Dynamic Scaling of Ion–Sputtered Surfaces

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Abstract

We derive a stochastic nonlinear equation to describe the evolution and scaling properties of surfaces eroded by ion bombardment. The coefficients appearing in the equation can be calculated explicitly in terms of the physical parameters characterizing the sputtering process. We find that transitions may take place between various scaling behaviors when experimental parameters such as the angle of incidence of the incoming ions or their average penetration depth, are varied.

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Recently there has been much interest in understanding the formation and roughening of nonequilibrium interfaces. Applications range from fluid flow in porous media to bacterial colonies or molecular beam epitaxy[[1\]](#page-9-0). A common feature of most rough interfaces observed experimentally or in discrete models is that their roughening follows simple scaling laws. The associated scaling exponents can be obtained using numerical simulations or stochastic evolution equations. The morphology and dynamics of a rough interface can be characterized by the surface width, $w(t, L)$, that scales as $w^2(t, L) = \langle [h(\mathbf{r}, t) - \overline{h}(t)]^2 \rangle = L^{2\alpha} f(t/L^z)$, where α is the roughness exponent for the interface $h(r, t)$ and the dynamic exponent z describes the scaling of the relaxation times with the system size L; $\overline{h}(t)$ is the mean height of the interface at time t and $\langle \rangle$ denotes both ensemble and space average. The scaling function f has the properties $f(u \to 0) \sim u^{2\alpha/z}$ and $f(u \to \infty) \sim \text{const.}$

Much of the attention has focused so far on the kinetics of interfaces generated in *growth* processes. However, for a class of technologically important phenomena, such as sputter etching, the surface morphology evolves as a result of *erosion* processes [\[2](#page-9-0)]. Motivated by the advances in understanding growth, recently a number of experimental studies have focused on the scaling properties of surfaces eroded by ion bombardment[[3–5](#page-9-0)]. For graphite bombarded with 5 keV Ar ions, Eklund *et al.* [\[3](#page-9-0)] reported $\alpha \simeq 0.2 - 0.4$, and $z \simeq 1.6 - 1.8$, values consistent with the predictions of the Kardar-Parisi-Zhang (KPZ) equation in $2+1$ dimensions [\[6,7](#page-9-0)]. Similarly Krim *et al.* [\[4](#page-9-0)] observed a self-affine surface generated by 5 keV Ar bombardment of an Fe sample, with a larger exponent, $\alpha = 0.52$. On the other hand, there exists ample evidence about the development of a periodic ripple structure in sputter etched surfaces (see e.g.[[8\]](#page-9-0)). Chason *et al.* [\[5](#page-9-0)] have recently studied the dynamics of such eroded surfaces for both $SiO₂$ and Ge bombarded with Xe ions at 1 keV, and found that it differs from the dynamics expected for the self-affine morphologies observed in [\[3\]](#page-9-0) and [\[4\]](#page-9-0).

In this paper we investigate the large scale properties of ion-sputtered surfaces aiming to understand in an unified framework the various dynamics and scaling behaviors of the experimentally observed surfaces. For this we derive a stochastic nonlinear equation that describes the time evolution of the surface height. The coefficients appearing in the equation are functions of the physical parameters characterizing the sputtering process. We find that transitions may take place between various surface morphologies as the experimental parameters (e.g. angle of incidence, penetration depth) are varied. Namely, at short lengthscales the equation describes the development of a periodic ripple structure, while at larger length-scales the surface morphology may be either logarithmically $(\alpha = 0)$ or algebraically $(\alpha > 0)$ rough. Usually stochastic equations describing growth models are constructed using symmetry principles and conservation laws. In most cases it is difficult to derive the equation from a given discrete model. In contrast, here we show that for sputter eroded surfaces the growth equation can be derived directly from a simple model of the elementary processes taking place in the system.

Most of the theoretical approaches focusing on the scaling properties of sputter roughened surfaces have assumed that essentially all relevant processes take place at the surface, and that nonlinear effects would appear only due to non–local effects such as shadowing[[9\]](#page-9-0). However, ion-sputtering is in general determined by atomic processes taking place along a finite penetration depth inside the bombarded material. The incoming ions penetrate the surface and transfer their kinetic energy to the atoms of the substrate by inducing cascades of collisions among the substrate atoms, or through other processes such as electronic excitations. Whereas most of the atoms which are actually sputtered are those located at the surface, the scattering events that might lead to sputtering take place within a certain layer of average depth a. As described by Sigmund's transport theory of sputtering [\[10](#page-9-0)], the average value of the ion deposition depth depends on the energy of the bombarding ions, their angle of incidence, the microscopic structure of the target material and the features of the scattering processes taking place inside the sample.

A convenient picture of the ion bombardment process is sketched in the inset of Fig. [1.](#page-12-0) According to it the ions penetrate a distance a inside the solid before they completely spread out their kinetic energy with some assumed spatial distribution. An ion releasing its energy at point P in the solid contributes an amount of energy to the surface point O , that may induce the atoms in O to break their bonds and leave the surface. Following [\[10,11](#page-9-0)], we consider that the average energy deposited at point O due to the ion arriving at P follows the Gaussian distribution

$$
E_D(\mathbf{r'}) = \frac{\epsilon}{(2\pi)^{3/2}\sigma\mu^2} \exp\left\{-\frac{z'^2}{2\sigma^2} - \frac{x'^2 + y'^2}{2\mu^2}\right\}.
$$
 (1)

In (1) z' is the distance measured along the ion trajectory, and x' , y' are measured in the plane perpendicular to it (see Fig. [1](#page-12-0); for simplicity in the figure x' has been set to 0); ϵ denotes the total energy carried by the ion, a is the average penetration length and σ and μ are the widths of the distribution in directions parallel and perpendicular to the incoming beam respectively. However, the sample is subject to an uniform flux J of bombarding ions. A large number of ions penetrate the solid at different points simultaneously and the velocity of erosion at O depends directly on the total power \mathcal{E}_O contributed by all the ions deposited within the range of the distribution (1). If we ignore shadowing effects among neighboring points, as well as further redeposition of the eroded material, the velocity of erosion at O is given by

$$
v = p \int_{\mathcal{R}} d\mathbf{r} \, \Phi(\mathbf{r}) \, E_D(\mathbf{r}), \tag{2}
$$

where the integral is taken over the region R of all the points at which the deposited energy contributes to \mathcal{E}_O , $\Phi(\mathbf{r})$ is a local correction to the uniform flux J and p is a proportionality constant between power deposition and rate of erosion. In the following we outline the basic stepsin the calculation of v; further details can be found in Refs. $[11,12]$ $[11,12]$.

The calculation of (2) is most efficiently perfomed in the *local* coordinate system (X, Y, Z) shown in Fig. [1](#page-12-0). The ion beam lies in the X-Z plane, forming an angle φ with the Z axis, Z being normal to the surface at O . To simplify the calculations, we assume that: (a) the radii of curvature (R_X, R_Y) of the surface at O are much larger than the penetration depth a, so that only terms up to first order in $a/R_{X,Y}$ are kept; (b) the curvatures attain their maximum and minimum values along either of the X and Y directions, and thus we can expand the value of the surface height at O by taking a second order approximation in the (X,Y) coordinates and consistently ignoring cross term contributions. Performing the integral [\(2\)](#page-3-0) the velocity $v(\varphi, R_X, R_Y)$ is found to be a function of the angle φ and the curvatures $1/R_{X,Y}$ [[11](#page-9-0)].

Finally, from the expression of v we can obtain the equation of motion for the profile. Now it is convenient to use the *laboratory* coordinate frame (x, y, h) . In the absence of overhangs the surface can be described by a single valued height function $h(x, y, t)$, measured from an initial flat configuration which is taken to lie in the (x,y) plane. The ion beam is parallel to the x-h plane forming an angle $0 < \theta < \pi/2$ with the z axis. The time evolution of h is given by

$$
\frac{\partial h(x, y, t)}{\partial t} \simeq -v(\varphi, R_X, R_Y),\tag{3}
$$

where φ is the angle of the beam direction with the local normal to the surface at $h(x, y)$. Now φ is a function of the angle of incidence θ and the values of the local slopes $\partial_x h$ and $\partial_y h$, and can be expanded in powers of the latter. We will assume that the surface varies smoothly enough so that products of derivatives of h can be neglected for third or higher orders.

At this stage additional relevant physical processes must be taken into account to describe the evolution of the surface. First, the bombarding ions reach the surface at random positions and times. We account for the stochastic arrival of ions by adding to (3) a Gaussian white noise $\eta(x, y, t)$ with zero mean and variance proportional to the flux J. Second, at finite temperature atoms diffuse on the surface [\[3,5](#page-9-0)]. To include this surface self-diffusion we allow for a term $-K\nabla^2(\nabla^2 h)$ [\[13](#page-10-0)], where K is a temperature dependent positive coefficient. Expanding (3) and adding the noise and the surface-diffusion terms we obtain the equation of motion [\[14](#page-10-0)]

$$
\frac{\partial h}{\partial t} = -v_0 + \gamma \frac{\partial h}{\partial x} + \nu_x \frac{\partial^2 h}{\partial x^2} + \nu_y \frac{\partial^2 h}{\partial y^2} + \frac{\lambda_x}{2} \left(\frac{\partial h}{\partial x}\right)^2 + \frac{\lambda_y}{2} \left(\frac{\partial h}{\partial y}\right)^2 - K \nabla^2 (\nabla^2 h) + \eta. \tag{4}
$$

From (3) we can compute the expressions for the coefficients appearing in (4) in terms of the physical parameters characterizing the sputtering process. To simplify the discussion we restrict ourselves to the symmetric case $\sigma = \mu$. The general case is discussed in [\[12](#page-9-0)]. If

we write $F \equiv (\epsilon Jp/\sqrt{2\pi}) \exp(-a_{\sigma}^2)$ $\sigma^2(z)$, $s \equiv \sin \theta$, $c \equiv \cos \theta$ and $a_{\sigma} \equiv a/\sigma$, we find for the coefficients in([4](#page-4-0))

$$
v_0 = \frac{F}{\sigma} c \ , \ \gamma = \frac{F}{\sigma} s (a_{\sigma}^2 c^2 - 1), \n\lambda_x = \frac{F}{\sigma} c \left\{ a_{\sigma}^2 (3s^2 - c^2) - a_{\sigma}^4 s^2 c^2 + 1 \right\}, \n\lambda_y = -\gamma \frac{c}{s}, \n\nu_x = \frac{F}{2} a_{\sigma} \left\{ 2s^2 - c^2 - a_{\sigma}^2 s^2 c^2 \right\}, \n\nu_y = -\frac{F}{2} a_{\sigma} c^2.
$$
\n(5)

Consistent with the direction of the bombarding beam and the choice of coordinates made, theterms in ([4\)](#page-4-0) are symmetric under $y \to -y$ but not under $x \to -x$, while for $\theta \to 0$ we get $\gamma = \xi_x = \xi_y = 0$, $\lambda_x = \lambda_y$ and $\nu_x = \nu_y$ [\[15](#page-10-0)]. The equation studied in Ref. [\[11](#page-9-0)] corresponds to thedeterministic linear version of ([4](#page-4-0)), i. e. $\lambda_x = \lambda_y = \eta = 0$.

If ν_x and ν_y are positive, the surface diffusion term is expected to contribute negligibly as a relevant surface relaxation mechanism when we probe the system at increasingly large length scales. Scaling properties are then described by the anisotropic KPZ equation (AKPZ), which predicts two possible behaviors depending on the relative signs of the coefficients λ_x and λ_y [\[16,17\]](#page-10-0). If $\lambda_x \lambda_y > 0$, then $\alpha = 0.38$ and $z = 1.6$, the surface width $w(L, t)$ increases algebraically, being characterized by the exponents of the KPZ equation in $2+1$ dimensions [\[7](#page-9-0)]. For $\lambda_x \lambda_y < 0$, the nonlinear terms λ_x and λ_y become irrelevant, and the width grows only logarithmically, i.e. $\alpha = 0$.

In our case ν_y is always negative, while ν_x can change sign as θ and a_{σ} are varied. A negative surface tension generates an instability in the system. The same kind of instability is known to take place in the Kuramoto–Sivashinsky (KS) equation [\[18\]](#page-10-0), which is the *noisyless* andisotropic version of (4) (4) (4) . It has been argued for the KS equation that in $1+1$ dimensions ν renormalizes to a positive value [\[19](#page-10-0)], and the large length scale behavior is described by the KPZ equation. In 2+1 dimensions it is not completely settled whether the large distance behaviors of KS and KPZ fall in the same universality class, different approaches leading to conflicting results[[20\]](#page-10-0).

In contrast to the KS equation, Eq. [\(4](#page-4-0)) is anisotropic, and explicitly contains a noise term. One can study Eq.([4\)](#page-4-0) using a dynamic renormalization group (DRG) procedure. The linear instability generated by the negative surface tension leads to additional singularities in the Feynman diagrams of the perturbation expansion. This is related to the presence of a characteristic length scale in the system, generated by the competition between surface tension and surface diffusion, $\ell_c = \sqrt{K/|\nu|}$, where ν is the largest in absolute value of the negative surface tension coefficients. Below we discuss the possible scaling behaviour predicted by([4\)](#page-4-0) based on results obtained from DRG calculations performed on its isotropic version [\[21](#page-10-0)], and the results available in the literature for other limits. The complete scaling picture should be provided by either a DRG analysis or a numerical integration of [\(4](#page-4-0)).

The scaling behavior depends on the relative signs of ν_x , ν_y , λ_x and λ_y [[22](#page-10-0)]. The variations of these coefficients as functions of a_{σ} and θ lead to the phase diagram shown in Fig. [2.](#page-12-0) In the following we discuss the various scaling regimes separately.

Region I — For small θ both ν_x and ν_y are negative. As discussed by Bradley and Harper[[11](#page-9-0)] and experimentally studied by Chason *et al.* [\[5\]](#page-9-0), a periodic structure dominates the surface morphology, with ripples oriented along the direction $(x \text{ or } y)$ which presents the largest absolute value for its surface tension coefficient. The wavelength of the ripples is $\lambda_c = \sqrt{2} \ell_c$.

The large length scale behavior $\ell \gg \ell_c$ is expected to be different. Now both nonlinearities and the noise may become relevant. A DRG analysis performed for the isotropic case of Eqn. [\(4](#page-4-0))[[21](#page-10-0),[23](#page-11-0)] suggests that inside region I, the nonlinearities $\lambda_{x,y}$ flow to zero. The surface is logarithmically rough, with a roughness exponent $\alpha = 0$. While in regions I_{a-c} the coefficients of the nonlinear terms may change sign as a_{σ} or θ vary, we do not expect this to modify the scaling behavior.

Region II — This region is characterized by a positive ν_x and a negative ν_y . Now the periodic structure associated with the instability is directed along the y direction and is the dominant morphology at scales $\ell \sim \ell_c$. Much less is known about the scaling behaviour for large length scales. Preliminary results of a DRG calculation indicate that the surface may be stabilized by the positive ν_x and the nonlinearities, and [\(4](#page-4-0)) reduces to the AKPZ equation. If this is the case, then the surface roughness increases algebraically in region IIa , whereas logarithmic scaling characterizes region *IIb*.

Even though several aspects of the scaling behavior predicted by [\(4](#page-4-0)) and [\(5](#page-5-0)) remain to be clarified, we believe that these equations contain the relevant ingredients for understanding roughening by ion bombardment [\[24\]](#page-11-0). To summarize, at short length scales the morphology consists of a periodic structure oriented along the direction determined by the largest in absolutevalue of the negative surface tension coefficients [[5\]](#page-9-0). Modifying the values of a_{σ} or θ changes the orientation of the ripples [[8,11\]](#page-9-0). At large length scales we expect two different scaling regimes. One is characterized by KPZ exponents, which might be observed in region IIa in Fig. [2](#page-12-0). Indeed, the values of the exponents reported by Eklund *et al.* [\[3\]](#page-9-0) are consistent within the experimental errors with the KPZ exponents in 2+1 dimensions. The other regime is characterized by logarithmic scaling $(\alpha = 0)$, which has not been observed experimentally so far. We believe that it can be experimentally tested for suitable (e.g, small) values of the angle of incidence. Moreover by tuning the values of θ and/or a_{σ} one may induce transitions among the different scaling behaviors. For example, fixing a_{σ} and increasing the value of θ would lead from logarithmic (regions Ia and Ic) to KPZ scaling (IIa) , and again logarithmic scaling (IIb) for large enough angles. Also, the measurement of the erosion velocity may help to test the applicability of Eqn.([4\)](#page-4-0). Taking spatial and noise averages in [\(4](#page-4-0)), the terms contributing to the erosion velocity are the ones proportional to v_0 , ξ_x , ξ_y , λ_x and λ_y . In the stationary state, modifying θ or a_{σ} changes the average yield, allowing for a comparison of the experimental yield curves with the ones predicted by([4\)](#page-4-0).

The experimental verification of the above possibilities would constitute an important step to elucidate the interplay between the mechanisms leading to the different morphologies and dynamics for sputter-etched surfaces. It will provide as well additional insight into the scaling behaviors to be expected from equation([4\)](#page-4-0).

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FIGURES

FIG. 1. Reference frames for the computation of the erosion velocity at point O. Inset: Following a straigh trajectory (solid line) the ion penetrates an average distance a inside the solid (dotted line) after which it completely spreads out its kinetic energy. The dotted curves are equal energy contours. Energy released at point P contributes to erosion at O.

FIG. 2. Phase diagram for the isotropic case $\sigma = \mu = 1$. Region Ia: $\nu_x < 0$, $\lambda_x < 0$, $\lambda_y > 0$; Region Ib: $\nu_x < 0$, $\lambda_x > 0$, $\lambda_y < 0$; Region Ic: $\nu_x < 0$, $\lambda_x > 0$, $\lambda_y > 0$; Region IIa: $\nu_x > 0$, $\lambda_x > 0$, $\lambda_y>0;$ Region IIb: $\nu_x>0,$ $\lambda_x>0,$ $\lambda_y<0.$

