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FAST TRACK COMMUNICATION

A continuous non-linear shadowing model of columnar growth

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Abstract
We propose the first continuous model with long range screening (shadowing) that describes columnar growth in one space dimension, as observed in plasma sputter deposition. It is based on a new continuous partial derivative equation with non-linear diffusion and where the shadowing effects apply on all the different processes.

Plasma sputtering is a common process for film growth which often exhibits wide columns that are more or less close packed and separated by thin deep grooves [1–4]. This columnar growth results mainly from a shadowing instability [5–7], where the elevated parts of the surface are more exposed to the sputtering while they shadow the incoming particles to the lower parts. The modelization of this shadowing instability has been well described by probabilistic Monte Carlo (MC) methods [8–10] and also with continuous models based on partial derivative equations (PDE) [6, 7, 11–16] including the seminal work of Bales and Zangwill [1]. However, both approaches fail to describe at long times the strongly non-linear columnar microstructures observed recently (see [2] for instance). In fact, although the continuous models give tall and well separated columns at early time, only few sharp peaks remain later on [6, 14, 16]. Columnar structure using PDE has already been obtained by Gillet *et al* [17] but in that case no shadowing effect was taken into account. On the other hand, discrete approaches using MC methods including shadowing have been developed and have shown a fair description of the columnar structure, particularly through the formation of sharp column sides. However, these models cannot avoid the coarsening of the columnar structures showing larger and larger plateau as time increases, in contrast to the experimental observations.

The goal of this paper is to present a new continuous non-local model which includes both non-linear shadowing and diffusion effects to simulate columnar-like growth. We consider a two-dimensional model where the one-dimensional (1D) surface described by $h(x, t)$ is subjected to receiving particles from all directions not shadowed by the surface itself. Our starting point is deduced from the models initially developed by Bales and Zangwill [1] and by Karunasiri *et al* [6]:

$$\frac{\partial h}{\partial t} = R\Omega(x, \{h\})\sqrt{1 + (\nabla h)^2} + \nu\nabla^2 h + \eta, \qquad (1)$$

where the deterministic deposition term R is multiplied by the solid angle $\Omega(x, \{h\})$ which modelizes the shadowing effect as a long-range screening (see figure 1). ν is the

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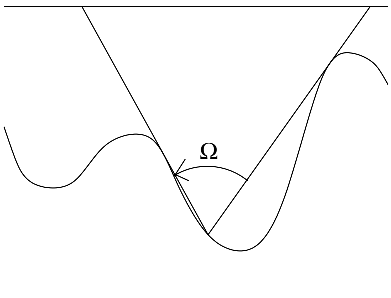

Figure 1. Shadowing process: interface $h(x,t)$ and solid angle $\Omega(x,\{h\})$.

diffusion/relaxation coefficient while η is the usual noise with zero mean $\langle\eta\rangle = 0$ and its correlation given by $\langle\eta(x,t)\eta(x',t')\rangle = 2D\delta(x,x')\delta(t,t')$.

For small surface angles, we retrieve a KPZ-like equation [18] with shadowing effects (defining $\lambda = \pi R$):

$$\frac{\partial h}{\partial t} = \nu\nabla^2 h + \frac{\lambda}{2}(\nabla h)^2 + R\Omega(x,\{h\}) + \eta. \qquad (2)$$

A complete study of these equations has shown that it is unable to reproduce columnar shapes corresponding to experiments and MC simulations [7, 14]. Indeed, in the most favourable situation, only broad peaks emerge instead of flat columns. Experiments thus suggest that the diffusion should be enhanced in the region more exposed to the flux. Moreover, we will assume that the flux also increases (greater than for normal shadowing) on the top of the columns compared with the grooves. Although we have no strong argument for it, we expect some point effect near the sharp edge to be responsible for this process. We then propose the following stochastic differential equation where the main ingredients are non-linear shadowing effects and diffusion:

$$\frac{\partial h}{\partial t} = g(\Omega(x,\{h\}))\,(R\sqrt{1+|\nabla(h)|^2} + \nu\nabla^2 h + \eta). \qquad (3)$$

In this equation $g(\Omega)$ is a given function of the solid angle Ω. Therefore, in order to increase the shadowing effect and the diffusion from the top to the edges and bottom, $g(\Omega)$ has to be stronger than linear, and we will take later on for the numerics $g(\Omega) = \Omega^2$. Multiplying the right-hand side by $g(\Omega)$ leads to an increase of the unshadowed top column growth rate while the shadowed bottom of the column cannot grow. The diffusion is also affected by the shadowing in the same way.

A plane-wave analysis performed on equation (3) shows that the solutions are unstable for large enough wavelengths λ, i.e. $\lambda = 2\pi/k > \lambda^* = \nu\pi^3/(\alpha R)$, with $\alpha \sim 0.724$ and the growth rate $\sigma = k\pi(2\alpha R - \nu\pi^2 k)$. Then, starting from a flat substrate, the noise will trigger the instability and will drive the system into a strong non-linear regime. Figure 2 shows the evolution of the interface profile for different times for $D = 1$, $\nu = 1$ and $R = 1$. It exhibits the desired columnar shape. This shape is characterized by flat column tops and vertical sides as compared with previous MC simulations and columnar growth experiments. The shadowed deposition favours the columnar growth and the anisotropic diffusion smoothes the top of the column very efficiently and leads to vertical sides.

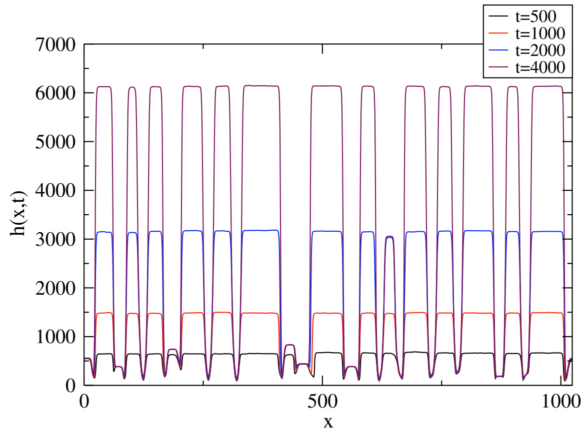

Figure 2. Continuous model. Snapshots of the interface given by the non-linear shadowing anisotropic diffusion model given by equation (3) at time $t = 500$, 1000, 2000 and 4000. The numerical simulation was done with $\Delta t = 0.01$, $\Delta x = 1$ and the total size of the system is $L = 1024$.

(This figure is in colour only in the electronic version)

The competition between these two effects leads to a columnar regime as expected. Moreover, most of the columns formed at the beginning of the simulation are still present at the end. This is also the case for 'Poisson/Wedding cakes' morphologies for which initial columns always remain [20] and for the step meander process [17].

For numerical simulation, equation (3) is integrated using the following explicit scheme:

$$h_i^{n+1} = h_i^n + (\Omega_i^n)^2\Big[\Delta t R\big\{1 + \big((h_{i+1}^n - h_i^n)^2 + (h_{i+1}^n - h_i^n)(h_i^n - h_{i-1}^n) + (h_i^n - h_{i-1}^n)^2\big)\big(3\,\Delta x^2\big)^{-1}\big\}^{1/2} + \frac{\nu\Delta t}{\Delta x^2}(h_{i+1}^n - 2h_i^n + h_{i-1}^n) + \sqrt{\frac{2D\Delta t}{\Delta x}}\varepsilon\Big], \qquad (4)$$

with the notation $h_i^n = h(i\Delta x, n\Delta t)$. ε is a random number picked with the uniform distribution between $[-1, 1[$. To obtain a discrete form of the gradient term, we follow the scheme proposed by Lim *et al* although their study strictly applied for the KPZ equation [19]. $\Omega(x,\{h\})$ is evaluated following [6].

The time evolution of the roughness W of the interface is given in figure 3. It shows the existence of different regimes. The first one, for $t < 1$, is driven by the fluctuations and W scales as $t^{0.5}$. For the second one ($1 < t < 100$), diffusion induced a relative reduction of the roughness which scales as $t^{0.4}$. Then, because of the shadowing instability described above, sharp canyons appear and the roughness quickly increases. Finally, after $t \sim 1000$, the columnar regime appears which leads to $W(t) \sim t$ as in the discrete model [14]. Even if $W(t)$ shows the same scaling as obtained in previous studies on the continuous columnar growth model [14–16], the column shapes are rather different and are now in closer agreement with the MC models and more important with the experiments [3, 4]. Indeed, experiments display

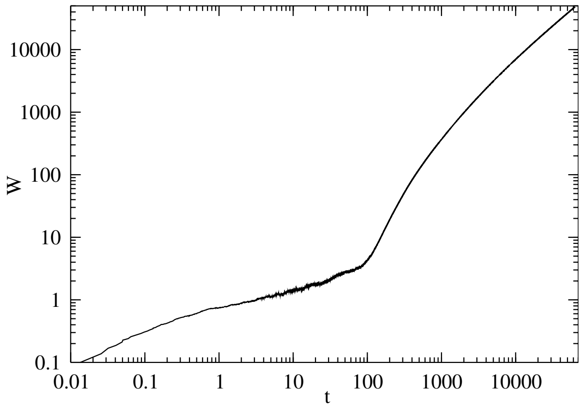

Figure 3. Continuous model. Roughness $W(t)$ as a function of time.

relative constant column width, while MC simulations lead to a column width which is increasing with time. In this respect, our model (3) exhibits better qualitative agreement with experiment than MC simulations.

We have presented the first continuous model, to our knowledge, that exhibits a columnar growth without the coarsening dynamics of the structures, in good agreement with experimental observations on sputtering deposition. For reproducing these wide flat columns with sharp edges, we have introduced an increase in the relaxation and in the flux on the top of the column compared with the grooves. For the sake of simplicity, we have considered a 1D surface and we have taken the same non-local and non-linear multiplicative factor $g(\Omega(x, \{h\}))$ for all the terms of the dynamics. Further on, we have considered a simple power law behaviour for this function $g(\Omega(x, \{h\})) = \Omega^n$. We argue that $n > 1$ is needed to enhance the shadowing effects on the protuberances. We have tested numerically $n = 2$ and $n = 3$ with no loss of properties of the results. However, a better choice of the shadowing function g should be obtained through further experimental comparisons. Similarly, different shadowing functions should be considered in the future for the diffusion term and the deposition term. Finally, this new continuous model, with $n = 2$, considered as a minimal model, already correctly reproduces the formation of flat wide columns, with sharp edges and thin separating grooves, as usually encountered in sputtering deposition. Further works should perform such approaches to two-dimensional surfaces in particular.

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