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Measurement of critical exponents of diamond films by atomic force microscopy imaging

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We have synthesized diamond films by plasma assisted chemical vapor deposition on silicon substrates. The roughness and dynamic critical exponents of these films have been measured using an atomic force microscope. [S1063-651X(98)14510-5]

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The growth of films by deposition is clearly of great technological interest. Fluctuations in the height $h(\mathbf{x},t)$, the surface location $\mathbf{x}$, and time t can be measured directly using a scanning probe microscope or indirectly by scattering. Analytical and numerical treatments of simple growth models suggest that, quite generally, the height fluctuations have a self-similar character and their average correlations exhibit a dynamic scaling form, named the Family-Vicsek scaling relation [1]. The roughness and dynamic exponents α and z, defined by this relation, are expected to be universal, depending only on the underlying mechanism that generates self-similar scaling [2]. The determination of the critical exponents α and z is one of the fundamental problems of statistical mechanics. Considerable effort, both theoretical and experimental, have been made to investigate the surface growth process. Many references on the subject are found in the excellent review of Barabási and Stanley [3]. Theoretical discrete models provided a substantial fraction of the driving force behind early investigations of the interface morphology. Discrete models, numerical simulations, and stochastic differential equations have been used to explain the growth mechanisms on d-dimensional substrates. Such equations typically describe the interface at large scales and times, which means that we neglect the short-range scale details and focus only on asymptotic coarse-grained (hydrodynamic) variables. We can, in some sense, say that for $d=1$ the growth phenomenon is reasonably well understood. Moreover, for $d>1$, there are many challenging problems that seem insurmountable. Numerical simulations are generally impracticable or extremely difficult and numerical integrations are questionable, leading to somewhat inconclusive results [2,3]. On the other hand, more detailed and accurate measurements of critical parameters of thin films are still lacking and experimental confirmation of dynamic scaling is scarce [2,4]. In this work we synthesized diamond films by microwave plasma assisted chemical vapor deposition and we measured the roughness and dynamic exponents of these films using an atomic force microscope (AFM).

A description of our equipment can be seen elsewhere [5,6]. The substrate for the films was silicon that had been scratched by 1-μm diamond powder and cleaned in an acetone ultrasonic bath. The following growth parameters were fixed: a 300-SCCM hydrogen flow rate (where sccm denotes cubic centimeter per minute at STP), 1.5-SCCM methane flow rate (0.5-vol % methane in hydrogen), 70-torr chamber pressure, 820 °C substrate temperature, and a nominal 850-W microwave power. The polished silicon substrate was divided into small pieces and eight films were synthesized with different growth times: 17, 20, 24, 26, 34, 48, 63, and 74 h.

The most economical way to characterize self-affine roughness is by appealing to a dynamic scaling form [1,2]. In many situations, we do not have information on the dynamics of the growth, nor do we have the possibility of producing interfaces with different sizes. Suppose that the only data we have are collected at the final stage of the experiment, consisting of the values of the height $h(\mathbf{x},t)$ of the interface, at different points $\mathbf{x}$ and times t. In this situation, we study the scaling of the ''local width'' or ''local roughness'' $w_L(l,t)$ defined by [1]

$$w_L^2(l,t)=\langle[h(\mathbf{x},t)-h_l(\mathbf{x},t)]^2\rangle_{\mathbf{x}}, \tag{1}$$

where L is the system size and l means that we select a portion (''window'') of length l on the interface and measure the width and the average height $h_l(\mathbf{x},t)$ in this window. The angular brackets $\langle\ \rangle_{\mathbf{x}}$ denote spatial (over $\mathbf{x}$) and ensemble averages. One can show that $w_L(l,t)$ obeys the scaling form [1–3]

$$w_L(l,t)\sim l^{\alpha}f(t/l^{z}), \tag{2}$$

where $f(u)$ is the scaling function of the argument $u=t/l^z$ and $z=\alpha/\beta$. The parameters α, β, and z are expected to be universal parameters, named critical exponents. For very small times $u\ll1$ we have $w_L(l,t)\sim t^{\beta}$ when the width grows as t^{β} and the different sites of the surface are practically independent. As time increases, different sites of the interface begin to be correlated. The typical distance over which the heights ''know about'' each other, the characteristic distance over which they are correlated, is called the correlation length, denoted by $\xi(t)$, which increases as $\xi(t)\sim t^{1/z}$. When correlations are significant we have $w_L(l,t)\sim l^{\alpha}$ for $l\ll\xi$; for $l\gtrsim\xi$ the width saturates, that is, $w_L(l,t)\cong\text{const}\sim\xi^{\alpha}$. In these conditions [3] the fractal dimension d_f of the film interface is given by $d_f=3-\alpha$. In the very long time limit, that is, $u\to\infty$, we expect that the width reaches its maximum (saturation) value $w_{\text{sat}}(L)\sim L^{\alpha}$. In what follows, omitting for simplicity the index L, the width will be indicated by $w(l,t)$; l will be measured in micrometers and w in nanometers.

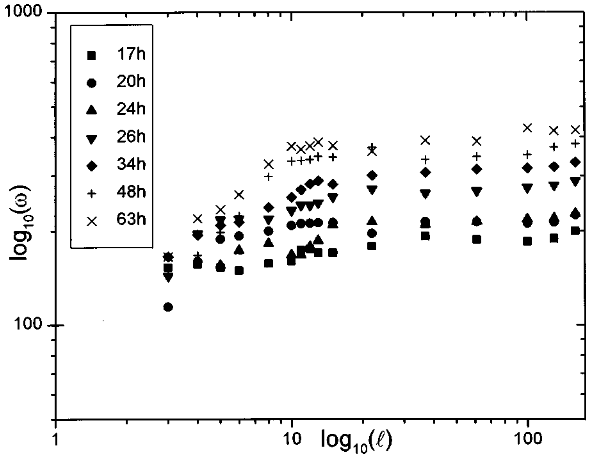

FIG. 1. Typical values of the roughness $w(l,t)$ as a function of the length l and of the growth time t; $w(l,t)$ is measured in nanometers, l in micrometers, and t in hours.

The roughness and fractal dimensions of the seven diamond films have been measured using an AFM, the NanoScope IIIA from Digital. We have analyzed about ten different regions of each sample, each one with 160×160 μm^2. With the zoom facility of the AFM, we have divided these (160×160)-μm^2 regions in smaller ones (windows) with $l\times l$ μm^2, l going from 3 up to 160 μm, and measured their local roughness $w(l,t)$. We must note that it was not possible to analyze the 74 h sample: Due to the formation of large microcrystalline diamond grains, about 8.5 μm in diameter, we found $w\sim 600$ nm, exceeding the z limit of the AFM. The average grain sizes are about 2.8 and 7.0 μm for 17 and 63 h, respectively.

Shown in Fig. 1 for each film are typical values of $\log_{10}w(l,t)\log_{10}(l)$ as a function of the growth time t. We verified that for $t<34$ h, the correlations between different sites of the interface are small and the α power law growth is not well defined.

To determine the exponent α we take into account only the roughness of the 63-h sample. We have analyzed 12 different regions of this sample. Shown in Fig. 2 is $\log_{10}[w(l,t=63\text{ h})]\log_{10}(l)$ for these 12 regions. We see that $\log_{10}(w)\log_{10}(l)$ shows clearly the α power law growth, which appears as straight lines from 3 μm up to $l=\xi\cong 15$ μm, where ξ is the correlation length. For $l\geqslant\xi$, the width saturates, that is, $w(l,t)\cong\text{const}$ according to Eq. (2), and is given by $w\simeq 400$ nm. With a straight-line best fit [7] we have found the following α values for 12 regions: 0.36 ± 0.06, 0.49 ± 0.04, 0.51 ± 0.04, 0.51 ± 0.08, 0.51 ± 0.03, 0.53 ± 0.03, 0.53 ± 0.06, 0.58 ± 0.07, 0.64 ± 0.08, 0.66 ± 0.11, 0.66 ± 0.08, and 0.70 ± 0.10. To analyze the goodness of the straight-line best fit we have followed the standard statistical procedure [7] calculating the χ^2_ν. The χ^2_ν values go from 0.97 up to 0.99, showing that our fit is appropriate to describe the data. From the above α values we get $\alpha=0.56\pm 0.09$.

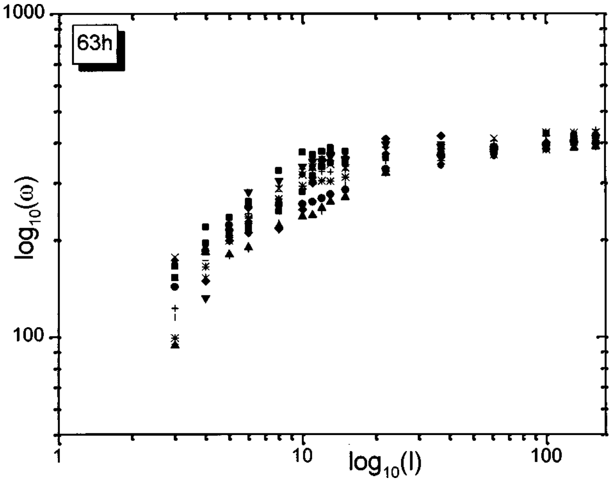

FIG. 2. Roughness $w(l,63\text{ h})$ as a function of the length l. The 12 different symbols seen for each curve correspond to the 12 different regions analyzed.

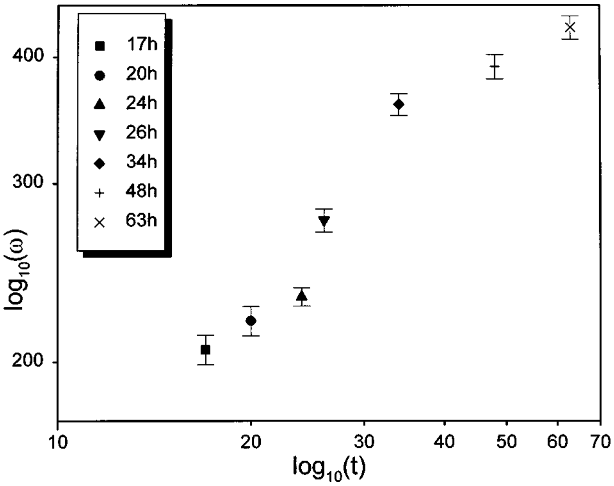

FIG. 3. Roughness $w(t)$ as a function of the growth time t.

The fractal dimension d_f of the 63-h sample was determined using the fractal algorithm of the AFM software, where the surface is analyzed by triangulation. We have considered only the (160×160)-μm^2 images. The following values were found for d_f: 2.43, 2.45, 2.46, 2.46, 2.47, 2.47, 2.48, 2.49, 2.49, 2.49, 2.50, and 2.50. So d_f would be given by $d_f=2.48\pm 0.02$ and, consequently, as $\alpha=3-d_f$, α is given by $\alpha=0.52\pm 0.02$. This result is in good agreement with that obtained using the width measurement.

Shown in Fig. 3 is $\log_{10}w(t)\log_{10}(t)$ for $t=17$, 20, 24, 26, 34, 48, and 63 h. Since for small growth times [1] we must have $w\sim t^\beta$, to determine the critical exponent β we take into account the roughness of the samples only for small growth times, that is, 17, 20, and 24 h: w (17 h)$=205.72\pm 6.90$ nm, w (20 h)$=219.63\pm 7.24$ nm, and w (24 h)$=232.01\pm 4.87$ nm. These values have been estimated by the average $w(t)=[w(100\ \mu\text{m},t)+w(130\ \mu\text{m},t)+w(160\ \mu\text{m},t)]/3$. With a straight-line best fit [7], the growth exponent is given by $\beta=0.34\pm 0.02$.Also in this case, estimating the variances and χ^2_ν we verified that $\chi^2_\nu=0.99$, showing that our straight-line fit is appropriate to describe the data.

Taking into account the above values obtained for α and β, we get $z=\alpha/\beta=1.65\pm 0.28$. Consequently $\alpha+z=2.21\pm 0.30$, which satisfies the condition $\alpha+z=2$, within the experimental errors. This could be an indication that the growth of diamond films is governed by the Kardar-Parisi-Zhang (KPZ) equation [2,3,8,9]. However, according to the KPZ predictions, for $d=2$ ballistic deposition [2], $\alpha\simeq 0.38$

and $\beta \simeq 0.24$. We have obtained $\alpha \simeq \frac{1}{2}$ and $\beta \simeq \frac{1}{3}$, which are critical exponents predicted by the KPZ equation for $d=1$. This could be a support for the superuniversality conjecture [10]. On the other hand, the above discrepancies could be produced by the formation of diamond microcrystallites and, in this case, a somewhat different equation would govern the growth of diamond films, as proposed by Hwa, Kardar, and Paczuski [11]. We intend to analyze these aspects in the future.

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