Mathematical formulation of rough surfaces at the nano-scale

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ABSTRACT: The surface morphology of oriented single crystals is investigated using a theoretical approach. An adaptive method is developed based on recent theoretical developments. This new method allows to generate parameterized random surfaces including various roughness levels. After a brief comparison to AFM measurements, the method is coupled to atomistic design in order to provide more realistic and tunable surfaces as an input for molecular dynamics simulations.

INTRODUCTION

Surface roughness has a fundamental impact on the physical response of materials. Rough surfaces have been studied at the macro-scale for friction, lubrication, machining, polishing, electromagnetic wave scattering and asphalt performances in highway roads[1-5]. At the micro and nano-scales, surface roughness has been investigated to quantify the geometrical factors affecting AFM characterization[6] and adhesion phenomena in MEMS[7,8]. Surface shape and roughness possibly influence nanomechanical properties too. Generally, it is assumed that the interaction between sample and flat punch indenter is produced through planar contacts when modeling nanoparticle compression tests[11]. However, most of the images obtained with characterization methods as SEM and TEM will contrast with the perfectly flat surface assumed in atomistic models. At the atomic scale, the sample design has recently shown a gained of interest due to its impact on the modeling of mechanical properties. Prakash et all.[9] studied the influence of the local interface curvature and the local chemical composition in the dislocation process under highly idealized simulation setups with planar interfaces. Their conclusions emphasize the role of the interface curvature description on atomistic simulations results. Amodeo and Lizoul[10] recently reported the effect of blunting nanoparticles on their mechanical properties.

Mathematical formulation of random surfaces has been widely reported. To generate random surfaces, the linear transformation[12], 2-D filters[13] and the time series[14] methods are among the most frequently used. Other geometrical approximations resulting in non-real surfaces can be mentioned as well. As an example, Bhattacharjee and colleagues[15] used spheres (that account for randomness in their radii) intercepted at half of their diameter with an originally flat plane. The center of the sphere is chosen randomly as well as the part of the half sphere to be kept. Thus, a random surface made of protrusions and depressions is generated. Surfaces can be characterized according to their composition, roughness direction and height distribution. In this work we will focus on homogeneous (isochemical) isotropic Gaussian random surfaces.

In what follows, we will shortly recall the main theoretical approaches available in the literature to generate random surfaces. Then, a more general method will be detailed and further characterizations and comparisons with real surface will be shown. Finally, we present how to apply the method to molecular dynamics sample fabrication.

ROUGHNESS MODEL THEORY

Patir[12] proposes a method to generate parameterized (i.e. including controllability) three-dimensional rough surfaces using an autocorrelation function (ACF) which imposes coherent modulations of the surface. Based on weighted random submatrices summations (each submatrix corresponding to a guess surface subset), biased-random rough surfaces with Gaussian or non-Gaussian distribution are obtained:

$$z_{ij} = \sum_{k=1}^{n}\sum_{l=1}^{m} a_{kl}\eta_{i+k,j+l} \qquad \begin{matrix} i = 1,2,\dots,N \\ j = 1,2,\dots,M \end{matrix} \tag{1}$$

Where $\boldsymbol{z_{ij}}$ is the roughness height matrix (i and j scaling with cartesian positions), $\boldsymbol{\eta_{i+k,j+l}}$ is an (N+n)X(M+m) matrix of independent and identically distributed random numbers with zero mean and unit standard deviation. $\boldsymbol{a_{kl}}$ are weight coefficients that will be determined based on the desired ACF.

By definition[16,17], the ACF can be expressed by sampling intervals Δx *and* Δy as:

$$R_{pq} = R(p.\Delta x, q.\Delta y) = E\left(z_{ij}.z_{i+p,j+q}\right) \quad (2)$$

Where R_{pq} is the autocorrelation matrix terms and $\boldsymbol{E\left(z_{ij}.z_{i+p,j+q}\right)}$is the expectation value of $\boldsymbol{z_{ij}.z_{i+p,j+q}}$. $\boldsymbol{\eta_{ij}}$ values are generated independently with variance and mean respectively equal to 1 and 0, the expectation value of $\boldsymbol{\eta_{ij}.\eta_{kl}}$ can be expressed as follows:

$$E\left(\eta_{ij}\eta_{kl}\right) = \begin{cases} 1 & if\ i = k, j = l \\ 0 & otherwise \end{cases} \quad (3)$$

Based on equations (1) to (3), the general expression of the $\boldsymbol{a_{kl}}$ autocorrelation coefficients can be written as:

$$R_{pq} = \sum_{k=1}^{n-p}\sum_{l=1}^{m-q} a_{kl}a_{k+p,l+q} \qquad \begin{matrix} p = 0,1,\dots,n-1 \\ q = 0,1,\dots,m-1 \end{matrix} \quad (4)$$

Equation (4) is equivalent to a system of nonlinear equations that can be solved using an iterative method (e.g. Newton-Raphson) to find the autocorrelation coefficients $\boldsymbol{a_{kl}}$. A complete detailed description of this method using linear transformation is provided by Patir[12].

Although the linear transformation method is a simple way to obtain random rough surfaces, it depends on an iterative method with doubtful convergence conditions. An improvement of this methodology was proposed by Hu et all.[13]. By taking a discrete Fourier transform of equation (1), the height distribution can be expressed in terms of their spectral components:

$$Z\left(w_x, w_y\right) = H\left(w_x, w_y\right)A\left(w_x, w_y\right) \quad (5)$$

Where $Z\left(w_x,\ w_y\right)$ and $A\left(w_x,\ w_y\right)$ are the respective Fourier transform of the height and the random number matrix. $H\left(w_x,\ w_y\right)$ is the transfer function related to the ACF (i.e. the Fourier transform of ACF coefficients) and in 2D can be expressed as:

$$H\left(\omega_x, \omega_y\right) = \sum_{k=1}^{n}\sum_{l=1}^{m} a_{kl}\ e^{-jk\omega x}e^{-jl\omega y} \quad (6)$$

Determining $\boldsymbol{a_{kl}}$ coefficients is equivalent to apply a digital filter constraining the expected frequency response. To generate a sequence of randomly distributed heights with a deterministic autocorrelation function $\boldsymbol{R_{pq}}$, the power spectral density (the PSD, the Fourier transform of the ACF) has to be computed[16,17].

We denote $\boldsymbol{S_{zz}}$ as the PSD of the ACF and $\boldsymbol{S_{\eta\eta}=C}$ (the PSD of $\boldsymbol{\eta_{ij}}$) as a constant due to $\boldsymbol{\eta_{i+k,j+l}}$ definition. Assuming a filter defined only by real numbers including coefficients that satisfy certain symmetry relationships, $H\left(w_x,\ w_y\right)$ can be rewritten as[16,17]:

$$H(w_x, w_x) = \left(S_{zz}\left(w_x, w_y\right)/C\right)^{1/2} \quad (7)$$

After specifying the desired ACF, obtaining the respective PSD ($\boldsymbol{S_{zz}}$) and calculated the frequency response (equation (7)), the height coordinates can be obtained using the convolution sequences expressed in equation (1) where the $\boldsymbol{a_{kl}}$ are obtained by applying the inverse Fourier transform to equation (7):

$$a_{kl} = \frac{1}{nm}\sum_{w_x=-n/2+1}^{n/2-1}\sum_{w_y=-m/2+1}^{m/2-1} H\left(w_x, w_y\right)e^{-jkwx}e^{-jlwy} \quad (8)$$

A rigorous detailed description of this method can be found in the original article of Hu[13]. In the same reference, an improvement of the method based on fast Fourier transform (FFT) and the generalization for non-Gaussian distributed random surfaces can be found. One of the conclusions reported by Hu is that this methodology constitutes an extension of the linear transformation method and is mathematically identical to the time series model proposed by Watson et al.[14]. This model avoids the non-linear system of equations used in Patir's method by introducing a change of domain. The $\boldsymbol{a_{kl}}$ (equation 8), will have a non-trivial expression if the filter proposed is not linear and perfectly symmetric which will affect the roughness of the surface.

In this work, we propose an alternative method adapted from the work of Sjodin[18]. Random surfaces can be described using two main constituents i.e. a random number generator and a periodic expression. These two contributions can be combined using a convolution equation very similar to the one proposed in equation (1). Usually, a periodic function has the form of a cosine (or a sine). 2D rough surfaces can be obtained as the superposition of elementary periodic functions:

$$z_{xy} = \sum_{m=0}^{M}\sum_{n=0}^{N} A * cos(2\pi(mx + ny) + \varphi) \quad (9)$$

A is the amplitude and $\boldsymbol{\varphi}$ the phase of the function. Inside the periodic component of equation (9), the wavenumbers *m* and *n* induce a gradual change in the spatial frequency. For generating random surfaces it is necessary to perturb the phase and amplitude component in a non-deterministic way. Compared to the models of Patir and Hu, the amplitude can be set as a matrix of Gaussian distributed random numbers with specified mean and standard deviation ($\boldsymbol{A_{mn}}$). However, in order to obtain a more realistic rough surface two more contributions to equation (9) should be made: (i) the phase angle $\boldsymbol{\varphi}$ can take any value in the interval $[-\boldsymbol{\pi/2}, \boldsymbol{\pi/2}]$ with the same probability, (ii) mathematically, frequency and amplitude are correlated.

Therefore, Sjodin[18] proposed to redefine $\boldsymbol{A_{mn}}$ as a matrix of random numbers multiplied by c :

$$fc = (m^2 + n^2)^{-b/2} \tag{10}$$

Now, *b* is the spectral exponent and it will parameterize how fast the high frequencies are attenuated. Putting all together; a final expression for the desired random surface can be written as

$$z_{xy} = C1 * \sum_{n=-N}^{N} \sum_{m=-M}^{M} fc * G(n,m) * \\ * cos\big(2\pi(mx + ny) + U(n,m)\big) \tag{11}$$

Where $\mathbf{G(n,m)}$ and $\mathbf{U(n,m)}$ are the matrix array of normal or Gaussian distributed and Uniform distributed random numbers. C1 is a normalization factor. *x* and *y* are the cartesian coordinates. Equation (11) is periodic in the interval $x,y \in \{0,1\}$. It can be avoided by taking values inside or outside of that boundary. The fact that *n* and m can be positive or negative makes the surface to be generated with no preferred oscillation direction. The spatial frequency is controlled by the maximum number that n or m can reach. It means; if n and m can go until maximum values *N* or *M* then the shortest wavelength in x and y direction will be in the order of $\lambda_x = 1/N$ and $\lambda_y = 1/M$. Euler relationship tells that equation (11) can be seen as a discrete cosine transform or discrete Fourier transform where just the real part is considered. This results in a familiar relationship obtained before during the introduction of Hu methodology. A more detailed description in the previous methodology which leads to equation (11) is provided by Sjodin[18].

The degree of roughness is associated with the variation of height with respect to a central line. In the case of an experimental surface, a mathematical expression for autocorrelation should be given. This is not necessary for modeling random surface since the correlation length is known a priori. Different statistical parameters (mean, MS , skewness and kurtosis) are used to characterize the roughness of the surface[19-21]. If the final height for random surface follows a Gaussian distribution the standard deviation *(σ)* represents the main parameter for such characterization. If the final height is not normaly distributed other parameters should be considered. We start defining the mean line value:

$$l = \frac{1}{N.M} \sum_{i=1}^{N.M} z_i \tag{12}$$

With this we can calculate the standard deviation (σ) and the root mean square (RMS) like

$$\sigma = \sqrt{\frac{1}{N.M} \sum_{i=1}^{N.M} (z_i - l)^2} \tag{13}$$

$$RMS = \sqrt{\frac{1}{N.M} \sum_{i=1}^{N.M} {z_i}^2} \tag{14}$$

If the mean line value is strictly zero the values of σ and RMS will match. These two parameters should be enough to characterize the roughness of any surface but they do not give any information about the final distribution. For a complete mathematical description we use skewness (sk) and kurtosis (K).

$$sk = \frac{1}{\sigma^3 . N.M} \sum_{i=1}^{N.M} (z_i - l)^3 \tag{15}$$

$$K = \frac{1}{\sigma^4 . N.M} \sum_{i=1}^{N.M} (z_i - l)^4 \tag{16}$$

The parameter *K* and *sk* characterize the shape of any data. The *sk* measures the lack of symmetry in the distribution shape while *K* characterizes the degree of flatness with $sk = 0$ and $K = 3$ in the case of normally distributed data.

For MD samples using rough surfaces, the atoms positions and height coordinates must be correlated. With that, the relation between the Δx and Δy to the cell parameter and the crystal structure used in the model is considered. The coordinate of the atoms can match the coordinates where the height of the random surface is obtained. Otherwise a tolerance in function of the atom size should be considered. Notice that better resolution can be found if a Nyquist criterion is considered. Once the surface is generated we should apply a displacement in Z direction in order to make the higher value in the random surface to match the Z value of the uppermost atoms layer. An algorithm will detect the number of layer affected by the interception with the random surface and remove the atoms over it. With that, the new atoms sequence is ready for MD with random surface at the top.

RESULTS AND DISCUSSION

We produce random surfaces using equation (11). The first step in the method is to define the number of iterations of the system. It will affect the spatial frequency of our output. We start setting *N*=*M*=30, and at the same time, defining the size of our random matrices. The random number generator algorithm guarantees a change in the internal state of the generator; otherwise identical surfaces will be obtained in any new generation process.

We allow the algorithm to generate four random surfaces without changing the parameters of equation (11) considering C1=0.01 and *b*=1.8.

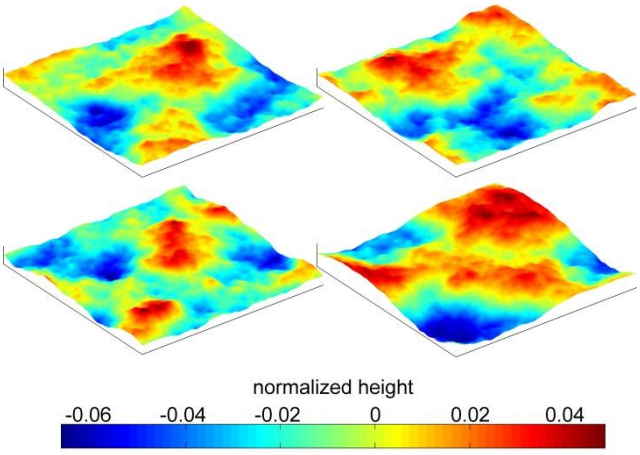

Figure 1. Four random surfaces produced with the same parameters (*N*=*M*=30, *b*=1.8, C1=0.01).

The images shown in Figure 1 are all produced with the same conditions, only the random number generator is affected. It means that this method is able to generate topologically different surfaces with the same physical parameters. It will be more useful if further characterization can be done in function of physical parameters (roughness) and not in function of topological information (height distribution).

Keeping the same values of *N* and *M* in equation (11) allows the spatial frequency to remain unchanged and the different morphologies at the surface to be controlled by the parameter b. We studied the topological changes in function of this parameter. Four surfaces with different roughnesses are observed in Figure 2.

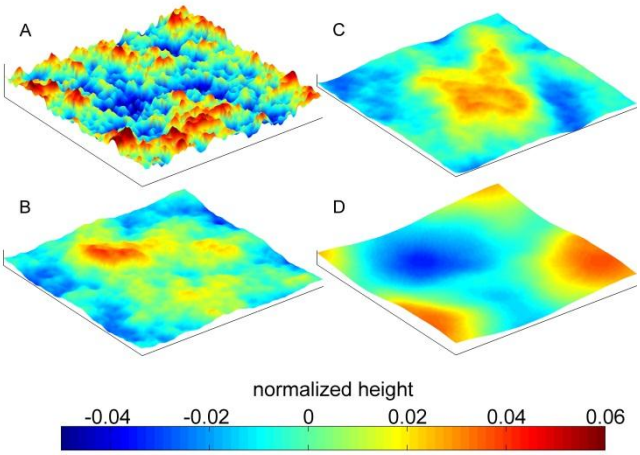

Figure 2. Changes in roughness degree in function of the spectral exponent considering *N*=*M*=30 andC1=0.01. A) *b*=1.2; B) *b*=1.8; C) *b*=2.2; D) *b*=2.8. The Cartesian coordinates in the figure are normalized

The degree of roughness vanishes with the increase of *b*. It implies that the model is able to mimic almost all kind of roughnesses with no similarity in topological information.

Figure 3 shows roughness versus *b* statistical dependency (***RMS***, mean central line, skewnees and kurtosis). The roughness characterization is generally defined by the ***RMS*** [19-21] (equation 14). Here we apply the method to the case where the mean central line (equation 12) is zero i.e. the ***RMS*** and σ have the same numerical value. Special attention should be paid to the parameter C1 in equation (11). It will directly affect the σ and ***RMS*** value according to the order of magnitude of C1. The ***RMS*** describes a descendent exponential shape followed by a linear behavior with the increase of b. The asymptotic part in *RMS* highlights the effective values of b between 0 and 3. $G(n,m)$ represents Gaussian distributed random numbers that will be linearly transformed by equation (11). It is expected that the final height will also be normally distributed. Figure 3B reveals the characteristics of Gaussian distribution for values of spectral exponent below 1 and progressive loss of symmetry in the distribution of final height while decreasing the roughness. This is mathematically expected since small roughness means fewer differences with respect to the central line.

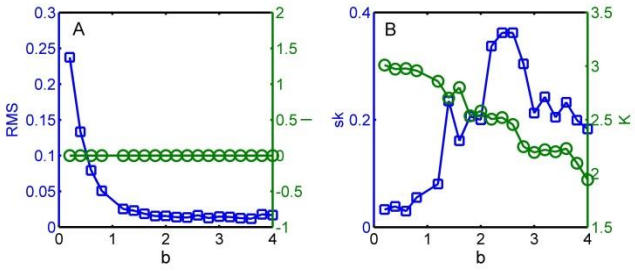

Figure 3. Statistical study of the spectral exponent. In blue and green respectively the ***RMS*** and mean central line (A); and the skewness and kurtosis (B) are shown.

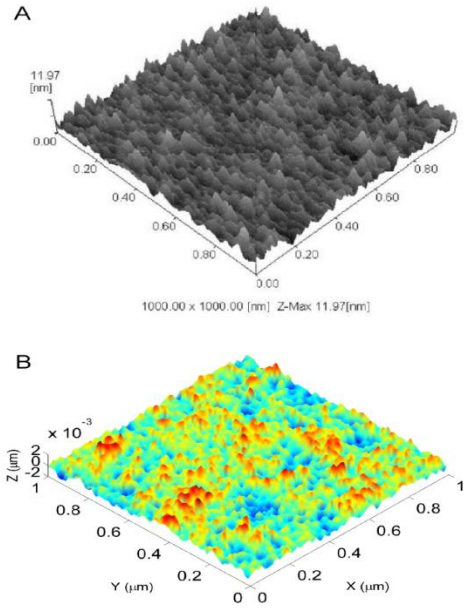

Figure 4. Comparison between AFM image A) and synthetic random surface B).

In order to verify the application of the model, we compare a real surface with one of our synthetically-generated ones. In other words, an AFM image of aluminum film reported by Fang[20] is compared with a

synthetic random surface generated with equation (11) (Figure 4). Light differences are observed due to the scanning system used to generate AFM images. It makes the output image to be produced with the maximum resolution depending on the scanned direction.

Next, we use equation (11) to design a sample made for MD simulations. The random surface is intercepted with the uppermost layer of a regular slab originally oriented along <100> directions (aluminum, 256000 atoms, fcc lattice, 4.02Å lattice parameter). This sample which is similar to a fully periodic thin film is the first step towards nano-object design based on the Sjodin random surface generation method. Ovito[23] is used to visualize the atomistic sample.

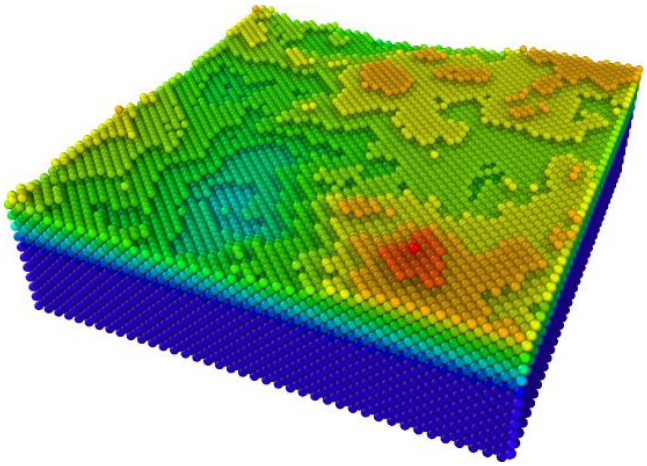

Figure 5. Surface design of an aluminum sample at the atomic scale using the roughness model (equation 11).

The structure showed in Figure 5 represents roughness at atomic scale. The conditions imposed for removing the atoms do not take any tolerance due to the atoms size. Furthermore only punctual position of the atoms center will be considered and insolated atoms will appear. Before being used for nanoidentation/nanocompression, the structure will have to be minimized and equilibrated under a given temperature to avoid unrealistic atoms positioning. Another way to change these conditions is to introduce tolerance for positions in function of the atoms radii; it is useful for the case when few atoms intercept the rough surface. Also it should be consider that, a much better resolution will be obtained if the number of atoms per layer in the sample increases.

Conclusions

Here are the main conclusions drawn by this study:

- Mathematical description of principal models for rough surfaces shows that the method proposed by Sjodin is the simplest and most complete one for random surface modeling. Statistical parameters are used for characterization of roughness in surface modeling indicating the changing law of roughness associated with the spectral exponent.
- Several configurations of roughness are provided and after comparison, a good correspondence with experimental AFM image is observed.
- Implementation of a periodic structure with rough surface at the atomic scale reveals that the method can be used for MD simulations.

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