Probing rough surfaces: Markovian versus non-Markovian processes

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Abstract. We demonstrate that Markov processes play a fundamental role in probing rough surfaces and characterizing their topography. The surface topography obtained by a probe, as in the atomic force microscopy (AFM) technique, is based on the probe–surface interactions. When the size of the probe tip is comparable with the height of the surface fluctuation, the surface image can be aberrated from its origin. Due to the tip effect, there is a crossover in the structure function of the surface. For scales smaller than the Markov length—the minimum length scale over which a process is Markovian—the stochastic function that describes a rough surface is non-Markovian, whereas for length scales larger than the Markov length, the surface may be described by a Markov process. Synthetic rough surfaces generated by fractional Guassian noise (FGN), as well as the rough surface of $V_2O_5$, obtained by AFM, confirm this.

Characterization of the topography of material rough surfaces, and the effect of the topography on the material properties, ranging from adhesion to fracture [1]–[6], have been the subjects of much interest over the past two decades. Profilometry techniques, such as atomic force microscopy (AFM), are powerful probing methods for high-resolution imaging and characterization of nano- to macroscale fluctuations in the height of surfaces of various materials [7]–[17]. Most imaging processes by profilometry techniques are affected by the interaction between the tip of the probe and the sample. When the resolution of the observation is comparable with the tip size, the scanned image is a ‘convolution’ of the tip and the sample.

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geometries. Convolution has been used by authors to describe the distortions caused by the tip shape [18, 19], although others [20] have suggested describing the distortions by the concept of dilation. In the literature, this phenomenon has been reported to be present in the images of a variety of materials, which are distorted by the tip-convolution effect [18], [20]–[24]. Aue and De Hasson [20] and Klapetek et al [25] reported on the appearance of multifractality (i.e. a crossover) in the structure function and concluded, based on wavelet analysis, that this effect cannot be efficiently removed by a theoretical analysis. Buzio and Valbusa [26] considered the effect of increasing tip curvature radius on the structure function and observed variation of structure function slope (that is a crossover) on length scales comparable with the tip size.

While the question of the reliability of the images of material surfaces obtained by any profilometry technique is important and must be addressed, as such images are a prime tool for obtaining much information about the surface structure of materials (particularly biological materials), there has not been much effort devoted to the issue. Addressing this issue is one of the main goals of this study. We introduce novel concepts, heretofore not used, in order to study the problem.

To reduce the influence of the probe geometry, efforts have been focused on producing smaller probe tips. Novel manufacturing techniques, such as electron beam deposition, crystal growth and ion beam etching, have produced tips with higher aspect ratios [22]. However, the smallest tip sizes that have been obtained are a few nanometres [22, 27]. Consideration of the tip–sample adhesive interactions suggests that there is a lower limit in the tip sharpness which may cause inelastic deformation of the sample. In addition, tips with higher aspect ratios can easily break when scanning hard and rough samples, or damage soft surfaces. In general, the probe’s aspect ratio is a prominent limiting factor in the accuracy of the image of a surface. A small probe angle makes it very fragile and, therefore, unusable on rough surfaces. This makes it necessary, for some samples, to use tips with lower aspect ratios. In such cases, the influence of the tips cannot be ignored, since the smaller the tip aspect ratio, the larger is its effect. Thus, some authors tried to develop techniques for image refinement in order to partially reduce the topographic distortions induced by the geometry of the tip [21, 22, 24]. Many phenomena were observed using various tips and sample geometries, such as broadening of the apparent width and rounding of sharp edges. Several simulation studies of the deconvolution of the tip and the surface were also carried out, in order to reduce the convolution phenomenon, although to a large extent the problem was not solved [22]. For example, when one uses AFM to study the scaling behavior of fractal surfaces, the issue is the extent to which the observed scaling exponent of the fractal surface is influenced by its distorted image [20].

The distortion in observed data arises from the fact that interactions which are assumed to take place at the tip apex, actually occur on the side of the tip. We can, therefore, conclude that if the probe is sharp enough, it will contact the surface properly and the image will be correct. But if it is not, then the image of some parts of the surface is distorted. The height fluctuations of the one-dimensional (1D) surfaces are shown in figure 1, before and after probing with a pyramidal probe (the angle of the probe was $\theta = 45^\circ$).

Scanned data are affected more by the peaks than by the valleys of the surface. Also, a surface image is smoother than the real surface ($\sigma_{\text{imaged}} < \sigma_{\text{real}}$ as figure 1 top-right). In other words, most of the information that may be lost due to the probe effect is in the valleys. This implies that the probe effect represents a nonlinear map from the actual surface to its imaged topography, and is related to the scale of the scan.
Figure 1. The height $h(x)$ of a surface generated by FGN before (thinner) and after (thicker) being scanned by a tip with $\theta = 45^\circ$. The inset shows how the PDF of the height fluctuations is changed as a result of probing.

Some parts of the probe with an arbitrary shape may interact with the surface before the apex actually touches it. Therefore, the height of the point which is below the apex depends on the height of the point which is actually in contact with the tip. Then, this effect could change the joint probability as a function of the heights of two points and their distance. In this paper, we show that there is a new characteristic scale—the maximum length scale on which the joint probability density function (PDF) exists. Physically, this length scale estimates the minimum scale which due to the probe’s influence vanishes and, thus, the topography of the sample obtained by the profilometer is reliable.

To identify this length scale, one can investigate the missing information by evaluating important statistical parameters that characterize the surface. For simplicity, we consider a pyramid-shaped probe, which is characterized by, $y = 2 \tan(\theta) |x|$, exploring a rough surface, where $\theta$ is the probe’s half angle, and $x$ and $y$ are the probe’s height and base length, respectively. To derive quantitative information about the surface morphology and probe effects, we use the surface structure function (which depends on the length scale $\Delta x = l$) as follows:

$$S(l, t) = \langle (h(x + l, t) - h(x, t))^2 \rangle. \quad (1)$$

First of all, surfaces with various Hurst exponents $0.1 < H < 0.9$ were generated using fractional Gaussian noise (FGN) [28]. Figure 2 compares the structure function for the typical FGN surfaces after being probed with tip angles from 1.1 to 1.6 radians. With increasing probe angle, the scale of its convolution increases. The structure functions for various aspect ratios indicate a crossover from a higher slope to a smaller one. The crossover point moves to larger scales with increasing aspect ratio. The results are in accordance with the studies of Aue and De Hosson [20] and Klapetek et al [25] on multifractality (i.e. a crossover) in the structure function.

To identify the length scale at which the probe effect diminishes, we utilize Markov processes. We determine the Markov length $l_M$ [29]–[32], [34] that is a length between points at which the heights of those points follow a Markov process. In other words, if the scale of observation is smaller than the Markov length, the stochastic process that describes the height fluctuations will be non-Markovian. Indeed, the probe interaction with the height fluctuation
Figure 2. The structure function deviation of a typical probed surface, before and after probing by the tip with various angles $\theta$ (in radian). It shows a crossover in structure function of observed data with respect to the original.

Figure 3. 1D simulation scheme for obtaining an AFM image (the tip interaction between a triangular tip and a hole).

of the surface is the main reason that the Markov length scale (non-Markovian scale) exists in the sample. Figure 3 shows how the probe reports the topographies from the original one. Because of the contact of each point from the probe with a point in the surface, the resulting topography differs from the original one. This means that the heights of the contact points were jointed with the height reported by the top of the probe. In fact, in this jointed length scale, the process is non-Markovian and the largest joint length scale in the surface is the Markov length scale. These points can be summarized in a sentence which is ‘the minimum length scale or the highest resolution scale in the scan image, which is certainly reliable, is the Markov length scale’. To estimate $l_M$, we note that a complete characterization of the statistical properties of height fluctuations of a rough surface $h$ in terms of a parameter $x$ requires evaluation of the joint PDF, i.e. $p(h_1, x_1; \ldots; h_n, x_n)$, for any arbitrary $n$. For a Markov process, $p$ is generated by a
Figure 4. Dependence of the Markov length on the Hurst exponent and the probe angle.

The product of the conditional probabilities \( p(h_{i+1}, x_{i+1}|h_i, x_i) \), for \( i = 1, \ldots, n - 1 \). As a necessary condition, for being a Markov process the Chapman–Kolmogorov (CK) equation

\[
p(h_2, x_2|h_1, x_1) = \sum_{h'} p(h_2, x_2|h', x') p(h', x'|h_1, x_1)
\]

must be satisfied for any value of \( x_i \), in the interval \( x_1 < x_i < x_2 \) [29]. The simplest way to determine \( l_M \) for the data is the numerical calculation of the quantity,

\[
D = |p(h_2, x_2|h_1, x_1) - \sum_{h'} p(h_2, x_2|h', x') p(h', x'|h_1, x_1)|,
\]

for given \( h_1 \) and \( h_2 \), in terms of, e.g. \( x_2 - x_1 \) (taking into account the possible errors in estimating \( D \)). Then, \( l_M \) is that value of \( x_2 - x_1 \) for which \( D = 0 \).

These analyses were performed for different surfaces with Hurst exponent \( 0.1 < H < 0.9 \) and different probes with semiangle \( 10^\circ < \theta < 80^\circ \) (figure 4). We also find, empirically, the relation between the Markov length \( (l_M) \), Hurst exponent \( (H) \) and the probe angle \( (\theta) \) as

\[
\ln(l_M) = a + bH^3 + c\theta^2,
\]

where \( a = -0.1698, b = 0.6398 \) and \( c = 0.0004 \).

Indeed, in our analysis, we have considered the unit roughness \( (\sigma = 1) \). The roughness effect appears in the angle of the probe. Higher roughness is equivalent to scanning with thinner probes. The relation between the angles of the probe with unit roughness \( (\theta_0) \) and original roughness of the surface \( (\sigma) \) is \( \tan(\theta_0) = \frac{1}{\sigma} \tan(\theta) \) (see figure 5), where \( \theta \) is the angle of the probe for the surface with \( \sigma \) roughness. Another important point that we must consider in probe convolution is the maximum resolution in the scanned image. Similar to the roughness problem, higher resolution is equivalent to a wider probe. In other words, if the resolution increases two times, this has the same tip effect as when the roughness increases two times. It means that we must consider not only the roughness, but also the resolution of the samples.

For considering the correlation length effect in our analysis, there are two cases with respect to the Markov length. The first is when the correlation length is larger than the Markov length. In this case, we find a crossover in the structure function because the tip-size effect leads to a smoother surface in the Markov length scale. Therefore, we find two roughness exponents (see figure 6). We can find the original exponent by knowing the probe angle, surface roughness and the Markov length of the reported data. The second one occurs when the correlation length

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Figure 5. 2D scheme of obtaining the relation between the probe angle and roughness. It means that the probe angle has roughness information. A thinner probe with a rougher surface has the same effect as a wider probe with a smoother surface. In other words, increasing the roughness results in the same behavior as increasing the probe angle.

Figure 6. V$_2$O$_5$ surface image, obtained by AFM, and its structure function. The crossover is obvious.

is smaller than the mock Markov length. In this case, all statistical information is lost in the scale smaller than the Markov length. In other words, only scales larger than the Markov length are reliable. Knowledge about the Markov length scale helps us to find the minimum scale or the maximum resolution scale at which the reported topography of the surface is reliable. Empirically, we can apply the moving average over all data with a Markov length window size. There may exist a Markov length scale in a real surface. In this case, we should explain the source of this length scale fundamentally. For example, for a growing surface it may come from the size or shape of particles which have been used for its growth, and our observation is of less than this size. In addition, if there exists a Markov length in the original case, after scanning...
with a probe, a Markov length may be produced from the probe convolution too which could be equal to the original Markov length or not. But the important point is that we should be careful in the scales smaller than the Markov length. Even if we know that this is an original length scale in the surface. We emphasize that ‘the minimum length scale or the highest resolution scale in the scan image, which is certainly reliable, is the Markov length scale’. This means that we should be careful in the scale smaller than the Markov length.

Now we test these concepts by considering a $\mathrm{V}_2\mathrm{O}_5$ layer, grown on a polished Si(100) substrate by the resistive evaporation method in a high-vacuum chamber [33]. This material has a highly porous surface, and has been the subject of intense study [34]. The pressure during evaporation was 5–10 Torr. The surface topography of the film was investigated using an AFM (Park Scientific Instruments model Autoprobe CP). The image was collected in constant-force mode and digitized into $256 \times 256$ pixels with a scanning frequency of 0.6 Hz. A cantilever of $0.05 \, \text{N m}^{-1}$ spring constant with a commercial standard pyramidal $\text{Si}_3\text{N}_4$ tip with $3 \, \mu\text{m}$ size was used. We consider a pyramid-shaped probe with $\theta \simeq 35^\circ$, where $\theta$ is the probe’s half angle and the aspect ratio $(2 \tan \theta)$ is about 1.4. Figure 6 presents the structure function of the $\mathrm{V}_2\mathrm{O}_5$ surface, together with the surface AFM image. As figure 6 indicates, there is a crossover in the structure function from a slope of $0.83 \pm 0.02$ over the scale smaller than about 16 nm, to $0.29 \pm 0.04$ in the range 20–40 nm. In addition, due to the probe effect, the measured Hurst exponents can be higher than the true values. As discussed above, this raises the question, to what extent are the observed scaling exponents influenced by probe shape? In other words, what is the scale over which the estimates are reliable? Analyzing the $\mathrm{V}_2\mathrm{O}_5$ surface, we estimate that the Markov length is $l_M \simeq 14 \, \text{nm}$, which is also in reasonable agreement with what figure 6 indicates.

In summary, the main reason that Markov length scale (non-Markovian) exists in profilometers is the probe interaction with the height fluctuation of the surface. The minimum trustable length scale (the highest resolution scale) in the scan image is the Markov length scale (see figure 3). If $\delta$ is a measure of the resolution of length scales (a minimum scale which is equal to a pixel size) in the scanning process, the probe effect can be expressed as follows,

\[
\delta < l_M \quad \Rightarrow \quad H_{\text{image}} > H_{\text{real}},
\]
\[
\delta \geq l_M \quad \Rightarrow \quad H_{\text{image}} = H_{\text{real}}.
\]

These relations mean that at scales lower than the Markov length, the evaluated Hurst exponents of AFM images are larger than the original ones. This means that the size effect leads to reporting smoother surfaces.

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Appendix. Estimation of the Markov length

Complete statistical information about the morphology of a rough surface would be available from the height knowledge of all possible $n$-points, or more precisely $n$-scales, joint PDF $p(h_n, x_n; h_{n-1}, x_{n-1}; \ldots; h_1, x_1)$ describing the probability of finding simultaneously, the height
$h_1$ on the scale $x_1$, $h_2$ on the scale $x_2$, and so forth up to $h_n$ on the scale $x_n$. Without loss of generality, we consider $x_1 < x_2 < \cdots < x_n$. In any case, the $n$-scale joint PDF can be expressed by the multi-conditional PDF \cite{29}

\begin{equation}
 p(h_n, x_n; h_{n-1}, x_{n-1}; \ldots; h_1, x_1) = p(h_n, x_n|h_{n-1}, x_{n-1}; \ldots; h_1, x_1) \\
 \times p(h_{n-1}, x_{n-1}|h_{n-2}, x_{n-2}; \ldots; h_1, x_1) \ldots p(h_2, x_2|h_2, x_2)p(h_1, x_1). \tag{A.1}
\end{equation}

Here, $p(h_{i+1}, x_{i+1}|h_i, x_i)$ denotes a conditional probability of finding the height $h_{i+1}$ on the scale $x_{i+1}$ under the condition that simultaneously, on the scale $x_i$, we find $h_i$ as the height. It can be defined with the joint probability $p(h_{i+1}, x_{i+1}; h_i, x_i)$ by

\begin{equation}
 p(h_{i+1}, x_{i+1}|h_i, x_i) = \frac{p(h_{i+1}, x_{i+1}; h_i, x_i)}{p(h_i, x_i)}. \tag{A.2}
\end{equation}

An important simplification arises if

\begin{equation}
 p(h_{i+1}, x_{i+1}|h_i, x_i; \ldots; h_1, x_1) = p(h_i, x_i|h_i, x_i). \tag{A.3}
\end{equation}

This property is the defining feature of a Markov process evolving from $h_{i+1}$ to $h_i$. So, in a Markov process the $n$-scale joint PDF is reduced to the $n$-conditional PDF

\begin{equation}
 p(h_n, x_n; \ldots; h_1, x_1) = p(h_n, x_n|h_{n-1}, x_{n-1}) \ldots p(h_2, x_2|h_2, x_2)p(h_1, x_1). \tag{A.4}
\end{equation}

The Markov property implies that the $x$-dependence of $h$ can be regarded as a stochastic process evolving in $h$. Here it should be noted that condition (A.3) holds for a process that depends only on the neighboring point or the previous step. Equation (A.4) also emphasizes the fundamental meaning of conditional probabilities for the Markov processes since they determine any $n$-scale joint PDF and thus the complete statistics of the process.

In the following, we describe the evolution of height, $h(x)$ in the scale $x$ as realizations of a Markov process. For a Markov process, the defining feature is that the $n$-scale conditional probability distributions are equal to the single conditional probabilities. Three procedures were applied to find out the Markov properties of a dataset, such as height fluctuations in a rough surface. From the results of all three tests, we find a minimal length scale $l_M$ for which this is the case. As is well known, a given process with a degree of randomness or stochasticity may have a finite or an infinite Markov length scale \cite{31}. The proposed method utilizes a set of data for a phenomenon which contains a degree of stochasticity. To determine the Markov scale $l_M$ for a rough surface, we note that a complete characterization of the statistical properties of stochastic fluctuations of a quantity $h$ in terms of a parameter $x$ requires the evaluation of the joint PDF $p(h_n, x_n; \ldots; h_1, x_1)$ for an arbitrary $n$, the number of data points. If the phenomenon is a Markov process, an important simplification can be made, as the $n$-point joint PDF is generated by the product of the conditional probabilities $p(h_{i+1}, x_{i+1}|h_i, x_i)$, for $i = 1, \ldots, n - 1$.

The first approach to estimate the Markov length scale via the $\chi^2$ measure has been presented in \cite{32, 35}. The second test \cite{13, 30} is the Wilcoxon test and the last one is the CK test. Next, we give a brief introduction to this procedure, CK, which will be used here. More detailed discussions of this test can be found in \cite{13}, \cite{29}–\cite{32}, \cite{34}. A necessary condition for a stochastic phenomenon to be a Markov process is the CK equation \cite{29}

\begin{equation}
 p(h_2, x_2|h_1, x_1) = \sum_{h'} p(h_2, x_2|h', x') P(h', x'|h_1, x_1). \tag{A.5}
\end{equation}

This should hold for any value of $x'$ in the interval $x_1 < x' < x_2$. One should check the validity of the CK equation for different $h_1$ by comparing the directly evaluated conditional
probability distributions $p(h_2, x_2|h_1, x_1)$ with the ones calculated according to right side of equation (A.5). The simplest way of determining $l_M$ for a stationary and homogeneous rough surface is the numerical calculation of the quantity,

$$D = |p(h_2, x_2|h_1, x_1) - \sum_{h'} p(h_2, x_2|h', x')p(h', x'|h_1, x_1)|,$$

(A.6)

for given $h_1$ and $h_2$, in terms of, for example, $x_2 - x_1$ and considering the possible errors in estimating $D$. Then, $l_M = x_2 - x_1$ for that value of $x_2 - x_1$ for which $D = 0$. From our data, conditional probabilities have been evaluated and the validity of the CK equation was checked for the height fluctuation of simulation samples and one of the experimental samples. With simulation, we can control the interaction of the probe–surface that according to our analysis, is the main source of the non-Markovian properties in the samples. In other words, the heights of the contact points in the samples were jointed (non-Markovian properties) with the height reported by top of the probe. The largest joint length scale in the surface is the Markov length scale. These results lead us to this point, ‘the highest resolution scale in the scan image which is certainly trustable, is the Markov length scale’.

To estimate the error in $D$, we must calculate the error in the conditional PDF. Let us consider a series $h(x)$, of height fluctuations with length $N$ and the number of data points in each discrete level, $n(h)$ in the space of $(h, h + dh)$. In general, the statistical error of the PDF (or conditional PDF), $p(h)$ is

\[
\begin{align*}
\text{Prob}(h) &= \frac{n(h)}{N}, \\
\text{Prob}(h) &= p(h)dh, \\
\Delta p(h) &= \frac{\Delta \text{Prob}(h)}{dh} = \frac{\Delta n(h)}{Ndh}, \\
\Delta n^2(h) &= N\text{Prob}(h)(1 - \text{Prob}(h)), \\
\Delta p(h) &= \sqrt{N\text{Prob}(h)(1 - \text{Prob}(h))}, \\
\sigma(p(h)) &= \frac{\Delta p(h)}{\sqrt{n(h)}} = \frac{\sqrt{(1 - \text{Prob}(h))}}{Ndh},
\end{align*}
\]

(A.7)

where $\text{Prob}(h)$ is the probability that $h \leqslant h(x) < h + dh$. For small $\text{Prob}(h)$, the error is

\[
\sigma(p(h)) = \frac{1}{Ndh}. 
\]

(A.8)

It is noted that we can replace $p(h)$ with any PDF or conditional PDF.

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