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Anisotropy and non-universality in kinetic roughening

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Abstract. – We introduce a new model for kinetic roughening which exhibits a non-universal behavior for the roughness exponent, in agreement with many experimental findings. The model, inspired by the chemical etching processes, takes explicitly into account the effect of anisotropy, say the dependence of the growth rule on the local environment conditions. The interplay between anisotropy and non-universality is investigated as well as the relationship with the known universality classes.

The problem of kinetic roughening received an increasing interest in the last few years in relation with non-equilibrium growth models [1] and in view of its practical applications: Chemical Vapor Deposition (CVD) and electro-chemical deposition are just two examples [2]. In this perspective one is interested in studying the dynamic universality classes and a lot of models have been defined starting from the models falling in the universality class of the Kardar-Parisi-Zhang (KPZ) equation [3] which describes the properties of an interface $h(x,t)$ driven by a stochastic noise giving a roughness exponent $\chi = 0.5$, up to the models describing the interface growth in random media, *i.e.* with a quenched disorder, driven by an extremal dynamics. In the last class fall the so-called Sneppen model [4] (in [4] referred to as model B) and the pinning model by directed percolation [5] which both predict a roughness exponent $\chi = 0.63$. We recall that the roughness exponent is defined by the ensemble-averaged width of the interface as $W(l,t) = \langle (h(x,t) - \langle h(x,t) \rangle)^2 \rangle^{1/2} \sim l^\chi f(t^{1/z}/l)$, where z is the so-called dynamical exponent, the angular brackets denote the average over all portions of the interface of length l for a given realization of total length L, and over different realizations, and f is a scaling function such that $f(y) \sim y^\chi$ for $y \ll 1$ and $f(y) = \text{const}$ for $y \gg 1$. The exponent $\beta = \chi/z$ describes the transient roughening.

It is worth stressing how in many experimental studies one measures several different values of χ which are different from the ones predicted by the KPZ and Sneppen universality classes [1]. To give some examples, we remember the experimental studies of paper burning for which one gets $\chi = 0.70 \pm 0.03$ [6], or the propagation of a forced fluid front in a porous medium, which exhibits a roughness exponent $\chi = 0.73 \pm 0.03$ [7] and $\chi = 0.88 \pm 0.08$ [8]. Whether these results represent just a crossover effect to the two quoted universality classes

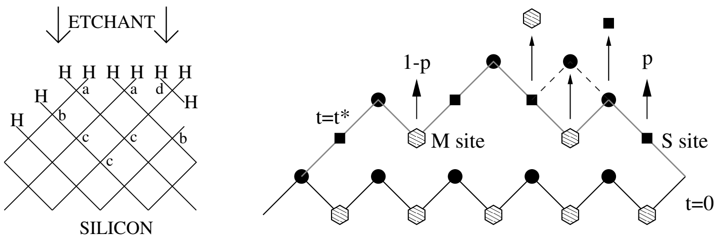

Fig. 1. – Schematic representation of: the crystalline silicon lattice as a square lattice (left); the interface dynamics (right).

or they display some new genuine universality classes not yet included in the usual theoretical approaches, is still an open problem. The second hypothesis suggests that the KPZ and Sneppen models capture the physics of kinetic roughening only partially, and that some other physical ingredients could be lacking. From this the need of more refined models in order to give a realistic description of such phenomena.

In this paper we try to follow this second hypothesis and introduce a new model for kinetic roughening that allows us to reproduce a non-universal behavior of the roughness exponents. In particular we consider a model which explicitly includes an *anisotropy* factor, say a growth rule dependent on the local environment of the growing site. The model thus presents a complex interplay between a global equilibrium and the conditions of a local dynamics.

This choice is motivated by the observation of roughening phenomena occurring in etching processes which represent an important tool either in academic research or in device technology, *e.g.* the preparation of single-crystal samples of desired dimensions, shapes and orientations. One of the most important properties of these processes is represented by the intrinsic anisotropy [9] of the etch rates. For instance, in samples of crystalline silicon etched in solutions of aqueous potassium-hydroxide (K-OH) with isopropil alcohol (IPA), depending on the concentration of the etchant and the temperature, the (111) direction etches slower than the others by a factor which can be of order 100 or more [10]. The degree of anisotropy affects the properties of the surface, which turns out to be rough with a non-universal roughness exponent.

Our model incorporates the effect of the anisotropy of the growth at a microscopic level. Although the definition of the model is very general, let us briefly consider the chemistry of the etching process in order to exhibit a physical framework allowing us to understand the meaning and the interpretation of the definitions.

The disorder in the etching process is related to the impurities in the lattice which change the distribution of binding energies in a quenched way: the etchant invades the system in a time scale much shorter than that of the time life of the impurities. We can thus look at the etching as an extremal process, where the etchant dissolves the atom with the smallest binding energy. This is correct for low etchant concentrations and corresponds to the situation experimentally more interesting, giving rise to rough surfaces.

In our model, the anisotropy is introduced by a phenomenological tunable parameter, p, which distinguishes sites with a different local environment. Let us look at the meaning of p in the case of etching. If we represent the crystalline lattice of the silicon on a two-dimensional plane, we can imagine a square lattice where (see fig. 1, (left)) the atoms can be found in each of the four positions marked in the figure by the letters *a*-*d*. The four positions correspond to different oxidation states: from the situation c (oxidation number 0) which occurs only in the bulk, to the situation d (oxidation number -3). All the surface atoms are passivated by hydrogen atoms. The atoms in the positions a and b, corresponding respectively to the oxidation numbers -2 and -1 (two and one heteropolar bonds, *i.e.* Si-H bonds), play an important role in explaining, at least from a heuristic point of view, the origin of the anisotropy in the etch rates [9]. The parameter p quantifies the ratio of the etch rates between the sites in the positions a and b. The basic idea is that in the (111)-plane of silicon, there is only one heteropolar bond per silicon atom. Therefore there are three bonds to break for dissolution, while other planes (except the (110)) have more than one heteropolar bond and accordingly a smaller number of bonds must be broken. The growth of the interface, in the etching process, represents the invasion of the etchants into the silicon wafer.

Let us turn to the detailed definition of the model. On a square lattice tilted at 45° we consider a $(1+1)$-dimensional interface $h(x) = h(x,t)$ with $x = 1, 2, \ldots, L$ whose initial condition is given by: $h(2x, 0) = 1\ \forall x \in [1, L/2]$ and $h(2x-1, 0) = 0\ \forall x \in [1, L/2]$. The interface, which locally satisfies the conditions [4] $|h(x,t) - h(x \pm 1, t)| \leq 1$, contains two classes of random variables that correspond to two separate classes of sites: the sites (M) for which $\nabla^2 h > 0$ holds (called minimum sites), which are, microscopically, the atoms with two heteropolar bonds, and the sites on a slope (slope (S) sites) for which one has $\nabla^2 h = 0$. These last sites correspond microscopically to atoms with one heteropolar bond. To each class of sites a class of Gaussian-distributed uncorrelated random variables is assigned which mimics the disorder: $\eta(x,h) \in [0:0.5]$ for S sites and $\eta(x,h) \in [0.5:1]$ for M sites. The sites with $\nabla^2 h < 0$, for which all the chemical bonds are homopolar, *i.e.* Si-Si, do not take part in the dynamics and they are assigned a zero value of the random variable. Periodic boundary conditions are assumed.

The parameter p can vary in the range $[0:1/2]$. For $p = 1/2$ all the sites which take part in the dynamics are equivalent and there is no anisotropy, whereas the case $p = 0$ corresponds to the maximal anisotropy in which $v_{(111)}/v_{(100)} = 0$, where $v_{(111)}$ and $v_{(100)}$ are the etch rates in the corresponding directions. The system evolves by updating the site i^* with the *biggest* r.v. in one of the two classes of sites chosen, at its turn, with a probability p. In practice, one updates a site (S) with probability p and a site (M) with probability $1-p$ according to the rules (see fig. 1, (right))

1) $h(i^*, t+1) = h(i^*, t) + 2, \quad \eta(i^*, h(i^*, t+1), t+1) = 0.$

2) Updating all the sites necessary to make the local conditions $|h(x,t) - h(x \pm 1, t)| \leq 1$ satisfied [11].

3) Updating of the random variables for the sites which changed their class of belonging. In particular $\eta(x,h,t+1) = 1/2 * RAN$ if $\eta(x,h,t) = 0$ and $\eta(x,h,t+1) = \eta(x,h,t) + 1/2$ if $\eta(x,h,t) \neq 0$, where RAN is a random value between 0 and 1.

4) Updating of the random variables of the sites which have changed their height but which did not change their class of belonging (sites S only): $\eta(x,h,t+1) = 1/2 * RAN$.

We have studied this cellular automaton by numerical simulations in order to analyze its dynamical roughening properties. We have chosen a size $L = 2048$ for our numerical simulations. For each value of the parameter p (we have considered $p = 0.0, 0.02, 0.2, 0.5$) we have computed the growth exponent β, which rules the time evolution of the width $W(t)$ of the surface ($W(t) \sim t^\beta$) before the stationary critical state is reached, and the roughness exponent χ, which gives the scaling of the width of the surface ($W(l) \sim l^\chi$), in the stationary state. The stationary state is called self-organized [12] in that it is reached spontaneously by the system independently of the initial conditions. It is moreover critical because the growth happens as a sequence of avalanches which are power law distributed [13].

TABLE I. – *Values of the roughness exponent χ and of the dynamical exponent β in our model for different values of the anisotropy parameter p.*

p	χ	β
0.0	0.93 ± 0.02	0.93 ± 0.01
0.02	0.75 ± 0.02	0.96 ± 0.01
0.2	0.65 ± 0.02	0.96 ± 0.01
0.5	0.63 ± 0.02	0.96 ± 0.01

To ensure that the system is in the stationary state, we have studied the behavior of the n-th moments of $h(x,t)$ normalized to the second momentum:

$$m_n(t) = \left\langle \sum_i (h(i,t) - \bar{h}(t))^n \right\rangle \bigg/ \left(\left\langle \sum_i (h(i,t) - \bar{h}(t))^2 \right\rangle \right)^{n/2}, \tag{1}$$

here $\bar{h}(t)$ is the mean surface height at time t. We get that, after a transient, all the odd moments vanish and the even ones tend to constant values. In particular, the condition for the skewness $m_3 = 0$, which characterizes the stationary critical state [4], is realized after about 10^3 time steps per site, independent of the value of p.

For each value of p, 10^2 simulations lasting 10^7 time steps have been performed. In table I we report the measured values for the exponents χ and β. When measuring the exponent χ, we performed the numerical fit on an intermediate region $l \in [2, 128]$, to eliminate the deviations from the power law scaling induced by the lower and upper cutoffs. Figure 2, (left) shows the scaling behavior of $W(l)$ for different values of p. One can see that χ varies continuously from the value $\chi = 0.93 \pm 0.02$ (for $p = 0.0$, *i.e.* maximal anisotropy) corresponding to the Sneppen model A [4], to $\chi = 0.63 \pm 0.02$ (for $p = 0.5$, *i.e.* no anisotropy) corresponding to the Sneppen model B. For values of $l > 128$ the curves deviate from the self-similarity due to a genuine finite-size effect. We have checked this phenomenon by performing very time-consuming simulations for values of $L > 2048$ which show how the self-similar region extends to larger regions. The value of χ for $p = 0.0$ is clearly a finite-size effect. We checked how this value tends, in the limit $L \to \infty$, to the value $\chi = 1.0$ found for the Sneppen model A [4]. The exponent β shows, instead, a more universal behavior, assuming a nearly stable value around 0.95 for all values of p (see table I).

We have also studied the time evolution of the distribution of random variables on the invading interface (the histogram $\Phi_t(\eta)$, where η is the generic value for $\eta(x,h)$), which is of great importance for models with extremal dynamics. One can see that $\Phi_t(\eta)$ self-organizes into a distribution which is the superposition of two theta-functions, one for each class of variables, each one characterized by a critical threshold $p_c(p)$ depending on the parameter p. A more detailed and extended analysis of the model will be reported in a longer paper [13]. The complete analysis, which includes a study of temporal correlations, avalanche distributions, and the power spectra of the asymptotic interface, shows quite clearly that the model has non-universal critical properties modulated by the parameter p.

In order to qualitatively check the reliability of the introduction of the anisotropy parameter p, we have performed small-scale experiments on crystalline silicon, by producing, under different conditions, and analyzing textured rough surfaces. The most important outcome is

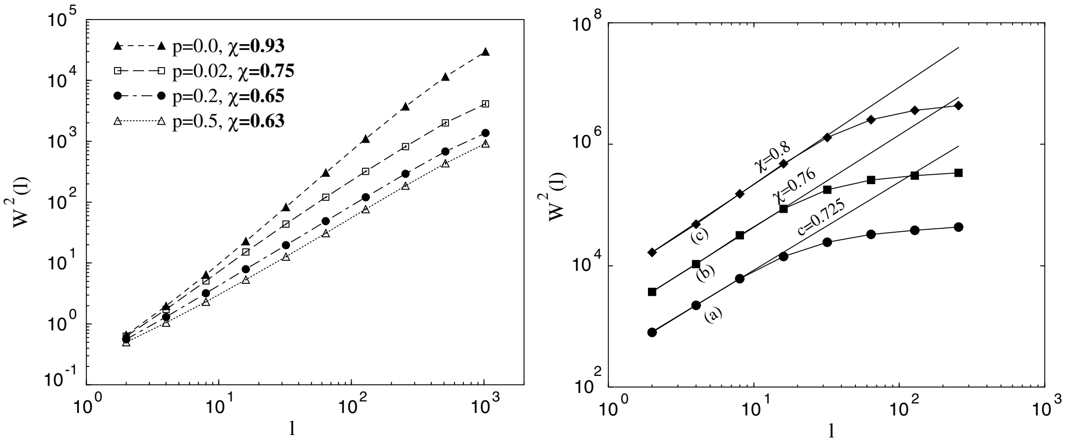

Fig. 2. – Left: Scaling behavior of the width $W(l)$ in the framework of our model for different values of p. $L = 2048$, total evolution time $T = 10^7$ steps. To avoid finite-size effects induced by the upper and lower cutoff, the values of the exponent χ reported in the figure are obtained by a least-square fit in the region $l \in [2, 128]$. Right: Experimental behavior of the width $W(l)$ *vs.* the length l for three different etched silicon samples. Curve (a) is obtained with a small concentration of the etchants (2% KOH, 10% IPA) and using ultrasound bath for the agitation. Curves (b) and (c) are obtained with higher concentrations of the etchants (5% KOH, 16.7% IPA) with (for (b)) and without agitation (for (c)). The deviation from power law in the final part of the data is attributed to saturation effects connected to the finite depth of the samples. The results of a numerical fit performed on the first part are shown.

the non-universal character of the roughness experiment of the surfaces produced with different degrees of anisotropy. At its turn the degree of anisotropy can be related to the temperature and to the environment, so it should depend on the etchants [9], [10].

The setup of the experiment was the following. A wafer of lapped silicon is immersed, with the specular face oriented upwards, in a solution of 600 ml of KOH at different concentrations and 100 ml of isopropil alcohol (IPA), whose temperature is kept fixed to 75 °C by a thermostat. A flat glass cover was used to close the vessel in order to keep the volumetric ratios constant during the etching process. The mixing process was achieved by using ultrasound bath.

The overall effect of the etching is the production of pyramidal structures of all sizes on the interface, with a distribution of dimensions depending on the anisotropy of the process, on the agitation, and on temperature.

We have studied in particular the roughness of the surfaces by analyzing the behavior of the root mean square $W(l)$ as a function of the length l of the sample. In order to do that we have used a Long Scan Profiler Tencor P-1 which allows measures of roughness with 1–25 Å resolution. In each sample we performed 25 scannings 100 μm long and 2 μm away from each other and we measured the roughness on the reconstructed surface by averaging over the 25 scannings. We have analyzed three samples and the results for the roughness are shown in fig. 2, (right). One can note how the effect of the ultrasound agitation is to globally decrease the dimensions of the pyramidal structures. The concentration has the same qualitative effect: the lower is the concentration of the solution, the smaller is the dimension of the pyramids. In general these two factors, the agitation and the decreasing of the concentration of the etchants, tend to reduce the anisotropy in the etch rates.

A quantitative measure of the properties of the surface is given by the roughness. In this case (see fig. 2, (right)) a numerical fit of the experimental data gives different values of the roughness, starting from the value $\chi = 0.8$ for the sample (c) (no agitation and high etchants concentrations) to the value $\chi = 0.76$ in the sample (b) and $\chi = 0.725$ for the sample (a) (agitation and low etchants concentrations). This is just a qualitative confirmation of our model, although the quality of the experimental data is not good enough to make a more quantitative comparison.

In summary in this paper we have introduced a new model for kinetic roughening whose real novelty lays in the explicit introduction of anisotropy in the growth rules which turns out to be dependent on the local environment. The introduction of the anisotropy, by means of a phenomenological parameter p, opens a new scenario in which the system exhibits non-universal critical properties dependent on p which is in line with the experimental available results [6]-[8] and which supports the existence of several universality classes.

Although the simple introduction of only one anisotropy parameter p is far from being able to capture all the characteristics of etching processes (*e.g.*, transport phenomena in the solution are likely to be important and both concentration and agitation have strong effects on transport), it nevertheless captures at least some basic elements of the relationship between anisotropy and the non-universality in etching processes. The general requirement of a microscopic dynamic rule depending on the local environment could be a key element in the observed non-universality in kinetic roughening phenomena. In the debated question whether the discrepancy between experimental results is due to crossover phenomena or to an intrinsic non-universal nature of kinetic roughening phenomena, our model suggests a possible *theoretical* way to have a non-universal phenomenology.

It is worthwhile to stress how our model suggests the possibility of several analytical approaches, from the treatment of the problem in terms of a continuous stochastic dynamical equation [13], to the application to the discrete model of a method recently proposed for dynamical models driven with an extremal dynamics [14].

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