

# Scaling behavior and mechanism of formation of SiO<sub>2</sub> thin films grown by plasma-enhanced chemical vapor deposition

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This paper reports a study of the kinetic roughening of SiO<sub>2</sub> thin films prepared by plasma-enhanced chemical vapor deposition (PECVD). Tetramethylsilane has been used as a precursor, and the synthesis has been carried out under remote and in-plasma configurations. The analysis of surface topography of the films by atomic force microscopy shows an anomalous scaling behavior that cannot be represented by the Family-Vicsek scaling relation of dynamic scaling theory. The application of different methods for obtaining the roughness exponent  $\alpha$  yields different values of this exponent ( $\alpha=0.7$  for the height-height correlation function and  $\alpha=1.3$  for the power spectral density function for long deposition times) in all experimental conditions. Moreover, a strong variation of the  $\alpha$  exponent with deposition time has been determined for the samples grown in remote mode. This correlates with the presence of a crossover region of the growth exponent  $\beta$ , which varies from a first value of 1.3 for low deposition times to another of 0.3 for longer deposition times. Such a variation is not found for the samples grown in the plasma, characterized by a  $\beta$  value of 0.28. The results obtained can be explained by the combined effect in the growth process of a low diffusivity of the physisorbed species along with the existence of nonlocal interactions due to shadowing effects. These two assumptions are in agreement with the empirical knowledge existing about the kinetics of the growth of SiO<sub>2</sub> thin films by PECVD and establish a link between the scaling properties of the films with the surface chemistry during the film growth.

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## I. INTRODUCTION

The study of the surface microstructure of SiO<sub>2</sub> thin films is of great interest in different technological fields such as microelectronics and protective and optical coatings.<sup>1-4</sup> Two important requisites for these applications are that the films present a good conformality and a low roughness, characteristics that impose a very precise control over the growing mechanisms of the films.<sup>5,6</sup> Among the different techniques used for the deposition of these materials, plasma-enhanced chemical vapor deposition (PECVD) has a great potential due to its fast deposition rate and the possibility of working at room temperature, a feature that is of great importance when the substrates and/or the films can be thermally degraded.

The deposition by PECVD of SiO<sub>2</sub> thin films has been intended with a number of different precursors for which information is available about the elemental reactions taking place in the plasma and about the nature of the possible fragments that can be involved in the film growth.<sup>7,8</sup> It is also known that a high mobility of the incoming species when they adsorb on the surface is critical for the development of homogeneous and conformal coatings with a low roughness. Thus, for example, by theoretical simulation of the deposition process and the analysis of the coatings, it has been found that the plasma species involved in thin film growth when using silane as a precursor are characterized by a low surface diffusivity.<sup>5</sup> Meanwhile, when using tetraethoxysilane (TEOS) as a precursor, the plasma fragments present a physisorbed state that favors the growth of smooth and conformal coatings.<sup>6</sup>

A possible way of assessing the growth mechanism of thin films is by analyzing their surface topography within the

concepts of the dynamic scaling theory (DST).<sup>9</sup> In general, the roughness of a film appears as a balance between roughening and relaxation mechanisms of the surface. As a consequence, from the study of the roughness evolution, it is possible to obtain information about the mechanisms controlling the growth of the films.<sup>10,11</sup> This provides a means to account for the influence of certain surface mechanisms (condensation and/or evaporation, diffusion, lateral growth, etc.) on the thin film growth and, therefore, on microstructural properties such as roughness and conformality. In many experimental cases, the roughness of the films presents a well defined scaling behavior as a function of both the deposition time and scale of measurement.<sup>12-15</sup> The scaling behavior, according to the conventional dynamic scaling theory, can be represented by the Family-Vicsek scaling relation<sup>9</sup>

$$\sigma(L,t) = L^\alpha f(t/L^{\alpha/\beta}), \quad (1)$$

where  $\beta$  and  $\alpha$  are the so-called growth and roughness exponents and  $f(u)$  presents the following two asymptotic behaviors:  $f(u) \sim u^\beta$  for  $u \ll 1$  and  $f(u) \equiv \text{const}$  when  $u \gg 1$ .

This scaling behavior appears as a consequence of the self-affine character of the interface, admitting a power dependence on the deposition time of the vertical and horizontal correlation lengths of the surface. Experimental evidences of such scaling behavior has been found for thin films prepared by different techniques such as evaporation,<sup>16</sup> thermal chemical vapor deposition (CVD),<sup>17</sup> sputtering,<sup>18</sup> or PECVD.<sup>15,19</sup>

However, in other experimental systems, the scaling relation defined by Eq. (1) is not fulfilled.<sup>20-22</sup> The system is said to present an anomalous scaling that generally causes the

appearance of distinct values of the exponents  $\alpha$  and  $\beta$  depending on the scale of measurement.<sup>23</sup> In this context, we have recently reported the modeling by kinetic Monte Carlo simulations of thin films grown by PECVD and the correlation with the actual roughness evolution of real SiO<sub>2</sub> and SiO<sub>x</sub>C<sub>y</sub>H<sub>z</sub> thin film surfaces.<sup>24</sup> A general conclusion from this modeling is that by PECVD, the particles arriving to the surface may span a large set of incoming angles and that, for oxide surfaces, they stick on the same place where they touch the surface. This renders an anomalous behavior characterized by a scaling law that cannot be represented by the Family-Vicsec relation.

In this work, the evolution of the roughness scaling properties is studied for SiO<sub>2</sub> thin films grown by PECVD using O<sub>2</sub>/tetramethylsilane (TMS) mixtures. The purpose of the study is, first, to understand the scaling properties of these PECVD films as a function of both deposition time and scale of measurement and, second, to obtain further information about the processes that are involved in the kinetic roughening of their surfaces. The intention is also to contribute to the general understanding of the processes controlling the growth of films prepared by PECVD, a procedure where the interpretation of the results is not so straightforward compared to other procedures such as molecular beam epitaxy or evaporation.<sup>15,19</sup>

This paper is structured as follows: Sec. II describes very briefly the basic concepts and terminology of the DST with a special mention to the situations that can be considered anomalous, Sec. III describes the experimental conditions used for the deposition and the analysis of the films, and Sec. IV reports the main experimental results obtained. In Sec. V, these results are discussed in terms of the current knowledge existing about the scaling behavior of thin films. A proposal about the possible mechanisms responsible for this behavior is also made in this section. Finally, some conclusions are presented in Sec. VI.

## II. THEORETICAL BACKGROUND

The conventional DST assumes the development of self-affine surfaces in which there exist scaling relations between the surface roughness [defined as the root mean square surface fluctuation  $\sigma = \langle (h(\mathbf{x}) - \bar{h}) \rangle$ ] and both the deposition time and the scale of measurement. As we have already indicated, according to this theory, the behavior of the surface roughness follows the Family-Vicsec relationship, which provides two asymptotic scaling behaviors for  $\sigma$  (Ref. 9):

$$\begin{aligned} \sigma &\sim t^\beta, & t \ll, \\ \sigma &\sim L^\alpha, & t \gg. \end{aligned}$$

Therefore, initially, the roughness increases with deposition time at a rate determined by the growth exponent  $\beta$  until a saturation value of the roughness is achieved. The saturation value depends on the scale of measurement through the roughness exponent  $\alpha$ . The crossover time between the two regimes changes with the scale of measurement through the dynamic exponent  $z$ , so that  $t_\times \sim L^z$ . Also, the correlation

length increases with time following the potential relation  $\zeta \sim t^{1/z}$ . In general,  $z = \alpha/\beta$ .

The presence of a scaling behavior described by Eq. (1) causes the appearance of some useful scaling properties for the height-height correlation function

$$H(L) = \sum_x [h(x+L) - h(x)]^2$$

and the power spectral density

$$\text{PSD}(k) = \langle \hat{h}(k,t) \hat{h}(-k,t) \rangle,$$

where  $\hat{h}(k,t)$  is the Fourier transform of the surface.

For systems that follow the Family-Vicsec relation, the following asymptotic behaviors are expected for these functions:

$$H(L) \sim \begin{cases} L^{2\alpha}, & L \ll \\ \text{const}, & L \gg \end{cases}$$

and

$$\text{PSD}(k) \sim \begin{cases} \text{const}, & k \ll \\ k^{-(2\alpha+d)}, & k \gg, \end{cases}$$

with  $d$  being the dimension in which the PSD( $k$ ) is taken. These two asymptotic behaviors are characteristic of statistical self-affine interfaces. In this sense, the growth exponent  $\alpha$  is equivalent to the Hurst surface exponent.

In some cases, the system does not obey the scaling relation given by Eq. (1) and presents an anomalous scaling behavior which, in some cases, has been explained in terms of more general scaling relations.<sup>25,26</sup> In these systems, although variations of the power spectral density of the type  $k^i$  are still expected for low values of  $k$ , the corresponding roughness exponent is different from that obtained using the height-height correlation function. According to the literature, such an anomalous behavior may appear either as resulting from the existence of nonlocal interactions during the surface growth (i.e., shadowing effects) or as a direct consequence of the local growing rules (intrinsic anomalous scaling).<sup>23</sup>

Another important point is that in the conventional DST,  $\alpha$  and  $\beta$  exponents do not change with deposition time. However, in the case of the growth exponent  $\beta$ , regions characterized by different values of this exponent have been found in systems where there is a transition between different growth modes, each of them satisfying independently the Family-Vicsec scaling relation.<sup>17</sup> Likewise, other authors have found a time-dependent  $\alpha$  exponent. This is, for example, the case of Zhu *et al.* studying the growth of diamondlike carbon films.<sup>27</sup>

## III. EXPERIMENT

Thin films were grown in a low pressure microwave surface-wave plasma reactor, which has been described in detail in a previous work.<sup>28</sup> The samples have been grown under two different conditions: the first one with the substrate holder placed in a remote configuration at 20 cm from

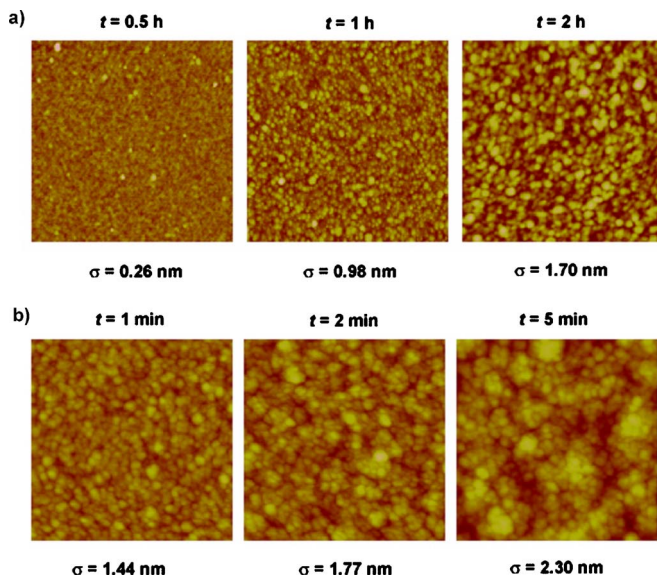


FIG. 1. (Color online) Evolution of surface microstructure for  $\text{SiO}_2$  films grown by PECVD ( $1 \times 1 \mu\text{m}^2$ ): (a) remote configuration and (b) inside the plasma.

the end of the discharge and the second one with the samples inside the plasma. Note that for the range of pressures used (10–100 Pa), the mean free path of neutral species is around 1–5 mm depending on the cross section.

Mixtures of  $\text{O}_2/\text{TMS}$  were used for the synthesis of the films. Typical working conditions were 15 SCCM (SCCM denotes cubic centimeter per minute)  $\text{O}_2$ , 5 SCCM TMS, and a working pressure of 10 Pa. In some experiments, a total working pressure of 100 Pa was also used by keeping the ratio between the flows of  $\text{O}_2$  and TMS constant. A power of 120 W was used for all the deposition experiments. The thin films obtained were characterized by Fourier transform infrared and x-ray photoemission spectroscopy. The results obtained will not be presented here, but in all cases, they indicate that a  $\text{SiO}_2$  composition was obtained.

The surface topography was analyzed by means of an atomic force microscope (AFM) Dimension 3100 instrument of Digital Instruments. Images were obtained in tapping mode using Al-backcoated Si cantilevers. The roughness of the films was calculated as the root mean square of the heights of the different surface points. This parameter, taken as a representative measure of the interface width, is valid as long as the higher moments of the height distribution are small. Histograms were taken for all the surfaces and, in all cases, they were fitted to Gaussian functions within the experimental error. In this way, the validity of surface roughness values as a measure of the interface thickness is ensured. Images taken at different points of the surface were used in order to improve the statistics and representativity of the values obtained.

The scaling properties of the sample were analyzed by determining both the power spectral density (PSD) of the surface<sup>17,29</sup> and the height-height correlation function.<sup>10</sup> Different values were obtained in each case. Curves were obtained for samples grown for different deposition times for both remote and in-plasma preparation conditions. Estima-

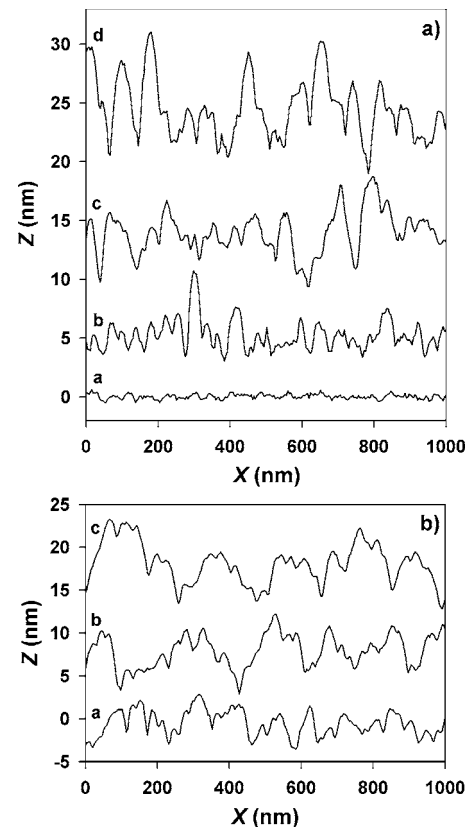


FIG. 2. Representative line profiles obtained for different deposition times: (a) remote configuration (a, 30 min; b, 1 h; c, 2 h; d, 3 h) and (b) inside the plasma (a, 1 min; b, 2 min; c, 5 min).

tion of the  $\alpha$  coefficient was also done by using the image-partitioning technique.<sup>30</sup>

The  $\beta$  exponents were calculated by direct fitting of the variation of surface roughness with deposition time. The dynamic exponent  $z$  was calculated from the variation with time of the crossover length between the two asymptotic behaviors of the height-height correlation function as defined in the previous section.

#### IV. RESULTS

In Fig. 1, the evolution of surface topography is presented for films grown in the remote configuration and inside the plasma. It can be seen in this figure that the time scales considered for preparation of the samples and recording of the images are very different for the two types of samples. This is due to the fact that the growth rate inside the plasma is higher than in the remote configuration (100 and 3 nm/min, respectively).

The films present a granular microstructure with similar grain sizes. In the case of the films grown inside the plasma, the grains seem to be gathered into higher order structures. This is better appreciated in Fig. 2, where line profiles along the images are presented. In the samples grown inside the plasma, there seems to be two length scales: the first one defined by the height fluctuations of the different grains and the second one formed by small aggregates of these grains. It

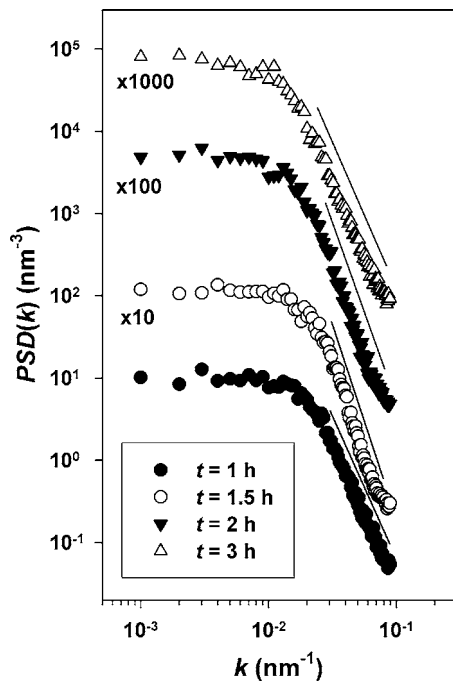


FIG. 3. Power spectral density plots obtained for the SiO<sub>2</sub> thin films grown in the remote configuration for different deposition times. The straight lines are a guide for the eyes.

is interesting to note that despite the differences in growth rate, the height oscillations defining the surface roughness fit in the two situations within the same scale. It is also interesting that, when comparing the profiles a and b in Fig. 2(a), it is possible to see the initial stages of development of the surface topography. Such an initial texturing of the surfaces is not appreciated in the profiles in Fig. 2(b) corresponding to the samples grown inside the plasma. This is likely due to the much higher growing rate inside the plasma that precludes the possibility of observing experimentally the initial stages of the thin film growth.

Interesting results about the surface roughness evolution are obtained when studying the PSD of the films. According to the scaling theory briefly introduced in Sec. II, the roughness exponent  $\alpha$  can be calculated from the slope  $i$  of the linear region of  $PSD(k)$  at low length scales as  $\alpha = (i - d) / 2$ , being  $d = 1$  for the one-dimensional PSD and  $d = 2$  for the two-dimensional PSD. The index  $i$  is the so-called Fourier index and is also related to the fractal dimension of the surface. For the remote configuration, the evolution of the PSD curves for selected deposition times is presented in Fig. 3. The values of the  $\alpha$  exponent in each case are plotted in Fig. 4(a). By looking at the slopes of the PSD curves in Fig. 3 and the evolution of the corresponding values of  $\alpha$  reported in Fig. 4(a), it appears that this parameter does not remain constant for different deposition times as it is predicted by the conventional DST.<sup>9</sup>

In Fig. 4(b), the values of the  $\alpha$  exponents calculated for the same samples by using the height-height correlation function are presented as a function of deposition time. This plot shows that the calculated  $\alpha$  values are almost two times smaller in this case (i.e., 0.7–0.9) as compared with those calculated from the PSD [i.e., between 1.3 and 2, note the

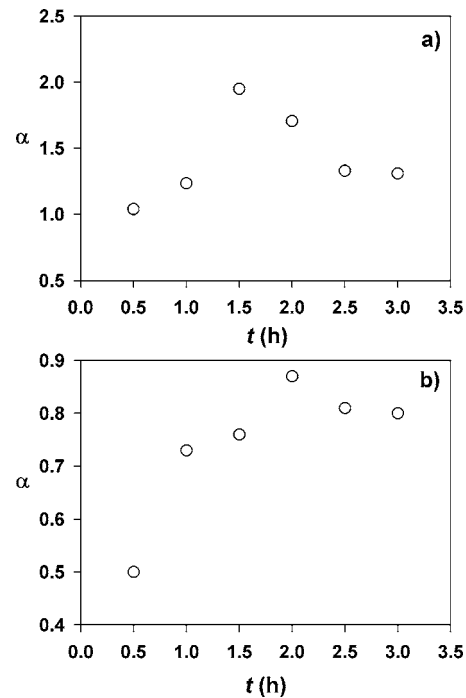


FIG. 4. Values of the roughness exponents of the SiO<sub>2</sub> films grown in the downstream configuration at different deposition times calculated from (a) the power spectral density and (b) the height-height correlation function.

different scales in Figs. 4(a) and 4(b)]. It is also interesting that the change with deposition time is less important for the values calculated from the height-height correlation function.

The different slopes in the points presented in Fig. 3 prevent the coalescence of the power spectral density and the height-height correlation function, the single normalized curve consequence of the scaling relation [Eq. (1)]. Therefore, although the surfaces can be considered self-affine, the scaling behavior observed does not correspond to that defined by the Family-Vicsek scaling relation.

A second interesting aspect is the existence of different exponents depending on whether they are calculated from the PSD or the height-height correlation function. According to Fig. 3, the power dependence of the power spectral density with the wave number  $k$  appears for wave numbers ranging from  $10^{-1}$  to  $10^{-2}$  nm<sup>-1</sup>, that is, to length scales of 10–100 nm, greater than those corresponding to the height-height correlation function.

Therefore, the different exponents obtained from these two techniques can be explained by assuming the existence of different behaviors at different length scales. Thus, global and local exponents would exist for, respectively, long and short length scales. Under this assumption, the exponents obtained from the PSD would reflect the global scaling of the surface, whereas those obtained from the height-height correlation function would reflect the so-called local scaling. Such a behavior has been referenced previously in the literature and attributed to a situation of an anomalous scaling.<sup>26</sup>

The films grown in the plasma also present different growth exponents when determined from the PSD and the height-height correlation functions, although, as it is evi-

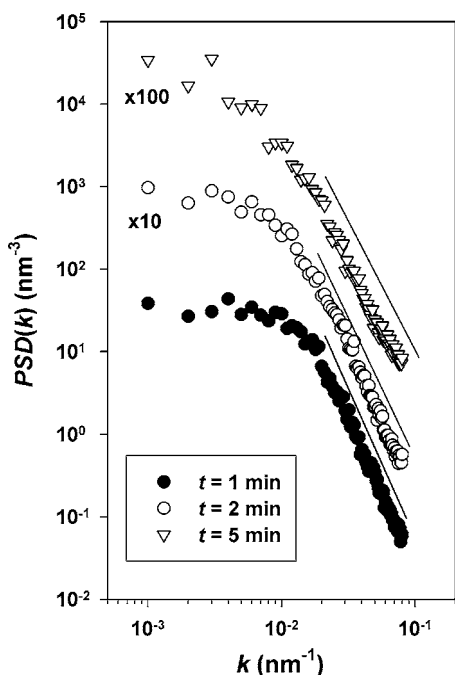


FIG. 5. Power spectral density plots obtained for films grown inside the plasma for different deposition times. The straight lines are a guide for the eyes.

denced by the similar slopes of the straight lines plotted in Fig. 5, there is no time dependence of the roughness exponent obtained from the PSD function. The  $\alpha$  values obtained are  $\alpha=1.3$  for the PSD and  $\alpha=0.7$  for the height-height correlation function, in both cases very close to those obtained in the remote configuration for long deposition times. A possible interpretation of the absence of such strong time dependence of the  $\alpha$  exponent for the films grown inside the plasma is that the variable and high values of the  $\alpha$  exponents in the samples prepared in the remote mode [cf. Fig. 4(a)] are associated with the existence of an initial transient stage of development of the surface topography. Owing to the different deposition rates for the two experimental configurations, such a stage can only be detected for the remote samples, but not for the samples grown inside the plasma. It is also likely that this initial transient stage detected in the former case is characterized by a situation where individual grains are still growing independently.

Fluctuations of the  $\alpha$  exponent with time and, consequently, an anomalous behavior were, for example, detected by Zhu *et al.*<sup>27</sup> during the IBAAD growth of diamondlike films. The working conditions used with the IBAAD technique are different from those used in the present work, where the energy of the incoming species to the surface would be much smaller, i.e., close to that of a Maxwellian energy distribution. One of the general interpretations given in the literature for the existence of different global and local exponents that would be compatible with our working conditions is that nonlocal interactions due to shadowing effects may be responsible for the appearance of an anomalous behavior.<sup>23</sup> A similar explanation can be invoked here to account for the surface topography developed by the samples prepared under our two experimental conditions. In the two cases, after long

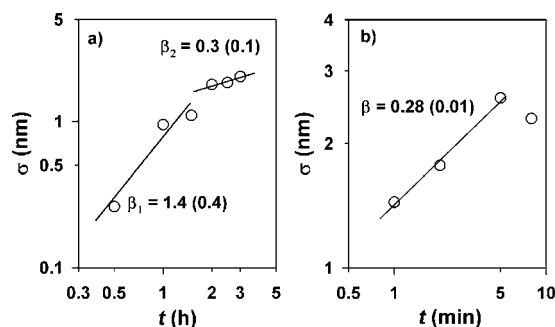


FIG. 6. Evolution of surface roughness with deposition time of  $1 \times 1 \mu\text{m}^2$  AFM images: (a) remote configuration and (b) inside the plasma.

deposition times, this topography is characterized by a well developed granular microstructure and a similar magnitude of the surface roughness. In this regard, it is also reasonable to assume that during the transient stage found for the remote samples, the observed anomalous scaling is related to a kind of columnar growth of individual grains.

The study of the variation of the surface roughness with the deposition time further supports the existence of a transient behavior for the samples grown under a remote configuration. As it can be seen in Fig. 6, two regions characterized by different values of the  $\beta$  exponent can be distinguished for the films grown in the remote mode. At low deposition times, the increase of roughness is very fast (i.e.,  $\beta=1.4$ ), a behavior that would agree with an independent growth of grains of the deposited material. Such a situation is actually evidenced by the AFM image of Fig. 1(a) for  $t = 1$  h, where very small and still separated grains can be observed. Analogously, the line profile b in Fig. 2(a), characterized by sharp and uncorrelated oscillations, would be typical of the roughness defined by individual and uncorrelated grains. Note that, although the surface roughness of the first points in Fig. 6 is very small, the minimum surface roughness measured is still five times greater than that coming from a flat surface.

After the transient regime, the value of the growth exponent changes to  $\beta=0.3$ . Such an exponent value is similar to that obtained inside the plasma [Fig. 6(b)] and also to that obtained for a remote configuration at higher pressures [cf. Fig. 6(a)], where the growth rate was higher and therefore the transient region cannot be detected. It must be mentioned that an analogous transient behavior characterized by a high value of the growth exponent has been experimentally measured by Dalakos *et al.* during the growth of Si:H thin films by PECVD.<sup>15</sup>

From the height-height correlation function, it is also possible to calculate the correlation lengths of the system for different deposition times [and therefore the dynamic exponent  $z$  defined in Eq. (3)]. From the analysis of the evolution of the correlation length with time, a value of the  $z$  exponent  $z=2.1 \pm 0.2$  is obtained for both the remote and the in-plasma configurations. Note that  $z=\alpha/\beta$  so that, from the values of the local  $\alpha$  exponent obtained ( $\alpha=0.7$ ), values of the growth exponent  $\beta$  of 0.3–0.4 are expected. This is in clear agreement with the values of  $\beta$  determined directly from the ex-

perimental AFM images (cf. Fig. 6), and justifies the differentiation made in Fig. 6 between the transient and the normal regions of growth when the samples are prepared in the remote configuration.

## V. DISCUSSION

### A. Local behavior of thin films and mechanism of growth

The films prepared here by PECVD present a well defined granular microstructure and a complex scaling behavior that cannot be explained in terms of the Family-Vicsek relation. An interesting finding is that for long deposition times and the two experimental configurations (i.e., inside the plasma and remote), the samples present a similar surface roughness characterized by equivalent values of the scaling exponents. The obtained values of the local exponents,  $\alpha=0.7$  and  $\beta=0.3$ , are close to those corresponding to the nonlinear molecular beam epitaxy universality class of Das Sarma–Tamborenea.<sup>31</sup> This suggests that the fragments of precursor species arriving to the surface have a certain mobility. This mobility would be higher than that found for the silane precursor,<sup>5</sup> but not as high as that assumed in the case of TEOS, where very smooth and conformal films are obtained.<sup>6</sup> Such intermediate behavior for the mobility of the incoming fragments is to be expected for TMS molecules or partially oxidized TMS fragments, as weakly bonded physisorbed states are expected when dealing with a complex molecular structure of this silicon precursor.<sup>5</sup>

Owing to the similar scaling exponents found after long deposition times for the two experimental configurations, it is also likely that similar types of plasma species are reaching the surface of the films under the two experimental conditions. Thus, in our experimental system, the concentration of active species in the plasmas would be the main factor controlling the growth rate, while the type of surface microstructure would mainly depend on the type of the species reaching the surface.

In previous works reporting the growth of SiO<sub>2</sub> thin films by PECVD using several precursor molecules, partially oxidized fragments of the precursor are supposed to incorporate to the films after condensation reactions with –OH terminal bonds on the surface.<sup>32</sup> In a previous work with plasmas of the TMS/O<sub>2</sub> mixture under slightly different experimental conditions than those used here, we have been able to detect by mass spectrometry and optical emission spectroscopy the formation in the plasma of SiO<sub>x</sub>(CH<sub>3</sub>)<sub>3-x</sub> and Si(OH)<sub>x</sub>(CH<sub>3</sub>)<sub>3-x</sub> fragments.<sup>33</sup> It is likely that these or similar TMS fragments are mainly responsible for the incorporation of silicon to the films, while the activated species of oxygen also reaching the surface would oxidize these adsorbed fragments and lead to the formation of SiO<sub>2</sub> as experimentally found.

### B. Anomalous scaling of SiO<sub>2</sub> thin film growth

Regarding the kinetic roughening of the films, two relevant experimental results deserve a specific comment: (i) obtaining two different values of the growth exponent  $\alpha$  from the height-height correlation function and the power

spectral density and (ii) the variation with deposition time of the roughness exponent  $\alpha$  for the samples grown in the remote position. The first result would, in principle, contradict the definition of a self-affine interface, since the asymptotic behaviors of  $H(L)$  and PSD( $k$ ) shown in Sec. II are both characteristic of self-affine fractal objects. However, following the interpretation of the experimental results provided in the previous section, this result could be explained in terms of an anomalous scaling behavior, so that each method would describe the scaling properties of the surface at different length scales. Regarding the second result, the variation with time of  $\alpha$  implies that, contrary to what it is commonly observed in the literature, the surface processes taking place during surface growth do not preserve the fractal properties of the surface, at least for shorter deposition times.

As it was mentioned in Sec. II, the anomalous scaling can be related either with the presence of nonlocal effects during surface growth or it can be an intrinsic anomalous scaling. The granular microstructure developed in our films and the experimental conditions used prompt us to propose that shadowing effects are the main factors responsible for the anomalous behavior found here. In fact, well developed granular microstructures are commonly observed in many systems depicting an anomalous scaling.<sup>20,27,34</sup> This type of microstructures is also expected for the relatively high pressures used for the synthesis of thin films in our PECVD reactor. Shadowing was also claimed by Ojeda *et al.*<sup>17</sup> to account for transient behaviors found during thermal CVD growth of SiO<sub>2</sub> films.

According to a previous theoretical model confirmed by Monte Carlo simulations and experimental results,<sup>24</sup> shadowing effects in films prepared by PECVD become more important when the plasma species arrive to the surface according to nonperpendicular directions. According to this model, the reactive species created inside the plasma are thermalized by successive collisions and reach the surface with an angular distribution probability, which is proportional to the cosine of the angle with the normal vector of the surface. Thermalization would be the result of the relatively low mean free path of the plasma species (i.e., 10<sup>-3</sup>–10<sup>-4</sup> m for the range of pressures used here, admitting a common value of elastic cross section for collision between molecules of 50 × 10<sup>-20</sup> m<sup>2</sup>). For this off-normal arrival of particles, well developed granular microstructures may appear as a combination of the nonperpendicular incidence of the incoming particles and the lateral growth of the grains.<sup>24</sup> Eventually, some re-emission of particles could also contribute to such granular microstructures.<sup>10</sup>

A well defined columnar growth, even for relatively thick films, is typical of evaporated films.<sup>35</sup> The main difference with respect to the PECVD films is that in this latter case, the off-perpendicular arrival of particles will also contribute to the lateral growth of the grains and, eventually, to their coalescence. Before coalescence, during the initial stages of growth, the microstructure is strongly affected by shadowing. Different results of our scaling analysis support such an assumption. During this initial transient regime, the grains would grow in a noncorrelated way from the thin film nuclei formed initially. Such an independent growth would agree with the high values of the  $\beta$  exponent measured for short

deposition times (cf. Fig. 6) that are typical of a columnar microstructure.<sup>23</sup> As the deposition time increases, the lateral growth of the grains favored by the nonperpendicular arrival of the plasma particles would cause the observed coalescence of the grains and hinder the appearance of separated columns. The influence of such initial growth instabilities would be observable at higher length scales (i.e., global  $\alpha$  exponent), reflecting the fact that before coalescence, the growth of each grain can be considered independently from that of the other grains. A similar explanation was given by Dalakos *et al.* to explain the transient behavior of a high  $\beta$  exponent found during PECVD of Si:H films.<sup>15</sup>

## VI. CONCLUSIONS

According to the results obtained, the growth of SiO<sub>2</sub> thin films by PECVD from a TMS precursor is described by an anomalous scaling behavior characterized by two features: (i) the height-height correlation function and the power spectral density provide different values of the roughness exponent  $\alpha$  and (ii) a time variation of the roughness exponent is observed.

The films grown in a remote configuration are characterized by a transient regime for short deposition times. The odd values of the growth and roughness exponents characterizing these initial stages are in agreement with a columnar growth of uncorrelated grains. The possibility that the plasma particles arrive to the surface in nonperpendicular directions is a key factor favoring this initial columnar growth.<sup>24</sup> Once these independent SiO<sub>2</sub> grains grow sufficiently both in height and laterally, they reach coalescence, a

point that defines a change from the transient regime to another one whose local scaling parameters are similar to the nonlinear molecular beam epitaxy universality class of Das Sarma–Tamborenea.<sup>31</sup>

The use of TMS as a precursor for the PECVD synthesis of SiO<sub>2</sub> thin films leads to the production of active plasma species of the types SiO<sub>x</sub>(CH<sub>3</sub>)<sub>3-x</sub> and Si(OH)<sub>x</sub>(CH<sub>3</sub>)<sub>3-x</sub>.<sup>33</sup> It is likely that these or similar plasma species arriving to the surface have an intermediate surface mobility as compared with that of the particles formed from silane and TEOS. This intermediate mobility is likely one of the reasons leading to the observed mechanism of growth found after the initial transient regime. In this regard, while the rate of growth of the thin films depends on the concentration of the active species in the plasma, the type of roughness and, in general, the microstructure of the films should be mainly connected with the type of precursor species arriving to the surface.

Finally, the fact that the height-height correlation function and the power spectral density have provided different values of the roughness exponent  $\alpha$  emphasizes the importance of using different methods to obtain a better picture of the kinetic roughening process and the scaling properties of surfaces.

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