecular Beam Epitaxy) can be considered as minimizing a chemical potential proportional to the surface curvature [13]. This approach is possible in a coarse grained description of the system [14], as the one we pursue here, in which the unit length is set by the finite penetration depth (or the spatial extent of the cascades), and therefore goes beyond the individual atoms.

In the above discussion, additional effects have been neglected, such as the shadowing of some surface features over others and redeposition of the eroded material. These assumptions have been shown to be consistent with the early stages of the surface evolution [4]: the late roughening is again consistent with them.

Finally, to model the sputtering phenomena, one has to take into account that in general the ion beam fluctuates in time and space. This can be described by assuming that at each time step (through which the average flux is implicitly fixed) a dynamical process takes place at a randomly chosen location along the interface.
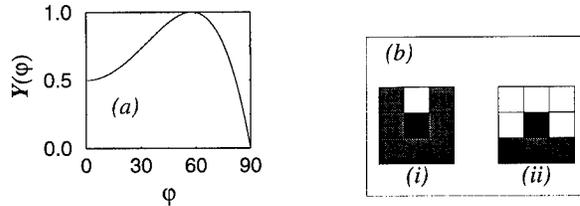
Figure 1: (a) Sputtering yield $Y(\varphi)$ as a function of the angle φ. (b) Box rule for erosion. We define p_e as the number of occupied neighboring sites (grey squares) inside the 3×3 box centered at site i (black square), normalized by 7. The examples shown correspond to (i) $p_e = 1$ and (ii) $p_e = 3/7$.

DISCRETE MODEL

To define our model, we introduce two dynamical rules, one to account for erosion and one to account for surface diffusion. The model for the case of $1 + 1$ dimensions is defined on a square lattice of lateral size L, with periodic boundary conditions in the horizontal direction. The initial interface is a horizontal line separating occupied sites (below) from empty sites (above). We choose randomly a site i at the interface where $i = 1, \ldots, L$. The chosen site is subject to erosion with probability p, or to diffusion with probability $1 - p$, where the rules are as follows:

(i) Erosion (probability p)— We compute $\varphi \equiv \tan^{-1}[(h_{i+1} - h_{i-1})/2]$, where h_i is the height of the interface at site i, and apply the erosion rule with probability $Y(\varphi)$, as given in Fig. 1a. To erode, we count the number of occupied neighbors inside a square box of size 3×3 lattice spacings centered in the chosen site i (box rule). We empty the site with an erosion probability p_e proportional to the number of occupied cells in the box (see Fig. 1b). Thus the box rule favors the erosion of troughs as compared to the peaks of crests, and therefore is the source of the instability in the ion-sputtered system.

(ii) Surface Diffusion (probability 1−p)— A diffusive move of the particle i to a nearest neighbor column is attempted with hopping probability $w_{i \to f} \equiv \left[1 + \exp\left(\Delta\mathcal{H}_{i \to f}/k_B T\right)\right]^{-1}$, where $\Delta\mathcal{H}_{i \to f}$ is the energy difference between the final and initial states of the move. Following [14], we choose $\mathcal{H} \equiv (J/2)\sum_{\langle i,j \rangle}(h_i - h_j)^2$.

The above model can be generalized in a straightforward way to the physical two dimensional case. We expect the results to be qualitatively similar to the one dimensional case discussed below. An additional ingredient in 2+1 dimensions is the anisotropy between the two substrate directions induced by the bombarding beam. This can be accounted for by imposing a finite angle of incidence, assumed in the above to be zero. For illustration, we show in Fig. 2 the result of two-dimensional simulations in which we have set $Y(\varphi) \equiv 1$. In Fig. 2a an isotropic box rule has been used, while in Fig. 2b the box does not count the number of neighbors in one of the two directions. Therefore, the surface only presents ripples with wavevector parallel to that direction in which the neighbors are counted in the corresponding box rule.

DISCUSSION

Next we consider the model with $Y(\varphi)$ shown in Fig. 1a. The results are not expected to depend strongly on the specific form of $Y(\varphi)$, so long as it preserves the existence of a maximum, and $Y(0) \neq 0$, $Y(90^0) = 0$ [15]. The initial stages of the evolution are dominated by an instability in which there exists a maximally unstable mode in the system, and the surface looks almost periodic, see Fig. 3a. Fig. 4a displays the structure factor
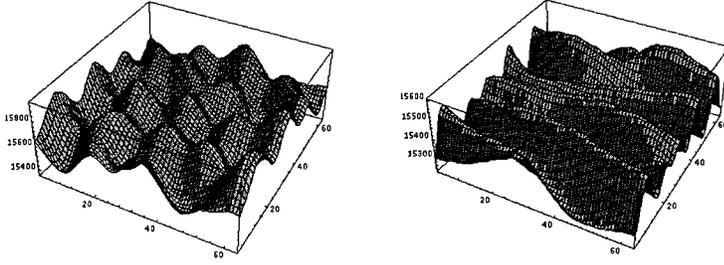
Figure 2: Two-dimensional simulations with $Y(\varphi) \equiv 1$. In Fig. 2a an isotropic box rule has been used, while in Fig. 2b the box does not count the number of neighbors in one of the 2 directions.

Figure 3: (a) Periodic surface morphology for initial stages (we have set $Y(\varphi) \equiv 1$) for $L = 50$ and $t = 1000$; (b) rough surface morphology for $L = 2048$ at late stages.

$S(\mathbf{k}) \equiv \langle \hat{h}(\mathbf{k}, t) \hat{h}(-\mathbf{k}, t) \rangle$ at the onset of the instability. Here $\hat{h}(\mathbf{k}, t)$ is the Fourier transform of $h_i(t) - \overline{h(t)}$, and $\overline{h(t)} \equiv L^{-1} \sum_{i=1}^{L} h_i(t)$. The solid line in Fig. 4a is a fit to the solution of the linear part of

$$\partial_t h(\mathbf{x}, t) = \nu \nabla^2 h - \kappa \nabla^4 h + \eta(\mathbf{x}, t) + f_Y[h(\mathbf{x}, t)], \tag{1}$$

where $h(\mathbf{x}, t)$ is the height of the interface at position $\mathbf{x}$ and time t, ν is a *negative* surface tension coefficient, κ is a *positive* coefficient that accounts for the surface diffusion, and $\eta(\mathbf{x}, t)$ is a Gaussian noise with short range correlations and strength $2D$, that accounts for the fluctuations in the ion beam. The functional $f_Y[h]$ takes into account the contribution of nonlinear terms, which appear in the equation of motion due to the effect of $Y(\varphi)$, itself a nonlinear function of the local slope $\nabla h \equiv \tan \varphi$ [16]. These nonlinear effects are triggered at later times by the large slopes built in by the instability, so that eventually the interface results in a rough morphology (Fig. 3b). In Fig. 4b, we present the time evolution of the total interface width $W(t) \equiv \langle L^{-1} \sum_{i=1}^{L} (h_i(t) - \overline{h(t)})^2 \rangle^{1/2}$ (the brackets denote an average over realizations of the noise). We observe a first scaling regime [17] $W(t) \sim t^{\beta_1}$, with $\beta_1 = 0.38 \pm 0.03$, consistent with the linear MBE equation [13, 14] (which is (1) with $\nu = f_Y \equiv 0$). This regime is followed by unstable erosion ($\beta_2 > 0.5$). For later stages, we find $\beta_3 = 0.23 \pm 0.03$, consistent with the scaling of the EW equation [18] (Eq. (1) with $\kappa = f_Y \equiv 0$, and $\nu > 0$), after which a crossover to $\beta_4 = 0.28 \pm 0.03$ is found. Finally, the width saturates due to the finite size of the system. Note that the value of the growth exponent for the KPZ equation is $\beta_{\mathrm{KPZ}} = 1/3$ [8]. As we see in Fig. 4b, there is a long crossover time from EW to KPZ behavior, responsible for the difference between β_4 and β_{KPZ}, and for the narrow window in which β_4 is observed—we find that the width of this window increases systematically with L. A similar phenomenon is well known to occur in the
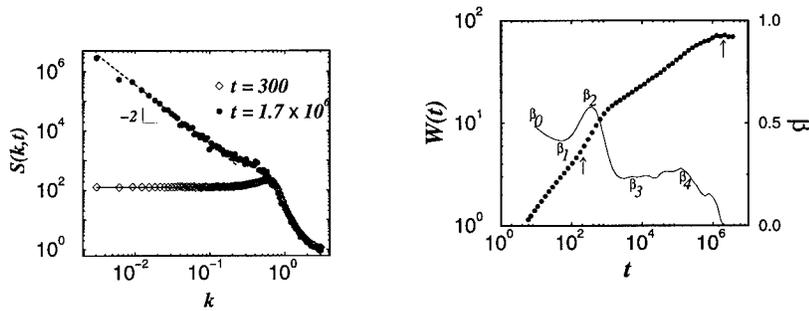
Figure 4: (a) $S(k)$ computed for a system with $L = 2048$. For $t = 300$, averaged over 2600 noise realizations ($\diamond$), and for $t = 1.7 \times 10^6$, averaged over 39 realizations ($\bullet$), see arrows in (b). The solid line is a fit to the exact solution of the discretized linear part of Eq. (1). The dashed straight line has slope -2; (b) $W(t)$ showing the regimes of the evolution for $L = 2048$. The solid line is the consecutive slope. The arrows indicate the times at which the structure factor is displayed in (a).
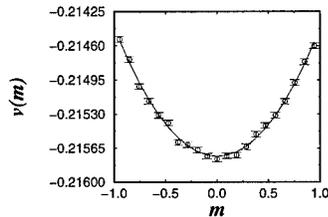

Figure 5: $v(m)$ as a function of the average tilt m of the interface, calculated in the saturated regime for $L = 128$, and averaged over 810 noise realizations. The solid line is a fit to a parabola.

deterministic KS equation in 1+1 dimensions, see Sneppen *et al.* in [10]. At saturation, $S(k)$ displays the small momenta behavior $S(k,t) \sim k^{-2}$ (Fig. 4a), consistent with the scaling of both the EW and KPZ universality classes. To determine if a KPZ nonlinearity is present in Eq. (1), we compute the mean velocity $v(m)$ of the interface in the saturated regime as a function of an average tilt $m \equiv \langle \nabla h \rangle$ imposed by using helical boundary conditions. If we assume that the relevant nonlinearity in (1) is of the KPZ type, then $f_Y[h] = (\lambda/2)(\nabla h)^2$. Taking spatial and noise averages in (1), $v = v_0 + (\lambda/2)m^2$, where v_0 is the velocity of the untilted interface [19]. The parabolic shape of $v(m)$ obtained in our simulations (see Fig. 5) leads to the conclusion that the long time and long distance behavior of the model falls into the KPZ universality class. Moreover, the continuum equation describing the model ion-sputtered surfaces is the *noisy* KS equation

$$\partial_t h = \nu \nabla^2 h - \kappa \nabla^4 h + \frac{\lambda}{2}(\nabla h)^2 + \eta(x, t). \tag{2}$$

To compare the *dynamics* of (2) with those obtained for the discrete model, we have integrated numerically Eq. (2) in $1 + 1$ dimensions [6]. We obtain the same crossover behavior for (2) as for the discrete model. Consistent with these numerical findings, the late scaling of Eq. (2) has been shown through a renormalization-group calculation [9] to

be that of the KPZ equation in 1+1 and 2+1 dimensions. Also, recently Eq. (2) has been derived for the present model through a master equation approach [20].

Finally, we compare the results of the model with observations of recent experiments. The experimental development of a ripple structure [4] is well understood in terms of the unstable linear theory of ion-sputtering describing the early stages of the time evolution of the model presented here. Moreover, the model predicts that in the late regime the large slopes generated by the unstable growth trigger the action of nonlinearities which stabilize the surface. The nonlinearity we find is of the KPZ type, consistent with the experimental observation of KPZ scaling reported by Eklund *et al.* [5]. To confirm the above picture, it would be of interest to study experimentally if both regimes do effectively take place in the time evolution of the *same* physical system.

References

[1] G. Carter *et al.*, B. Navinšek and J. L. Whitton in *Sputtering by Particle Bombardment*, Vol. II, edited by R. Behrisch, (Springer-Verlag, Heidelberg 1983), p. 231.
[2] M. C. Cross and P. C. Hohenberg, Rev. Mod. Phys. **65**, 851 (1993).
[3] P. Meakin, Phys. Rep. **235**, 189 (1993); T. Halpin-Healey and Y.-C. Zhang, *ibid.* **254**, 215 (1995); A.-L. Barabási and H. E. Stanley, *Fractal Concepts in Surface Growth* (Cambridge University Press, Cambridge 1995).
[4] E. Chason *et al.*, Phys. Rev. Lett. **72**, 3040 (1994); T. M. Mayer *et al.*, J. Appl. Phys. **76**, 1633 (1994).
[5] E. A. Eklund *et al.*, Phys. Rev. Lett. **67**, 1759 (1991); E. A. Eklund *et al.*, E. J. Snyder and R. S. Williams, Surf. Sci. **285**, 157 (1993); J. Krim *et al.*, Phys. Rev. Lett. **70**, 57 (1993); H.-N. Yang, G.-C. Wang, and T.-M. Lu, Phys. Rev. **B 50**, 7635 (1994).
[6] R. Cuerno, H. A. Makse, S. Tomassone, S. T. Harrington, and H. E. Stanley, Phys. Rev. Lett. in press.
[7] Y. Kuramoto and T. Tsuzuki, Prog. Theor. Phys. **55**, 356 (1977); G. I. Sivashinsky, Acta Astronaut. **6**, 569 (1979).
[8] M. Kardar, G. Parisi and Y.-C. Zhang, Phys. Rev. Lett. **56**, 889 (1986).
[9] R. Cuerno and K. B. Lauritsen, Phys. Rev. **E 52**, 4853 (1995). For $d > 2$, L. Golubović and R. Bruinsma, Phys. Rev. Lett. **66**, 321 (1991); *ibid.* **67**, 2747 (E) (1991).
[10] S. Zaleski, Physica **D 34**, 427 (1989); K. Sneppen *et al.* Phys. Rev. **A 46**, R7351 (1992); F. Hayot *et al.*, Phys. Rev. **E 47**, 911 (1993).
[11] P. Sigmund, Phys. Rev. **184**, 383 (1969).
[12] P. Sigmund, J. Mat. Sci. **8**, 1545 (1973); R. M. Bradley and J. M. E. Harper, J. Vac. Sci. Technol. **A 6**, 2390 (1988).
[13] C. Herring, J. Appl. Phys. **21**, 301 (1950); W. W. Mullins, J. Appl. Phys. **28**, 333 (1957); in the context of surface roughening, see D. E. Wolf and J. Villain, Europhys. Lett. **13**, 389 (1990); S. Das Sarma and P. I. Tamborenea, Phys. Rev. Lett. **66**, 325 (1991).
[14] M. Siegert and M. Plischke, Phys. Rev. **E 50**, 917 (1994).
[15] G. Carter *et al.*, Surf. Interface Anal. **20**, 90 (1993); A. N. Protsenko, Nucl. Instr. and Meth. **B 82**, 417 (1993).
[16] This argument can be made more precise, see [20].
[17] An initial random erosion regime ($\beta_0 = 0.5$) is also observed before correlations build up in the system.
[18] S. F. Edwards and D. R. Wilkinson, Proc. R. Soc. Lond. **A 381**, 17 (1982).
[19] J. Krug and H. Spohn, Phys. Rev. Lett. **64**, 2332 (1990).
[20] K. B. Lauritsen, R. Cuerno, and H. A. Makse, preprint.