Computational nanometrology of nanostructures: the challenge of spatial complexity

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Abstract

Several applications of nanotechnology are based on the surface nanopatterning of materials and the new functionalities it brings about. Not surprisingly, the novel material properties are tightly linked to nanostructure morphology and very sensitive to its geometrical characteristics. Therefore, the measurement and characterization of nanostructure morphology is very critical in order to get control of the added value of nanopatterning on material properties and functionalities. In other words, there is an emergent need for accurate and concise metrology of all kinds of nanostructures.

Up to now, the main tools for imaging and measuring the nanostructures are the well-known and widely used probe microscopes (AFM, SEM, TEM). However, due to the minute size of the measured structures, the measurement results are strongly dependent on the effects of measuring device and process. In order to get as more accurate measurement as possible, it is necessary to deconvolute the true structure from the effects of measurement. This can be made through the development and implementation of mathematical modeling methods able to get control of the measurement effects on the result and aid the acquisition of the true morphology. Furthermore, novel mathematical and computational methods are required in the characterization of complex surface nanostructures created by deposition, etching, ion bombardment or laser treatment of surfaces. The mathematical and computational methods needed to aid the accurate and complete metrology of surface nanostructures are collectively defined by the term computational nanometrology.

In this paper, first we shortly introduce the field of computational nanometrology and define its content. Then we focus on two specific applications to demonstrate the benefits of computational nanometrology. In the first, a new mathematical transform is proposed to enable the simultaneous characterization of both periodicity and feature width in almost periodic arrangements of nanodots on a surface. In the second, the multifractal spectrum of complex nanomorphologies is calculated to quantify their multiscale hierarchical structuring. Both methodologies are motivated and applied to the characterization of polymer surfaces after their treatment in plasma reactors.

1. Introduction

Nanometrology is the branch of metrology science which aims at providing methods and techniques for accurate and complete measurements of nanostructure properties including dimensions, morphology (shape), electric/magnetic fields, optical spectra, chemical composition, material hardness, etc. Since the recent advances in nanotechnology are mainly related to our ability to nanopattern material surfaces and bulk, the metrology of dimensions and surface morphology is of chief importance to get controlled and reproduced nanopatterning and therefore device performance. However, along with the increasing importance of dimensional and morphological metrology at nanoscale, we have the emergence of some specific issues which challenge both the accuracy and completeness of nanomeasurements.

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The first challenge has to do with the enhanced impact of measurement probe to the measurement results given the tiny size of measured object which is almost comparable to the size of the probe. This effect is very critical in scanning probe microscopy techniques (AFM, STM, etc.) but also affects the image-based metrologies such as SEM, STEM and TEM [1]. Another aspect of this challenge is the degradation that the energy of the used probe can cause on nanosized specimen. This is one of the greatest challenges in the use of SEM metrology in resist (polymer) nanogratings which are used in nanolithography for patterning transistor structures [2]. In this challenge we can also encompass the enhanced effects of the noise of measurement (instrument and process) on the measurement result emanated from the weakening of true measurement signal due to nanoscale sizes [3].

The second challenge of nanometrology is to achieve measurements of nanostructured surfaces and materials which at the same time have high resolution and cover large ranges of the measured surfaces. This is needed in many applications where nanostructuring is applied at large scales (for example nanoimprint lithography patterning of large polymer sheets) where we should have accurate measurements of the dimensions and shapes of billions of nanostructures (dots, wires,…) covering areas of millimeter scales. Recently, this challenge has been faced up by the combination of different techniques either at instrumental or data level. In the latter case, data fusion methods and algorithms have been applied properly adapted to the needs of nanometrology [4].

The third challenge comes from the complexity of nanostructure shape and morphology. Although this is more evident in self-assembled nanostructures delivered by bottom-up techniques, one can find aspects of this complexity (for example sidewall roughness) in well-organized lithographic nanostructures. The stochastic characteristics of nanomorphologies may require the proposal of new metrics or the adaptation of older and more well-established methodologies. Fig. 1 shows a collection of AFM and SEM images of polymer surfaces after their etching in Oxygen plasmas. One can notice the rich complexity of the imaged morphologies and deduce the need for new mathematical and computational tools dedicated to their characterization [5].

The consideration of above challenges in nanoscale metrology demands the invention and development of new measurement instruments and techniques but also requires better understanding and more accurate and complete characterization methods and tools. For example, in the first challenge the understanding of probe-sample interactions can be advanced by the modelling and simulation of the measurement process while in the second and third challenge developing new data analysis methods can have catalysing impact on compromising resolution and range and on providing more complete characterization of nanostructure complexity. The collection of all mathematical and computational methods devised for the modelling and characterization of measured nanostructures is usually called Computational Nanometrology (CNM). Fig. 2 displays a schematic of the measurement process indicating the two main areas of CNM dedicated to the modelling/simulation of measurement process (CNM1) and measurement data analysis (CNM2) respectively.

This paper focuses on CNM2 and more specifically shows two examples of CNM methods targeting to provide more complete nanostructure characterization aspects. The first regards the metrology of almost periodic arrangements of nanodots formatting during the first minutes of plasma etching of plasma films (Section 2) while the second considers the multiscale features of the same polymer morphologies when longer plasma treatment times

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are applied and multifractal analysis of SEM images is performed and discussed (Section 3). The paper closes with a summary of our results in the Section 4.

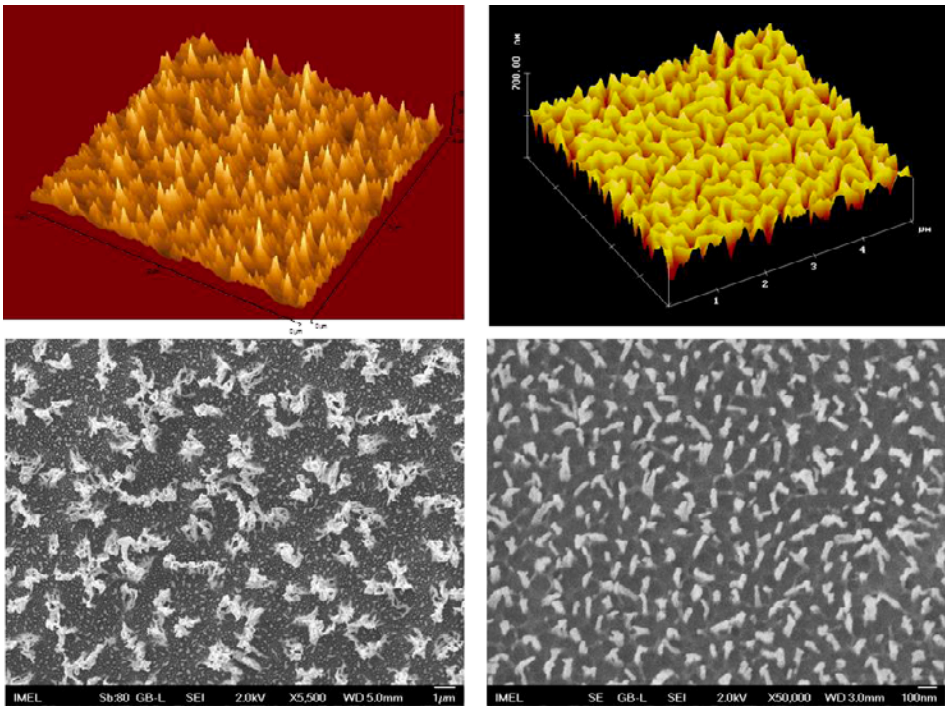

Figure 1. AFM (top row) and top-down SEM (bottom row) images of complex surface morphologies of polymers after their treatment (etching) in oxygen industrial plasma for different treatment times. Notice the multiple aspects of spatial complexity of the images nanostructures.

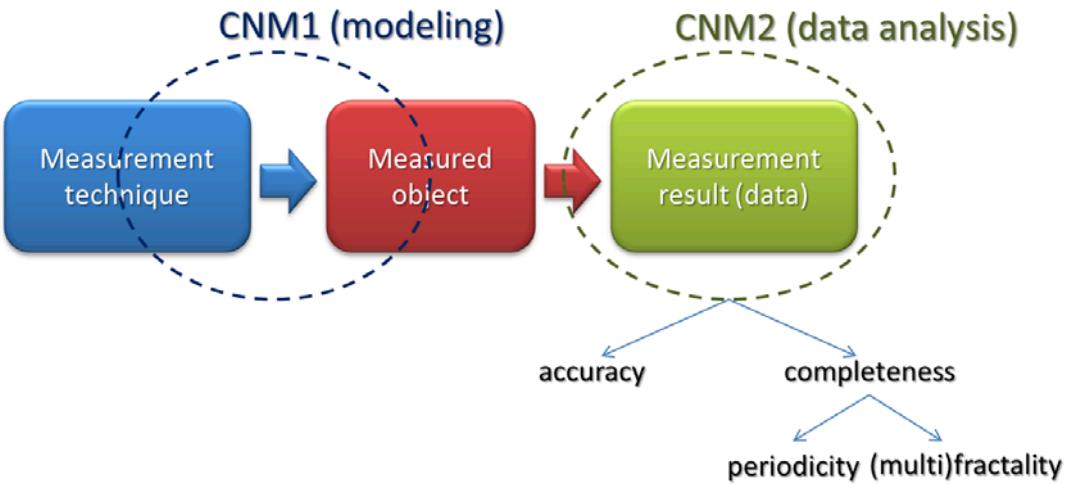

Figure 2. A schematic of the measurement process and the areas of application of Computational Nanometrology (CNM) methods: modeling of measurement process (CNM1) and measurement data analysis (CNM2). Also, the two branches of CNM2 dedicated to accuracy and completeness issues of nanometrology are shown along with the focus of our paper on periodicity and multifractality.

2. Characterization of almost periodic nanostructures: Period-Scale Transform

One of the key concepts in nanotechnology is the self-assembly of nanomaterials used in fabrication processes. In surface nanopatterning applications, self-assembly may result in the formation of almost periodic nanodots on surfaces with a characteristic size (see Fig. 3). The characterization of such surface morphologies can be done conventionally with two ways. The first is to consider the surface topography as roughness and calculate the standard surface roughness parameters found in relevant parameter and/or proposed by ISO [6,7]. The second approach is to make Fourier analysis transforming the surface to spatial frequency domain. The benefits of this transformation are to reveal and quantify the existed periodicities while by the inverse of the width of the peak in Fourier transform one can characterize the degree of periodicity. However, both approaches suffer from drawbacks. The first ignores the dominant periodicity (translational symmetry) and handles the surface morphology as random fluctuations without any kind of symmetry. The second approach, though closer to the periodic organization of surface, fails to distinguish the two different types of deviations from periodicity coming from variations in nanodot positions and changes in their size scales (widths). In nanotechnology to have an assessment of the different kinds of deviations from periodicity can be critical since they may be related to different fabrication conditions and/or surface functionalities [8-9].

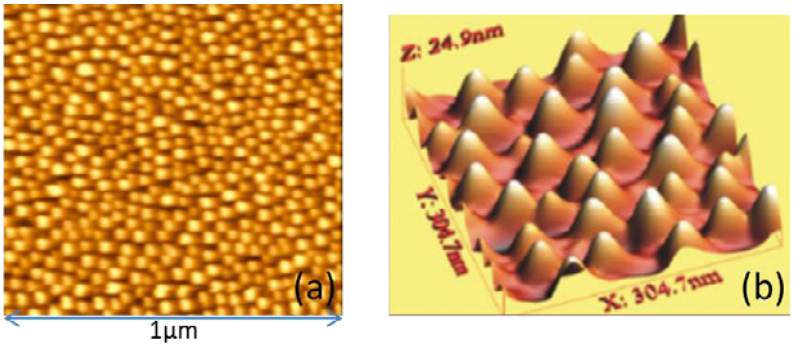

Figure 3. AFM measurements of the surface topographies of polymer (PMMA) surfaces after 1min Oxygen plasma etching in a plasma reactor [10,11].(a) Top-down image and b) magnification 3D topography

In order to face up this challenge and provide a method for the characterization of the different aspects of periodicity and of the deviations from it, we propose an alternative transform called Period-Scale Transform (PST) which is able to output info for both types of periodicity and deviation from it. Actually, PST is a combination of the Fourier transform with the localized concept of wavelet transform. The mathematical definition of the PST of a 1D function *f(x)* is given below:

$$PS_g f(s,\lambda) = \int_{-\infty}^{\infty} f(x)\,(G(x;s,\lambda) - \langle G\rangle)\,dx \qquad (1)$$

Where the kernel $G(x;s,\lambda)$ is the sum of localized functions $g(x;s,\lambda)$

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$$G(x;s,\lambda)=\sum_{n=-\infty}^{\infty}\tau_{n\lambda}g_s(x)=\sum_{n=-\infty}^{\infty}g\left(\frac{x-n\lambda}{s}\right). \quad (2)$$

One common example of *G(x;s,λ)* is shown in Fig. 4 where *g(x;s,λ)* are Gaussian peaks with width s and centered around integer multiples of λ.

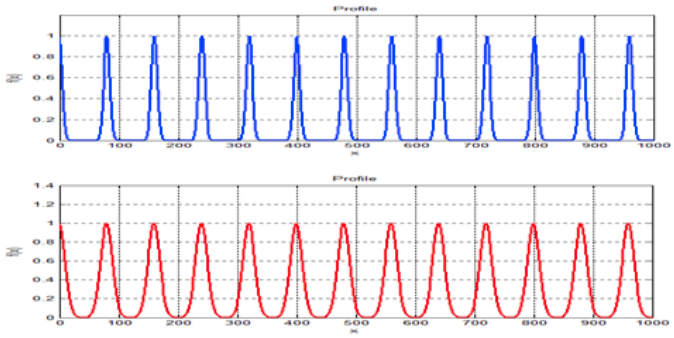

Figure 4. An example of PST kernel *G(x;s,λ)* consisting of a sum of Gaussian peaks for two different peak scales s.

As shown from the definition in (1), PST maps a 1D function to a 2D plane with axes the wavelength λ and the peak scale s. The first parameter discloses the periodicities of the analysed function while the second is used to identify the spatial scales of the peaks/protrusions comprising the surface. For a 2D surface, the PST is a 4D function and its representation should be carefully displayed.

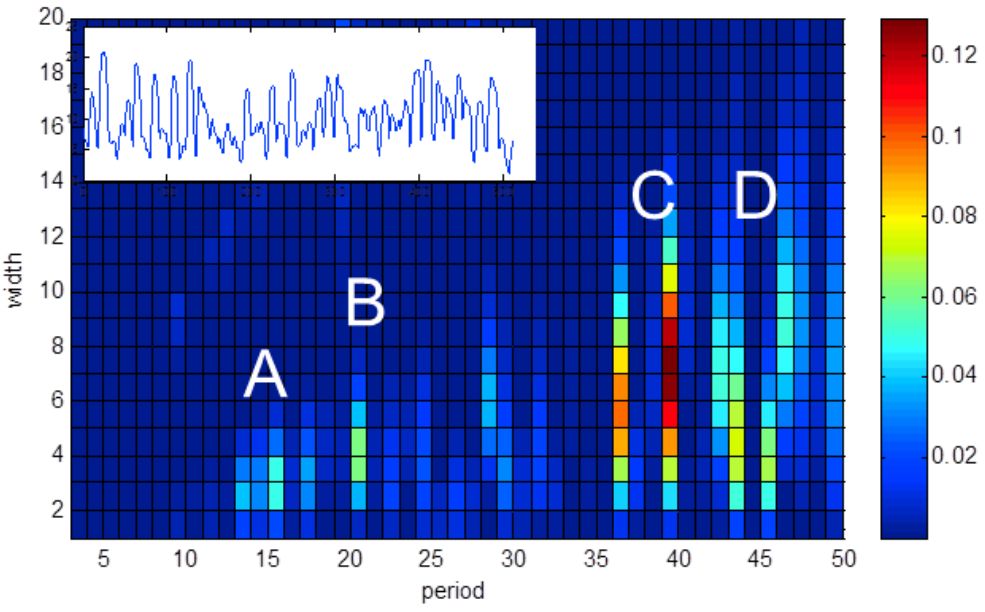

Figure 5. PST of the profile shown in the inset which has been taken from the AFM surface of Fig. 3.

Fig. 5 shows the PST of a profile (see inset) extracted from the surface morphology shown in the AFM image of Fig. 3. The transformation captures the existed periodicities in the profile morphology which are also found in the Fourier analysis of the profile (not shown here). In addition, PST provides an evaluation of the involved widths of peaks at each selected

wavelength. Not surprisingly, the scales of the peaks enhance as we are moving to longer periods although interesting deviations are observed in D peaks with respect to C ones.

3. Multifractal analysis of complex surface nanostructures

The second challenge of CNM2-Data analysis regards surface structures with complex appearances and the involvement of features with different size scales and at different position scales. Since the size and positional scales of a surface morphology is a critical feature affecting its functionalities, the fractal geometry seems to be a useful method when some scaling symmetries are present. However, frequently in the formation of a nanostructured surface, a wide spectrum of mechanisms is involved with their own spatial and time scales. As a result, the fabricated surfaces are characterized by the co-existence of multiple scales and fractal symmetries interweaved in complicated manner. A method to dissolve this complexity and quantify the co-existence of multiple scaling symmetries in a surface is the multifractal analysis (MFA) which is actually an extension of the fractal geometry when more scaling symmetries are involved. The output of the multifractal analysis is not a single fractal dimension but a spectrum of fractal dimensions (multifractal spectrum) whose width indicates the extent of scales and fractal structures involved in surface morphology.

In literature, one can find several methods for the implementation of multifractal analysis. For nanostructured surfaces, the most commonly used method is a properly modified version of the Box-Counting method (BCM) which measures the scaling behavior of the surface mass to give the gradient of fractal behavior versus the amount of surface mass [12].

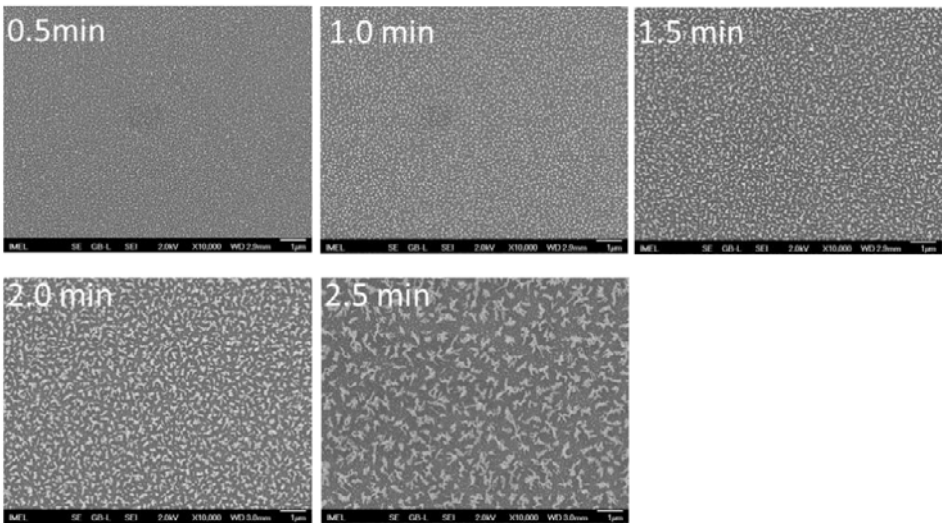

Figure 5. Top-down SEM images of PMMA polymer surfaces after oxygen plasma etching for 0.5, 1, 1.5, 2 and 2.5 min. One can notice the increased complexity of surface morphology versus etching time characterized by the emergence of multiple scales organized in a hierarchical manner.

We have applied the MFA in a series of polymer (PMMA) surfaces for increasing times of plasma etching (see Fig. 5). The first surface of this series has been used for the application of PST in Section 2. Here, instead of using AFM measurements, we have acquired SEM images in order to get larger scannig regions and have better statistics at long etch times. At large

etch times, the simple almost periodic arrangements of nanodots are replaced by more complex structures where multiple scales are involved and characterize the final nanostructured surface.

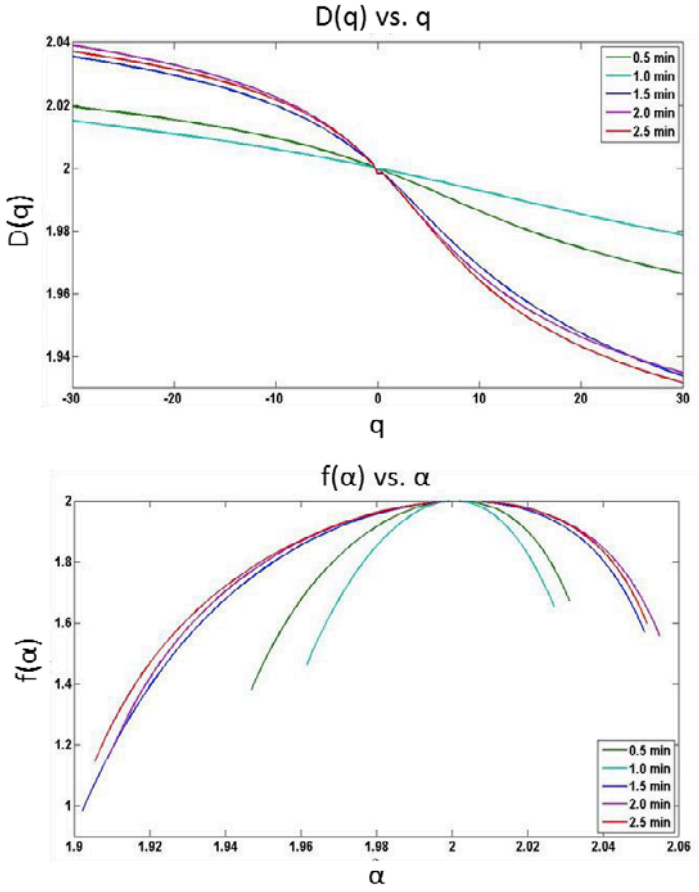

Figure 7. Multifractal spectra of generalized fractal dimensions D(q) vs. q (top) and f(α) vs. α (bottom) for the SEM images of PMMA surfaces displayed in Fig. 6. The apparent increase of spatial complexity vs. etching time is captured in multifractal analysis by the widening of multifractal spectrum at long etch times.

The results of the application of the BCM for the MFA of the SEM images of Fig. 6 are shown in Fig. 7. The top diagram displays the generalized fractal dimensions D(q) vs. q where the exponent q spans the whole range image intensities and the corresponding D shows the fractal behavior for the specific range. For example, the D(q) at the smallest negative q (=-30) characterizes the scaling behavior of the dark image regions while at positive q the spectrum is dominated by the scaling analysis of bright image regions. Therefore, the difference $\delta D=D(q=-30)-D(q=30)$ quantifies the degree of multifractality in a surface. Large δD is a hallmark of the co-existence of multiple scales in surface structuring while small δD indicates limited multifractality with the limit of monofractality at $\delta D=0$. The diagram with the plots of D(q) vs.q for all surfaces reveal clearly the wider range of scales emerged in polymer surfaces with etching time. More interestingly, it seems to exist a grouping of D(q) curves at etching times>1min which means that critical involvement of other scales in surface morphology takes place at etch times between 1 and 1.5 min.

Similar conclusions are reached when we inspect f(α) curves for different etch times. These spectra are calculated by te Legendre transforms of D(q) and they are considered more generic than D(q). Again, wider spectra indicate more multifractlity i.e. more feature scales contribute to build the final surface morphology.

5. Summary

The main goals of this work have been:

a) To underline and illustrate the importance of Computational Nanometrology and propose a classification of its different methods and implementations such as in the modeling of measurement process or in the analysis of measurement data.
b) To focus on specific open issues and demonstrate the capabilities of CNM methods and applications. The first example has been the development of an alternative surface transformation called Period-Scale Transform which can be applied to almost periodic surfaces and characterizes the different aspects of periodicity (position and scale of repeated features) as well as the different types of deviation from periodicity corresponding to these aspects.
c) To analyze the scaling symmetries of apparently complex polymer surfaces by applying the multifractal theory properly adapted for the analysis of SEM images of surfaces. We found that MFA analysis captures the changes in surface morphologies vs. etch times of polymer films and indicates a critical threshold in etching time for the insertion of other scales in surface morphology.

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