**Minkowski Measures for Image Analysis in Scanning Probe Microscopy**

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**ABSTRACT**

This article describes the capabilities of a mathematical toolkit, namely the Minkowski measures, for the analysis of images acquired by means of scanning probe microscopy. The considered method, which is still largely unknown in this field, allows for a novel morphological characterization somewhat independent of the image size. One example of its application is reported, using homemade routines based on a widespread software platform, routines for which can be provided free upon request. The analysed images represent tapping-mode atomic force microscope topography of spin-coated thin films of organic molecules that are currently of great interest for the fabrication of organic light-emitting diodes.

**KEYWORDS**

global analysis, scanning probe microscopy, integral geometry, morphology, connectedness, object boundaries

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**INTRODUCTION**

Conventional analysis and manipulation of panchromatic monochrome (i.e. greyscale) pictures extracted from microscopy are based on filtering and/or processing performed either in direct space or in Fourier space [1]. When working in the direct (x-y) space, the brightness of the image, i.e. the height of the sample surface in the case of scanning probe microscopy (SPM) images, usually undergoes some local averaging or ranking operation taking place in a limited neighbourhood, called a kernel (e.g. a 3 x 3 data-point matrix), around the considered value (pixel). On the other hand, for operation in the Fourier space of image spatial frequencies the treatment is not local but global, and the peculiarity of an image containing periodic features (either properties of the sample such as an atomic lattice, or undesired artifacts such as an electronic noise) is identified by averaging its local morphological properties [2].

In this study we describe a method which is based on processing in the direct space, but accounts for global properties of the processed images. This technique is based on the calculation of three quantities, called Minkowski measures, which come from the field of integral geometry [3].

The definition of the Minkowski measures is based on splitting the considered images into two regions, H and L, of high and low levels of the third dimension z respectively, according to a threshold, \( z = \rho \), chosen appropriately within the full range of the z values. The simplest Minkowski parameter \( F \) is nothing else but the surface coverage. For example, when the high level surface subset H is supposed to consist of a different chemical or physical phase from the low level surface subset L, the respective area \( A_H \), normalized to the total image area \( A \), represents the percentage of image surface covered by phase H. Obviously, the quantity \( F = A_H / A \) is a pure numerical value, changing between 0 and 1. The second Minkowski measure \( U \) is usually described as the boundary length, and represents the global perimeter of either the H-type or the L-type domains (this is common to both regions). Therefore it has the dimensions of length, and its numerical value depends on the adopted unit (e.g. micrometres or nanometres). However, if the physical x-y sizes of the image are expressed in pixels (actually representing an area), \( U \) can be expressed in a universal, dimensionless ‘image’ unit, as the number of the square root of a pixel. Finally, the third Minkowski parameter \( X \) is the so-called Euler characteristic. This is the difference of the number of H-type and L-type domains in the image. This quantity is originally dimensionless, as it is described by a pure numerical, integer value. However, to account for the quantity of analysed sample surface area, \( X \) can be normalized to the total area, and henceforth expressed in units of length², or in universal image units of pixel¹.

While the physical meaning of the surface coverage \( F \) is clear, and this quantity is usually reported in most SPM characterizations of two phase (or simply two level) sample surfaces, the innovative content of the Minkowski analysis is contained in the other two measures, \( U \) and \( X \). The latter actually represents purely topological properties of the imaged pattern: its sign identifies the type of prevail- ing domains (H-type for \( X>0 \) and L-type for \( X<0 \)), while the absolute value \( |X| \) is correlated to the disconnectedness of the background phase (i.e. the opposite of the identified image features, for example the H regions in Figure 1). Indeed, on the same surface area the larger the \( |X| \) the higher is the number of separated domains of the two types. On the other hand, \( U \) is correlated to the curvature of the boundaries: for a given \( X \), the larger \( U \) becomes the more the respective domains are bent, such that they can fit a longer boundary length into the same surface area. In fact, \( U(X) \) represents a mean boundary length, or, from a different point of view, \( X/U \) is an index of mean convexity of the H domains (negative if they are predominantly concave).

**MATERIALS AND METHODS**

The simplest situation for which the analysis provided by the Minkowski measures is of interest, is the investigation of samples which show a two-phase separation on their upper surface. This is often the case, for example, for thin films of organic materials.

As a sample system, blends of a diamine derivative (TPD: N,N'-diphenyl-N,N'-bis(3-methylphenyl)-1,18-biphenyl-4,4’-diamine) and a thiophene-based compound (STO: 2,5-bis(trimethylsilyl)-thiophene-1,1-dioxide) were used in this study. The blends were deposited by spin-coating from toluene solutions with different relative concentrations.

Tapping-mode atomic force microscope (AFM) images were then acquired, which are shown in Figure 1. These blend films are of interest in the field of organic light-emitting diodes (LEDs) as active materials for the devices [4]. Their efficiency is negatively affected by the occurrence of phase separation [5] which depends on the relative concentrations of the blend components. While the overall molar concentration was kept constant (equal to 50 mM), the relative STO:TPD molar ratio was varied in the range of several orders of magnitude.
concentration was changed from 20% (Fig. 1a) to 60% (Fig. 1b) and 80% (Fig. 1c). Unfortunately, while both real-space image processing based on kernel operations and frequency-space processing based on Fourier transformations are widely available in most commercial software packages, this is not yet the case for the U and \( H \) Minkowski measures. Therefore, dedicated home-built routines were developed for their computation. A comparatively easy recipe for extraction of the Minkowski measures from pixel based image maps was found in [6]. This was used for writing dedicated processing routines in Matlab (unsupported routines are freely available on e-mail request). The most natural choice of the thresholding value \( \rho \) is taking it as equal to the median value of the scanned z range, i.e. \( \rho = \rho_{\text{med}} = \frac{z_{\text{max}} - z_{\text{min}}}{2} \). This criterion, when applied to the images in Figure 1, results in the posterization of the above pictures into the binary (black and white) ones shown in Figure 2. The values of the Minkowski measures for the binary images in Figure 2, as calculated from our routines, are reported in Table 1, together with other characteristic quantities typical of a more usual SPM morphological analysis. In particular, the dominant length \( \lambda \) obtained by Fourier analysis [8] has been calculated from the greyscale images in Figure 1. Further, from the binarized images (Fig. 2) the mean size \( D \) of the features has been derived. \( D \) is accompanied by the respective value of the standard deviation \( \sigma \) calculated within each image. This is quite high (up to 76% of \( D \)) due to the fact that the features are not round, and \( D \) is the result of isotropically averaging over \( 2\pi \) in the image plane around the feature centre. On the other hand, the deviation of the feature shape from circular symmetry is expressed by the mean eccentricity factor \( e \) (the ratio of longest feature diameter - major axis - to its size in the orthogonal direction - minor axis).

**RESULTS AND DISCUSSION**

Minkowski measures (fixed threshold) and relation to standard morphological parameters

The correlation between morphology and phase separation for the samples in Figure 1 has been demonstrated elsewhere [7], and here it is assumed as a starting point that the different levels of height in the images correspond to different phases of the film material. From Table 1 it turns out that the surface coverage \( F \) is decreasing for increasing STO concentration, from Fig. 1a to 1c. Indeed, in Figure 2a the H regions are largely dominating (79% of the image), whereas the progressive decrease with increasing STO is such that in Figure 2c the H and L regions share equally the sample surface area (49% and 51%, respectively). Obviously, the presence of excess STO in the blend generates some phase separation of the film components. A deeper insight is provided by \( \chi \). This is negative for all of the three samples, meaning that the regions of the images that can be identified as ‘features’ are the black (L) ones. In other words, the patterns are dominated by holes as the main characteristic, while the top (H) regions represent a background. Furthermore, the absolute value of \( \chi \) is in general rather low (approximately constant in Figs 2a and 2b, 0.016 and 0.017). This means that these holes generate only a low disconnectedness in the high background. In particular in the case of highest STO concentration (Fig. 2c), \( \chi \) is still negative but very small (|\( \chi | = 0.002\)). This means that neither the holes nor the elevated ripples of Figure 1c prevail in the image, such that the kind of dominant feature is undefined. Therefore, both the H and the L regions have a quite high connectivity, and the film shows a bicontinuous morphology. Finally, looking at U one can
see that the second and the third samples in Figure 2 have an almost identical U value, higher than the first one. Indeed, the two morphologies of Figures 1b and 1c are somewhat similar to each other, as they present elongated features (holes and holes/ripples, respectively), while in Figure 1a the holes are rather round. This is also consistent with the lower width of the feature size D distribution of Figure 2a (relative standard deviation of 41%). The difference in shape of the features between Figures 2a and 2b is well represented by their mean eccentricity \( e \), which changes significantly. Finally, one can observe that \( \chi \) is roughly the same for all the three cases in Figure 1, thus it does not help to discriminate among them differently from \( U \) and \( X \). This is so since, even if the mean feature size changes, the typical (averaged) sum of the distance among adjacent features plus feature size (a sort of pseudo-period) remains obviously rather constant.

Plots of Minkowski functionals (varying threshold)

Even if the simple choice of the threshold as equal to the median \( z \) value seems to give a reasonable transformation of Figure 1 into Figure 2, some level of arbitrariness still remains in this procedure of splitting the image areas into H-type and L-type regions. As a result, for example, in Figure 2c some of the tiny H domains inside the large L islands have disappeared. It is for this reason that a more thorough Minkowski analysis has to account for the results of thresholding the original images at all the possible values of \( \rho \). The Minkowski measures as a function of the threshold value (Minkowski functionals) are represented by the respective plots \( F(\rho) \), \( U(\rho) \) and \( \chi(\rho) \) (see Fig. 3).

Taking one step back to the grey-scale images of Figure 1, the resulting Minkowski functionals have been calculated with our routines. As the images were 8-bit grey levels in the 3rd (z) dimension, there were 256 possible values for \( \rho \) (between \( z_{\min} \) and \( z_{\max} \)), conventionally set to zero, and \( z_{\max} \). However, it is worth noting that for all images, see legend to Fig. 1). These dimensionless ‘image’ \( z \) units have been represented on the horizontal axis of Figure 3. For \( U \) and \( \chi \), integer multiples of their values have been represented in the plots, in order to use the same numerical scale as for \( F \). Obviously, the previously considered individual values of \( F \), \( U \) and \( \chi \) (Table 1) correspond to intercepts of the plot curves for \( \rho = 128 \).

Generally, the first feature that can be extracted from the Minkowski plots is the nature of the \( z \)-value distributions in the considered images, if mono- or multi-modal. In particular, for a single mode distribution only one maximum shows up in \( U(\rho) \), as is clearly the case for Figures 3a and 3b. Accordingly, the \( f(z) \) has only one flex point, roughly at the same position of the U maximum. In fact, in Figure 3c, even if no separated peaks for \( U(\rho) \) show up, still the only maximum appears as a wide plateau, with the left edge sharper than the right one. This can be due to a superposition of two maxima that cannot be separated.

The hypothesis is confirmed by the two flex points in \( F \). These are all hints of phase separation between the two blend components in Figure 1c. Even if the value of the \( U(\rho) \) maximum is rather similar to that of Figure 3b, the shape of the plot here tells us that the two patterns are quite different, and can be classified as two different types. On the contrary, for Figures 3a and 3b the peak values of \( U \) are more different but the shapes of the profiles are similar, such that no real hint of complete phase separation can be found in the intermediate case (Fig. 1b), but the only effect of the feature elongation is an increase in the boundary length (along with the mean curvature \( \chi \)).

In conclusion, the Minkowski measures can help gain a deeper understanding of the patterns that are formed in the SPM images of complex systems, like, for example, thin films of molecular blends which are likely to segregate and/or separate into islands due to crystal formation. In one particular case of AFM images it has been demonstrated that the Minkowski measures account for significant morphological information contained in the images which are not easily accessible to other parameters determined with the classical tools of image analysis.

It is our opinion that calculation of the Minkowski measures will soon be integrated into most software packages for the analysis and processing of three-dimensional scanning probe microscope images.

### Table 1: Minkowski measures extracted from the images in Figure 2.

<table>
<thead>
<tr>
<th>STO - TPD Ratio</th>
<th>( \chi(\rho) )</th>
<th>( U(\rho) )</th>
<th>( D )</th>
<th>( \lambda )</th>
</tr>
</thead>
<tbody>
<tr>
<td>20%</td>
<td>-0.016</td>
<td>0.128</td>
<td>370 ± 150 (41%)</td>
<td>1.0 ± 0.2</td>
</tr>
<tr>
<td>60%</td>
<td>-0.017</td>
<td>0.216</td>
<td>380 ± 290 (76%)</td>
<td>1.1 ± 0.3</td>
</tr>
<tr>
<td>80%</td>
<td>-0.002</td>
<td>0.204</td>
<td>430 ± 260 (60%)</td>
<td>0.9 ± 0.2</td>
</tr>
</tbody>
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### REFERENCES

8. The dominant length is defined as \( l = 1/k_{max} \) where \( k_{max} \) is the position of the maximum of the curve derived by isotropically averaging the radial profiles of the 2D FFT (fast Fourier transform) maps, calculated from the AFM images. The reported uncertainties come from averaging over several similar images of the same samples.