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LETTER TO THE EDITOR

Roughness scaling of plasma-etched silicon surfaces

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Abstract. Atomic force microscopy reveals scaling behaviour of silicon surfaces etched by plasma. The experimental results are compared with some theoretical models. It is shown that plasma-induced roughness is driven by a phenomenon that can be described by shadowing instabilities resulting in columnar microstructure growth. The same scaling properties as are predicted by a growth model are obtained.

The problem of interface roughness has received particular attention, especially on a theoretical basis [1]. This is due to its practical connection to thin-film growth. However, rather little effort has been made to interpret experimental data in terms of kinetic roughening as can be done for the interaction of plasma with materials [2]. Our aim in the present work is to analyse plasma-etched silicon surfaces in terms of various roughness growth models.

The possibilities offered by scanning probe methods as regards this problem have been stressed in various articles [3, 4].

In a previous paper [5], we have described experiments dealing with a quantitative study of the roughness induced by SF_6 plasma etching of silicon surfaces. The advantage of independently varying the plasma parameters was found to carry over. As a major result, an empirical analytical form describing the surface roughness as a function of these parameters has been established. What we want to discuss here is the time dependence of this roughness.

Using an atomic force microscope, topographical maps of the surface have been recorded. The root mean square value σ of the roughness height $h(x, y)$ was found to obey the following phenomenological power law [5]:

$$\sigma(t) \propto \frac{1}{\sqrt{E}} \left(\frac{J^+}{J_F} \right)^{\eta} t^{\beta} \tag{1}$$

with

$$\eta = 0.45 \qquad \text{and} \qquad \beta = 1$$

where J^+ is the ion flux impinging on the substrate, E the kinetic energy of the ion and J_F the fluorine-atom flux.

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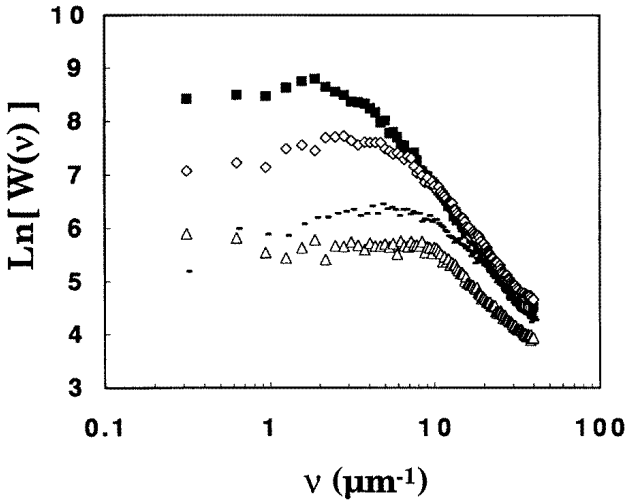

Figure 1. The power spectral density behaviour versus exposure time: (■) 600 s; (◇) 240 s; (−) 120 s; (△) 30 s.

Additional information can be gained by performing a power spectral density (PSD) analysis of the etched surfaces [3, 4], and typical PSD graphs of $W(\nu, t)$ are displayed in figure 1. The relation between W and h is given as follows:

$$W(\nu, t) = \frac{1}{\text{area}} \left| \int\int \frac{\mathrm{d}^2 r}{2\pi} \, \mathrm{e}^{-\mathrm{i}\nu \cdot r} h(\boldsymbol{r}, t) \right|^2 \qquad \boldsymbol{r} = (x, y) \tag{2}$$

where ν is the spatial frequency.

Several other statistical values can be derived from $W(\nu, t)$. In particular, $\sigma(t)$ can also be obtained by integration of $W(\nu, t)$ in the 2D ν-space, but two other quantities which summarize the general behaviour of $W(\nu, t)$ are very important.

The first one is the correlation length ζ_0 ($=1/\nu_0$, which defines the lateral extent of the roughness). If we define the corrugation as the slope of a line connecting two points on the surface, then the corrugation becomes small for points separated by a length longer than the correlation length ζ_0. For lengths longer than ζ_0, the surface can be considered to be flat. Roughly, one can thus expect that the PSD function should be independent of ν for $\nu < \nu_0$ (with a low-frequency value W_0), while it should decrease for $\nu > \nu_0$. This behaviour is observed in most cases (see figure 1).

The other relevant information is the behaviour at large ν. From the ν-power-law dependence of the PSD function tail, a local fractal dimension might be inferred from the value of the exponent in the case where it extends over a large enough range of ν-values.

Our main goal is to identify the basic mechanisms of the roughness induced by plasma etching by testing various growth models. The simplest model is purely stochastic: the system is random and uncorrelated. In this case, as for a Brownian process, $\sigma(t) \propto t^{0.5}$.

This is not what we have observed. The analysis of the experimental data plotted in figure 2 has led to $\beta = 1$, and the correlation length $\zeta(t)$ was found to follow a power law: $\zeta \propto t^{0.66}$. This means that a simple stochastic approach is not sufficient for explaining the roughness evolution induced by the plasma. This has also been observed for ion-beam erosion of various materials [4, 6].

Several other approaches can be derived from the random model simply by adding

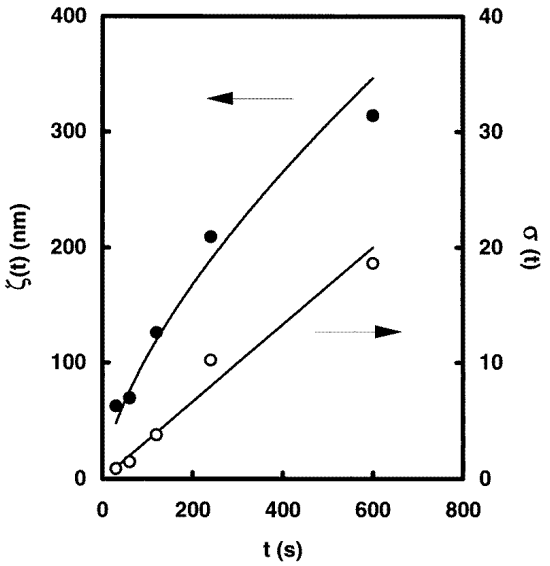

Figure 2. The correlation length $\zeta(t)$ and rms height $\sigma(t)$. The solid line is the best fit for each set of data points. $\sigma(t)$ is linear and $\zeta(t) \sim t^{0.66}$.

smoothing processes like, for instance, surface diffusion [7]. Without going into details, we already know that they predict smoother surfaces (i.e. a lower exponent of $\sigma(t)$ versus time) [7]. Since they also fail to represent the actual process, they are not considered further.

The other kind of theory uses a scaling-law formalism developed from analysis by renormalization group techniques of non-linear Langevin diffusion equations for interface growth evolution [4, 8]. We are aware that growth processes are different to erosion and sputtering, but the scaling formalism is expected to be of great help in analysing the present experimental data. This latter theory leads to the equation

$$\frac{\partial h(r,t)}{\partial t} = D_s \nabla^4 h(r,t) + D_v \nabla^3 h(r,t) + \nu \nabla^2 h(r,t) + \lambda(\nabla h(r,t))^2 + R\Omega(h,t) + \eta(r,t) \tag{3}$$

where D_s expresses surface diffusion, D_v volume diffusion, ν evaporation and redeposition, and λ is a non-linear coupling term (here it encompasses growth or etching parallel to the substrate), R is the growth (or etching) rate, Ω the shadowing solid angle (which is a non-local term) and η a noise term, uncorrelated in space and time, which generates the roughness. Each of the above-mentioned theories makes certain of these terms dominant. The most popular are the Kardar–Parisi–Zhang (KPZ) theory ($D_v = D_s = R = 0$) [1, 9–12], the Kim–Kosterlitz theory [13] (developed from a restricted solid-on-solid growth model) and models based on Villain conjectures [14, 15] on one hand and shadowing instability theories [16–19] (either $D_s = D_v = 0$ or $D_v = \nu = 0$) on the other hand. Shadowing theories are used for explaining columnar growth (such as occurs in ion sputtering, explosive crystallization and viscous fingering), while the others (called hereafter KPZ-like theories) deal with kinetic roughening (such as occurs in molecular beam epitaxy [8, 14, 15], ballistic deposition [20–22], Eden growth [20], wetting [23] and adsorption [24, 25]). Generally, such theories have been tested for both $d = 1 + 1$ and $d = 2 + 1$ dimensions.

Within the framework of KPZ-like theories, the kinetic roughness width $\sigma(L, t)$ is

thought to obey the dynamic scaling hypothesis [20]

$$\sigma(L,t) \propto L^{\alpha} f(t/L^{z}) \tag{4}$$

where L is the size of the image and $f(x)$ is the dynamic scaling function, with the following asymptotic properties:

$$f(x) = \begin{cases} 1 & \text{for } x \gg 1 \\ x^{\beta} & \text{for } x \ll 1, \text{ with } \beta = \alpha/z. \end{cases}$$

Thus the kinetic roughness σ for a substrate of size L grows as [1]

$$\sigma \propto L^{\alpha} \qquad \text{for } t \gg L^{z}$$

and

$$\sigma \propto t^{\beta} \qquad \text{for } t \ll L^{z}, \text{ where } \beta = \frac{\alpha}{z}.$$

The lateral correlation length is given by $\zeta \propto t^{1/z}$.

Another important relation is often used: $\alpha + z = a + \alpha/\beta = 2$ and this is considered as an exact result resulting from the invariance of the KPZ equation on tilting the interface by a small angle (galilean invariance) [1, 9].

Since in our case $\beta = 1$, from the above-mentioned relations, one immediately finds $\alpha = z = 1$. This is in contradiction with the time evolution of the correlation length measured experimentally, i.e. $1/z = 0.6$. Thus a KPZ-like model does not describe the roughness evolution in our case and another kind of theory has to be searched for.

Recently, very interesting calculations of columnar growth were carried out in $d = 2 + 1$ dimensions [18], involving shadowing instabilities leading to the growth of columnar microstructures. In this case the scaling law is somewhat different and originates from the dynamics of an order–disorder transition with a non-conserved order parameter; see [18, 19, 26]. In that case, we have the following relation:

$$W(\nu, t) \propto t^{\delta} F(\nu t^{1/z}) \tag{5}$$

which (using $\sigma \propto t^{\beta}$) leads to [18]

$$\delta = 2\beta + 2\frac{1}{z} \qquad \text{in } 2 + 1 \text{ dimensions.} \tag{6}$$

One can thus deduce

$$\zeta \propto t^{1/z} \qquad \text{and} \qquad W_0 \propto t^{\delta}. \tag{7}$$

One of the more striking consequences of this model is that the interface width $W(t)$ increases linearly with time ($\beta = 1$). Moreover, in the framework of this model, ζ is found to scale as $t^{0.66}$ in $2 + 1$ dimensions [18] and equation (6) gives $\delta = 3.2$ as a final result. The values deduced from our experimental data are precisely $\beta = 1$ and $1/z = 0.66$. Moreover, an independent direct measurement of δ is possible through the time evolution of W_0 (not shown) [5]. We find $\delta = 3.4$, which is in close agreement with the theoretical prediction.

Thus our experiments, surprisingly, provide a direct test of the shadowing instability model. This agreement cannot be considered as merely fortuitous, because we actually do observe columnar growth in the images.

However, we must recall that the shadowing model describes growth and that our experiments deal with etching. If we recall that ions continuously create defect sites at the outermost surface where fluorine atoms are more easily bound to silicon, the rate of formation of a volatile compound, say SiF_4, is accelerated compared to what happens on

a flat surface. This means that etching occurs predominantly at these sites, due to the lower binding energy of silicon and high local electric fields (ionic bonds between Si and F atoms). Thus the bottom of a native column is etched more quickly than the top, which is consistent with these mechanisms. Such a phenomenon has already been encountered, in fabrication of microporous silicon using electrochemical dissolution with electrolytes containing hydrofluoric acid [27, 28]. In fact, such behaviour is similar to the columnar growth that occurs as a result of shadowing during deposition: in this case, the top of the native column grows more quickly than the bottom, which is equivalent to a faster etching of the bottom. Finally, this explains why the shadowing model is successful in describing the roughness growth during etching.

In conclusion, plasma-induced roughness growth provides a good experimental test for scaling-law formalisms involved in growth models. For the first time, we present experimental results which are consistent with columnar structure growth driven by long-range shadowing instabilities. It is worthwhile to point out that this remarkable agreement has been obtained without the use of any adjustable parameter.

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