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A comparative study of correlation methods for determination of fractal parameters in surface characterization

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ABSTRACT
An analysis of several methods of extraction of fractal parameters from the simulated, artificial surfaces and AFM images of the real, polycrystalline diamond films is presented in the paper. The methods involve the cube count method, the roughness method, the autocorrelation function method, and the structure function method. By comparing the four methods, the roughness method is found to be superior for its high numerical accuracy, whereas the cube count method appears to be inferior in that aspect. The changes in the fractal dimension and the anisotropy ratio values observed over deposition time are also shown and discussed in the paper.

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1. Introduction

Precise description and control of 3D surface topography is of prime importance in engineering applications as it has strong influence on such materials properties as: fracture toughness, wear resistance, lubrication and others. Currently, surface characterization basically relies on a large series of statistical parameters derived using various methods, among which atomic force microscopy (AFM) now became one of the most popular. Since its discovery, AFM significantly evolved toward characterization of surface topography of solids of all types down to the nanoscale level, and nowadays this non-destructive method provides topographical information probed over an area from several square nanometers up to thousands of square micrometers. Moreover, this method can discriminate among a large diversity of interactions occurring between the surface and the scanning tip (electrical, magnetic, adhesive, friction, etc.).

Surface topography can be characterized using an excessive number of statistical parameters depicting various aspects of the surface lay, roughness, waviness and the form. Unfortunately, many of them strongly depend on how they are actually measured, including for instance the sampling and the scan lengths, and the instrumental resolution. In order to overcome this problem, description of engineering surfaces in terms of fractal geometry was suggested [1]. Fractals are virtual, self-similar geometrical objects that appear identical independent of the scale of magnification. Such objects are characterized by the fractal dimension \(D\) [1]. However, due to some physical confinements (finite instrumental resolution, finite observation time), fractal properties of objects of natural origin are often reduced to a limited range of scale lengths. Such objects (called self-affine) need to be described by three parameters: fractal dimension \(D\), corner frequency \(f_c\), and the topothesy \(A\), and differ from perfect fractals described by the fractal dimension \(D\) solely.

Fractal dimension is found to be correlated with surface roughness parameters [2–5], it is related to various material properties [6], and even to mechanisms leading to surface formation [7]. On the other hand, several experimental methods have been proposed for estimation of fractal dimension, including for example: AFM, Scanning Electron Microscopy [8], diffuse X-ray reflectometry [8], adsorption measurements, electrochemical impedance spectroscopy and others, which usually hardly converged into a consistent picture. What is worse, a little is known about possible influence of each numerical procedure on final results. In previous work we have compared the effect of the AFM tip geometry and the scan mode on results of a fractal analysis of well-established surfaces (calibration gratings) [4]. In this study we report the systematic comparison of different numerical procedures used to estimate the fractal dimension from the same AFM images recorded from the crystal surface that evolves with elapsed time. Such an attempt should exhibit numerical peculiarities allowing the quantitative assessment of processing procedures, which might be especially useful in fulfilling lacking knowledge about their accuracy, precision and terms of applicability.

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The selection of thin diamond films grown by microwave plasma was motivated by several reasons. First, the deposition process significantly affects the crystal morphology that starts from a bare substrate surface with tiny nuclei, and evolves into a closed although polycrystalline film with increasing roughness, which is expected to influence the fractal properties. In addition, a little is known about fractal properties of diamonds itself. Salvadori et al. [9] reported D value close to 2.5 for plasma-deposited diamond films on silicon substrates, whereas Silva et al. [10] obtained lower D equal to 2.2 for boron-doped diamond films grown by the hot-filament CVD method. Recently, Tsyrar [11] found fractal dimension equal to 2.36, and 2.73 for [111]- and [001]-oriented diamond grains in HF-CVD films, respectively. Finally, a modeling of the topography evolution in terms of fractal parameters might shed some light onto the kinetics of the growth process, including the early stages of the nucleation phenomena, and subsequent diffusion of contaminants and defects along grain boundaries.

2. Materials and methods

Diamond films were deposited on 1 mm thick substrates made of fused quartz. Pre-treatment procedure involved mechanical seeding with 250 nm diamond powder on a vibrating plate. The growth process was carried out in a microwave plasma CVD reactor (ASTeX AX 6590) described in details elsewhere [12]. Gas mixture contained methane largely diluted with molecular hydrogen (CH4/(CH4 + H2) = 5% (vol.)) Other deposition parameters were as follows: substrate temperature 900 °C, gas pressure 6650 Pa (50 Torr), microwave power 3000 W, and deposition time varying from 5 min to 5 h.

AFM measurements were carried out at ambient conditions using Multimode 8 instrument with Nanoscope V controller (Bruker). The tip (SNL-10 (Bruker)) with the radius 2 nm scanned across the surface in a contact mode. To determine the fractal properties, square AFM images with the lateral resolution of 512 points and scan lengths from 1 up to 150 μm were taken. The images were then flattened to remove line tilt and image bow prior to further numerical processing.

3. Evaluation of the fractal dimension

3.1. The cube counting method

An evaluation of the fractal dimension by counting the cubes directly explores the definition of a box-count dimension. The algorithm iteratively halves an initial cubic cell with the edge length L equal to the scan length into smaller cubes, and counts N(L) – the number of all cubes that contain at least one sample of a 3D topography. The process continues until L approaches the image resolution, i.e. the distance between two adjacent samples [13]. Since:

\[ N(L) \propto L^{-D} \]  

(1)

the slope of a log–log plot of N(L) versus L gives the fractal dimension referred to as the cube count fractal dimension D_{CC}.

3.2. The roughness method

The fractal dimension can be also estimated using the root-mean-squared value of the surface height variance S_q defined as [14,15]:

\[ S_q = \sqrt{\frac{1}{N_xN_y} \sum_{i=0}^{N_x-1} \sum_{j=0}^{N_y-1} (z(i,j) - \langle z \rangle)^2} \]  

(2)

where \( \langle \cdots \rangle \) denotes mean value, N_x, N_y is the number of samples along rows and columns in the AFM image, while \( z(i,j) \) is the measured height in pixel \((i,j)\) of an image. Assuming that the roughness \( S_q \) measured over surfaces with different edge lengths L scales as:

\[ S_q \propto L^{3-D} \]  

(3)

the fractal dimension D_{RMS} can be computed from the slope of a least-square regression line fit in a log–log plot of \( S_q \) vs. \( L \) [16].

3.3. The structure function method

The surface topography recorded in the form of discrete height samples \( z(i,j) \) in an AFM image allows us to compute the three-dimensional structure function (SF) defined as:

\[ S(x,y) = \langle (z(x,y) - z(x+\tau_x,y+\tau_y))^2 \rangle \]  

(4)

\[ \tau_x = \frac{L_x}{N_x} \quad (0, 1, \ldots, N_x - 1), \quad \tau_y = \frac{L_y}{N_y} \quad (0, 1, \ldots, N_y - 1) \]

where \( \langle \cdots \rangle \) denotes the spatial average, \( N_x, N_y \) – are the numbers of samples along scan axes, \( L_x, L_y \) – scan lengths, whereas \( (\tau_x, \tau_y) \) – the discrete spatial lag along scan axes between an original image and its delayed copy. Any profile of the structure function derived from the image is assumed to obey the approximate scaling-law behavior:

\[ S(x,y) = A \cdot x^{(2-D)} \]  

(5)

where \( D \) is the profile fractal dimension, while \( A \) – is the topohesy. According to Wu [17], the topohesy can be expressed explicitly as:

\[ A = \frac{\pi (D-1)}{2D(5-2D) \sin[\pi(2-D)]} \]  

(6)

where \( G \) is a scale constant with the dimension of reciprocal length, \( D \) – is the profile fractal dimension (1 < D < 2), and \( \Gamma \) – is the Euler function.

Note that any section through the structure function at an arbitrary angle around the origin would be equivalent to an ensemble average of profile structure functions measured at the angle \( \theta_r \) with respect to the x-axis:

\[ \theta_r = \tan^{-1} \left( \frac{\tau_y}{\tau_x} \right) \]  

(7)

The profile fractal dimension \( D \) is then calculated from the least-square regression line in a log–log plot of the profile structure function versus separation lag for each \( \theta_r \) angle, and the surface fractal dimension D_{SF} is computed according to:

\[ D_{SF} = (D) + 1 \]  

(8)

where \( \langle \cdots \rangle \) denotes tangentially averaged mean fractal dimension. Likewise, the topohesy \( A \) can be determined from the intercept of the above plot with the y-axis.

3.4. The autocorrelation function method

According to Nayak [18], any surface represents a real random process with the spatial variation described by the autocorrelation function (ACF). Assuming stationarity and ergodicity, the ACF can be computed through spatial averaging over a limited number of AFM samples. The stationarity condition requires that both the autocorrelation function and the mean value are independent of the position, whereas the ergodicity requires the mean to converge to a constant value with increasing sampling period. Even surfaces with apparent curvature and waveform (i.e. non-stationary) can be
treated in this manner providing that the AFM image is previously flattened. The autocorrelation function is defined as:

\[ R(t_x, t_y) = \frac{1}{\sigma^2} \langle (z(x, y) - \bar{z}) \cdot (z(x + t_x, y + t_y) - \bar{z}) \rangle, \]

\[ t_x = \frac{L_x}{N_x} (0, \ldots, N_x - 1), \quad t_y = \frac{L_y}{N_y} (0, \ldots, N_y - 1) \]

where \( \langle \ldots \rangle \) denotes the mean value, \( \sigma \) is the root-mean-square surface roughness (normalizing term), \( N_x, N_y \) – the numbers of samples along scan axes, \( (t_x, t_y) \) – the discrete spatial lag along scan axes, \( L_x, L_y \) – the scan lengths.

The ACF exhibits several useful properties; it is a real, symmetric function with the maximum value at the origin (zero lag), follows the periodicity of the surface, asymptotically decays toward zero with increasing spatial lag, and can be computed using very efficient FFT algorithms. Apart from that, the ACF gives insight into the surface anisotropy using the ACF decay lengths along various orientations. For isotropic surfaces, the decay lengths are equal in all directions, whereas for anisotropic surfaces the decay is faster across the lay orientation (\( a1 \) direction) than along it (\( a2 \) direction). Hence, the normalized anisotropy ratio \( S_t \) is defined as [14]:

\[ 0 < S_t = \frac{t_{a1}}{t_{a2}} \leq 2 \leq 1 \] (10)

where \( t_{a1} \) and \( t_{a2} \) – are the smallest and the largest decay lengths, respectively. For \( S_t > 0.5 \) the surface is said to be isotropic, while for \( S_t < 0.3 \) the surface is said to be strongly anisotropic [1].

Assuming that the ACF is Gaussian, and the surface is stationary, there is a straightforward relation between autocorrelation and the structure function [19]:

\[ S(t_x, t_y) = 2 \langle R(0) - R(t_x, t_y) \rangle \] (11)

Having the SF derived from the ACF and calculated for various \( \tau \) directions, it is possible to determine the fractal dimension and the topology according to the method described in Section 3.3. The fractal dimension obtained in that way is referred to as \( D_{ACF} \).

3.5. Comparison of the estimation methods

In order to assess how the numerical accuracy of the discussed methods affects calculated fractal parameters, a set of artificial surfaces has been generated numerically using the equation proposed by Yan and Komvopoulos [20]:

\[ z(x, y) = L \left( \frac{c}{E} \right)^{0.25} \left( \frac{\ln y}{M} \right)^{1.2} \sum_{m=1}^{M} \sum_{n=0}^{\max} y^{(0.3)n} \left[ \cos(\Phi_{mn}) - \cos \left( \frac{2\pi y^n (x^2 + y^2)^{1/2}}{L} \cdot \cos \left( \tan^{-1} \left( \frac{y}{x} \right) - \frac{\pi m}{M} \right) + \Phi_{mn} \right) \right] \] (12)

The surfaces were simulated with the following parameters: fractal dimensions \( D \) equal to 2.25, 2.5, and 2.75, fractal roughness \( G = 0.2 \) pm, the density of frequencies in the profile \( \gamma = 1.5 \), the number of superposed ridges \( M = 10 \), the cut-off length (of the order of around six lattice distances) \( L_5 = 2 \) nm, and homogeneously distributed random phase shift. Simulated surfaces turn out to be relatively flat, with the roughness equal to 74.4 nm, 1.1 nm, and 1.75 pm corresponding to the fractal dimension 2.25, 2.5, and 2.75 respectively.

Having these artificial rough surfaces, the fractal dimension was then back-calculated, and obtained results are summarized in Table 1. Taking into account the numerical accuracy, the roughness method is found to be superior. Indeed, the \( D_{RMS} \) values always hold to the reference ones within the error limit, and the largest relative error does not exceed 3 per cent. The SF method is found to be less accurate, as it is not consistent with the lowest \( D \) value within an error limit, and because its largest relative error approaches 4.5 per cent. As expected, the ACF method is found less accurate than SF method, especially for lower \( D \) values, due to the maximum relative error being as high as 9 per cent. Additionally, the analysis of the surface anisotropy points at distinctive peculiarity of the \( S_t \) value for the surface with \( D = 2.5 \), which is specific of anisotropic surfaces. As noted by Yan and Komvopoulos [20], such an artifact is an intrinsic property of the algorithm due to possible unmatch- ingness of opposing boundaries of the simulated surface, and does not necessarily mean that the ACF method is inappropriate. Finally, the cube count method turns out to be the most inaccurate among the others since it yields the highest relative error equal to 14 per cent.

Note, however, that in this case the relative error increases with increasing \( D \) value, which might suggest that this method is highly sensitive to fractal dimension.

4. Results and discussion

Fig. 1 shows a series of AFM images of a diamond film on quartz substrate at various stages of the deposition process that exhibits significant changes in its topography: Fig. 1A presents initial state, that is flat quartz surface with numerous scratches, Fig. 1B, and Fig. 1C display poor-faceted, minute diamond grains, while Fig. 1D–F present closed diamond layers comprised of well-faceted diamond crystals. Initial seed concentration left after the mechanical pre-treatment is about \( 2 \times 10^6 \) cm\(^{-2}\), and is the same as the nucleation density seen after 5 min of the deposition process. After that, however, the grain density steadily decreases due to increasing competitive crystal growth (average grain diameter raises from about 250 nm up to 1000 nm), approaching finally 0.8 \( \times 10^5 \) cm\(^{-2}\). Another important issue is the grain alignment. As seen in Fig. 1B, shapeless diamond nuclei appear mostly around scratches, and this gives rise to high surface anisotropy. With elapsed deposition time, however, random-oriented diamond grains with sharp edges cover the whole surface diminishing anisotropy of the film.

![Fig. 1C–E](image-url). Surprisingly, competitive diamond growth for 300 min (Fig. 1F) ends up in the linear alignment of the uppermost crystal edges.

Fig. 2 presents the plot of the film thickness (closed circles), and the surface roughness (open circles) against the deposition time. From the beginning, diamond crystals can grow up freely until coalescence occurs somewhere between 5 and 15 min. From that moment crystals grow up competitively since there is no empty

### Table 1

<table>
<thead>
<tr>
<th>( D )</th>
<th>( G ) [m]</th>
<th>( S_t ) [m]</th>
<th>( D_{RMS} )</th>
<th>( D_{ACF} )</th>
<th>( D_{CC} )</th>
<th>( D_{RMS} )</th>
<th>( S_t )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Surf 1</td>
<td>2.25</td>
<td>( 2 \times 10^{-11} )</td>
<td>74.4 ± 10^{-9}</td>
<td>2.31 ± 0.04</td>
<td>2.24 ± 0.04</td>
<td>2.22 ± 0.09</td>
<td>2.24 ± 0.03</td>
</tr>
<tr>
<td>Surf 2</td>
<td>2.50</td>
<td>1.1 ± 10^{-9}</td>
<td>2.52 ± 0.05</td>
<td>2.55 ± 0.01</td>
<td>2.34 ± 0.11</td>
<td>2.49 ± 0.04</td>
<td>0.46</td>
</tr>
<tr>
<td>Surf 3</td>
<td>2.75</td>
<td>1.75 ± 10^{-11}</td>
<td>2.74 ± 0.08</td>
<td>2.72 ± 0.01</td>
<td>2.47 ± 0.08</td>
<td>2.73 ± 0.06</td>
<td>0.85</td>
</tr>
</tbody>
</table>
space left between them. On the other hand, time-dependent changes in the root-mean-square surface roughness measured over an area of $10 \times 10 \mu m^2$, increases from 1.3 nm (quartz substrate) and asymptotically raises within two orders of magnitude up to 133 nm after 5 h of the deposition process.

**Fig. 3A** presents time-dependent changes in the fractal dimension calculated using the four methods described in Section 3: the cube count method ($D_{CC}$), the roughness method ($D_{RMS}$), the structure function method ($D_{SF}$), and the autocorrelation function ($D_{ACF}$). Note that obtained results follow the same path, namely that in the first 30 min the fractal dimension increases, and then it asymptotically falls down approaching a constant value. Unfortunately, each method yields different fractal dimension. Based on the results obtained from the artificial rough surfaces, the $D_{RMS}$ value was chosen as the reference. As can be seen from **Fig. 3A**, the roughness method yields generally higher results than the other methods. More specifically, the $D_{ACF}$ value initially differs less than 1 per cent with respect to the reference, but with increasing deposition time the discrepancy steadily increases up to 10 per cent. On the contrary, the $D_{ACF}$ turns out to be systematically underestimated from 9 up to 20 per cent with respect to the reference. Similar behavior is observed for the $D_{CC}$, which is at least 11 and not more than 17 per cent less than the $D_{RMS}$ value. In addition, changes in $D_{CC}$ are relatively small, which agrees well with low sensitivity of this method observed for the simulated surface.

In order to verify, how the fractal dimension depends on the scan length, **Fig. 3B** shows a semi-log plot of $D_{CC}$ and $D_{SF}$ values versus the scan lengths from 1 nm up to 150 nm recorded for the quartz substrate. Both plots behave in a similar manner, that is they follow non-monotonic trend on scan size with clearly visible cross-over. It might be due to possible transition between different roughness components (roughness, waviness, form) with different scaling exponents, which is also seen in **Fig. 1C–F**. Unlike large number of grown surfaces, these images cannot be transformed into each other by similarity, that is, by changing the zoom. As a result, changing the size of an object (crystal grains) with constant scale of observation, should influence the fractal dimension. Apart from that, however, the crystal structure may be also important – flat single-crystal facets in diamond grains exhibit significantly lower fractal dimension than rough polycrystalline material. Another explanation involves the fingerprint of multifractal behavior specific of surfaces formed by different formation mechanisms. It is also possible that the methods cannot distinguish between local regions with different fractal dimensions and non-fractal ones, because each of them averages the topography to a certain degree. Unfortunately, the above explanations remain speculative, and the issue needs to be addressed in further studies (by comparison with artificially generated surfaces, for example).

For better clarity, results from **Fig. 3A** are re-plotted in **Fig. 4** using the $D_{RMS}$ values as the reference (x-axis). Dotted straight line with the slope equal to 1 helps to compare the results, and to verify, how each numerical procedure copes with the same data. As mentioned previously, each method generally underestimates the fractal dimension with respect to $D_{RMS}$. However, the roughness method requires at least several AFM scans with various scan

**Fig. 1.** AFM images (10 $\mu m \times 10 \mu m$) of a diamond film on quartz substrate at various stages of the deposition process: (A) quartz substrate after mechanical seeding and (B)–(F) steadily developing diamond film.

**Fig. 2.** Temporal changes in the film thickness (closed circles), and the surface roughness (open circles) of the diamond film as a function of the deposition time.
lengths to calculate the fractal dimension, which may be difficult to implement, especially on rough samples with extreme changes in vertical topography. Another issue is that the roughness is insensitive to directional properties of the surface, and hence it assumes the perfect isotropy of the surface.

The SF method also exhibits reasonable numerical accuracy in deriving fractal parameters from AFM measurements. The main advantage of the structure function follows from its numerical stability, and independence of the mean plane, which implies that the singularities are not created at the origin. What is also important, this method estimates the fractal dimension as an average over the polar angle, giving rise to its low sensitivity to surface anisotropy. Apart from that, however, the structure function suffers from serious drawbacks. This function is known to be reliable up to only one tenth of the image size, and thereafter it starts to fluctuate due to digital limitations. In addition, algorithm of the SF can be hardly optimized with respect to that of the ACF. Also, the structure function is produced by subtraction of a given image and its lagged copy, but it is commonly known that the more nearly equal two numbers are, the more precision is lost in the subtraction. What is worse, obtained results are then raised to the power of 2, and the fractal parameters are estimated using the SF values around the origin (i.e. those with the largest error), which may even decrease the accuracy of the method.

On the contrary, computation of the autocorrelation function is very fast and numerically efficient since it can be implemented using the Fast Fourier Transform algorithms. Therefore, in its simplest form that involves a single AFM measurement, this method may serve either as a rapid surface characterization tool or as an on-site process monitoring tool even if its numerical accuracy is lower than that of the SF. However, some difficulties might arise due to ACF dependence on the mean value (and hence on the procedures of the flattening of AFM images), and possible presence of singularities at the origin. Another problem arises that simplistic assumption about the Gaussian distribution of the surface heights used to re-compute the ACF into the SF and further into fractal parameters might not hold in case of anisotropic surfaces. The ACF is highly sensitive to anisotropy, and the same concerns to the SF derived indirectly from it, which means that obtained fractal dimensions can be substantially different in directions parallel and perpendicular to the surface lay. This effect might not be necessarily compensated by the averaging procedure leaving the final $D_{ACF}$ value largely underestimated.

The cube count method is found to be the poorest among those studied. It is hardly sensitive to fractal dimension, and significantly deviates from the reference. Hence, this method is not suggested for an accurate fractal analysis.

Fig. 3. (A) Changes in the fractal dimension of the diamond film during the deposition process calculated with the methods discussed in the paper: the cube count method ($D_{CC}$), the autocorrelation function method ($D_{ACF}$), the roughness method ($D_{RMS}$), and the structure function method ($D_{SF}$). (B) Fractal dimension determined using the cube count method and the structure function method for the quartz substrate with increasing scan length.

Fig. 4. Comparison of the fractal dimensions $D_{ACF}$, $D_{RMS}$, and $D_{SF}$ to the $D_{CC}$ value taken as a reference. Straight line with the slope equal to 1 is shown for convenience.

Fig. 5. Time-dependent plot of the anisotropy ratio of the growing diamond film measured from the decay lengths of the autocorrelation function.
decay of the autocorrelation function according to Eq. (10) versus deposition time. The surface appears to be highly anisotropic at early growth stage ($S_{\tau} \leq 0.2$), but then it turns into isotropic one ($S_{\tau} \geq 0.6$). The ratio achieves the maximum value equal to 0.89 after 2 h of the deposition process. Afterwards, it gradually falls down to 0.63.

5. Conclusions

Obtained results clearly show the supremacy of the roughness method over the others studied in the paper. The autocorrelation method turned out to be slightly less accurate, but its low computational costs, numerical discrepancy below 9 per cent, and sensitivity to anisotropic properties of the surface make this method preferable in rapid characterization of the materials with the use of the AFM measurements. Unfortunately, they all use certain averaging procedures to compute the fractal parameters, and hence, they cannot distinguish local regions with different fractal dimensions as well as non-fractal regimes. As a result, obtained fractal parameters are found to be dependent on the scan size, and critical grain size.

Apart from that, the fractal analysis together with the anisotropy analysis reveals some information about the kinetics of the deposition process. In the beginning, the fractal dimension raises until the crystal grains remain separated by empty spaces from each other, but once they meet to form the closed layer, the fractal dimension gradually falls down. The anisotropy ratio exhibits similar behavior concerning the morphological change from anisotropic surface into isotropic one, although both quantities are rather poorly correlated.

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