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Morphology of composite films: a computer study

Stanislav Novák
Department of Physics, J.E.Purkynì University,
Èeské mládeže 8, 400 96 Ústí nad Labem, Czech Republic

Rudolf Hrach
Department of Physics, J.E.Purkynì University, Èeské mládeže 8, 400 96 Ústí nad Labem, Czech Republic
and Department of Electronics and Vacuum Physics, Charles University,
V Holešovièkách 2, 180 00 Praha 8, Czech Republic

Miloš Sobotka
Department of Electronics and Vacuum Physics, Charles University,
V Holešovièkách 2, 180 00 Praha 8, Czech Republic

Four models, all with spherical objects, were prepared to study the possibility of at least partial reconstruction of 3D structures from their 2D sections. The criterion to judge whether the structure consists of constant size objects is given. The errors caused by the influence of limited numbers of objects and of digitization are discussed. It is illustrated the usage of the integral transform method to unfold the structures. The results of the film analysis are presented.

1. Introduction

Morphological analysis of composite metal/dielectric thin films is important both for the characterization of the films themselves and for the analysis of their properties. Theoretical approach to the problem often leads to nearly invincible difficulties.

The main goal is to derive some spatial characteristics of the film such as the average number of objects per unit volume N_V, the distribution function of the characteristic dimensions of objects D_V, or the mean characteristic dimension d_V. We usually are able to find out some statistical parameters from planar sections of the films or from their projections. Assuming further spherical objects and knowing the mean number of section circles per unit area N_A and the distribution function of diameters of section circles D_A it is possible to solve the important problem, first solved by Wicksell 0. This leads to Abel integral equation

$$d_A(r) = \frac{r}{d_V}\int_r^{\infty}\frac{d_V(x)}{\sqrt{x^2-r^2}}\,\mathrm{d}x \tag{1}$$

where δ_A and δ_V is density function of the distribution function D_A and D_V, respectively. Its solution can be writen [2] as

$$d_V(r) = -\frac{2d_V r}{p}\int_r^{\infty}\frac{1}{\sqrt{x^2-r^2}}\frac{\mathrm{d}}{\mathrm{d}x}\left[\frac{d_A(x)}{x}\right]\mathrm{d}x \tag{2}$$

for $r \geq 0$. In the special case of the constant sphere diameters, we find out that the equation (1) leads to

$$d_A(r) = \frac{r}{d_V\sqrt{d_V^2-r^2}} \qquad \text{for } 0 \leq r \leq d_V\,. \tag{3}$$

If we have the section then the mean and the variance of the circle diameters are

$$d_A = \frac{pd_V}{4} \tag{4}$$

$$s_A^2 = \frac{\left(32-3p^2\right)d_V^2}{48}\,. \tag{5}$$

We can use these equations to test whether the structure has the constant sphere diameter.

A computer model is more convenient for morphological analysis of more complex systems. The islands in composite films with low metal volume fraction can be nearly spherical and randomly distributed [3]. In our paper we deal with such a kind of composite films, islands of which have different diameter and are randomly distributed in a polymer matrix.

2. Models

We prepared four different models of composite films to analyse the influence of various conditions and phenomena on the unfolding process. All basic models have the main region 1000×1000×100 pixels (*x*, *y*, and *z* directions) and 100 pixels wide border for a compensation of boundary effects. The number of generated objects is always the same - 1000 objects in the inner part (except for the discussion of errors). They are randomly generated, not touching each other – in feet, the minimum distance between objects is another parameter of our models. From the chosen filling factor (0.2) of the film and the number of objects, we could calculate the sphere diameters.

Once we have generated the composite structure we make a random section parallel to the (*x*, *y*)-plane. In the next four figures, we can always see the projection of different

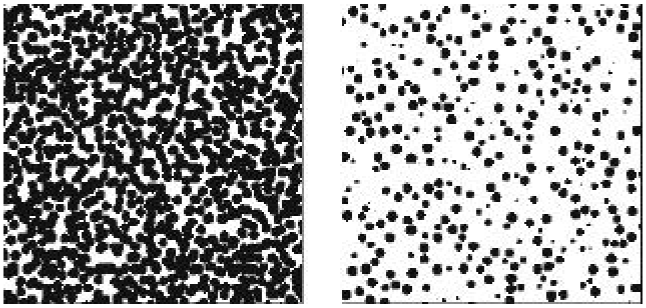

FIG. 1. Model M1 with constant radii of spheres, R = 16.84.

structures generated by our models, on the left side, and the random section of each structure, on the right side. The pixel is the basic length unit everywhere.

Fig. 1 shows the model M1 of the film with constant object radii R, whil Fig. 2 shows the model M2 where the radii of sphere objects are uniformly distributed in the interval $<R_1, R2>$. In Fig. 3 we can see the model M3 of the film with Gaussian (normal) distribution of the diameters of objects, and the superposition of two Gaussian curves is ground for the generating of object diameters in the Fig. 4 - model M4. Here are one part of spheres generated with a mean diameter R_1 and second part of spheres with another mean diameter R_2.

3. Moments of the object distribution

When we take into account a random section of a composite film with a constant diameter D of spheres, we can see a set of circles with diameter d in the interval <0, D>. If we consider these circle diameters as random variables then their mean and the variance have to be given by Equations (4) and (5), respectively. As was previously noted we can use these formulae to test whether the object diameters are actually constant. In this case it is also possible to derive the size of the sphere objects. The condition to be the sphere objects constant, is:

$$\frac{\sigma_A^2}{\left(\bar{d}_A\right)^2} = \frac{32-3\pi^2}{3\pi^2} \approx 8.0759 \cdot 10^{-2} \qquad (6)$$

Applying the same analysis to the structures in our four models M1 to M4 with the parameters presented above we

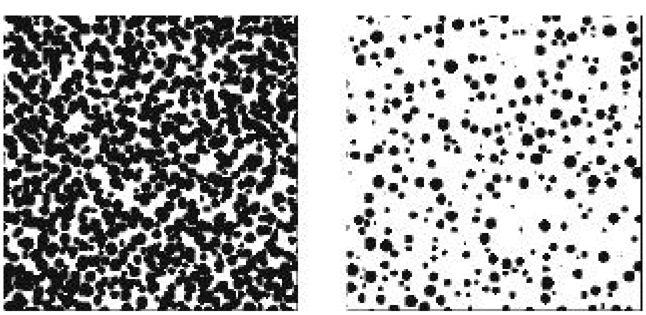

FIG. 2. Model M2 with uniform distribution of sphere radii in the interval $<R_1, R_2>$, R_1 = 10.0, R_2 = 23.0.

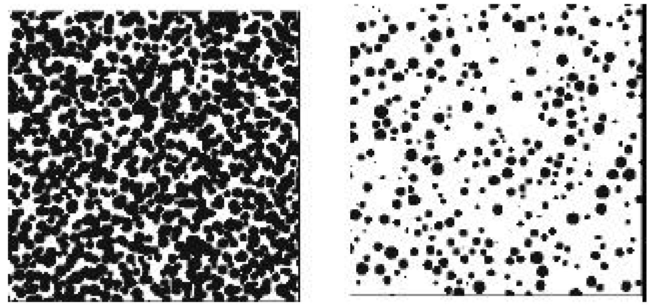

FIG. 3. Model M3 with Gaussian distribution of sphere radii with parameters: mean radius R = 15.8, standard deviation σ = 0.25

get the next values of the ratio variance to mean squared: $7.88\cdot10^{-2}$, $1.20\cdot10^{-1}$, $1.38\cdot10^{-1}$ and $1.88\cdot10^{-1}$. In accordance with the given models, we see that we can exclude the models M2 to M4 from our further thinking because of the formula (6). However, the value for model M1 corresponds to the criterion (6) not exactly. It leads to consideration about the precision of the results taken from experimental analysis of the planar sections of the composite films. The possible uncertainty has the two reasons:

- fluctuations caused by the limited number of objects in the sample
- image digitization error of the sample

The next information, that the analysis of the section of composite structure yields, is the structure-filling factor. It is equal to the coverage determined from the section, in the case of uniformly distributed objects. That fact holds for any size and form distribution of objects, as was showed already by a French geologist Delesse in 1847 [4]. Nevertheless, in this case the results are influenced by the both above-mentioned possible uncertainties.

In the next two sections, we discuss the errors of analysis of composite film sections based in all cases on the model M1.

Influence of limited number of objects

In left Fig. 1 is shown the structure consisting of 1000 objects in active region. The planar section (in the

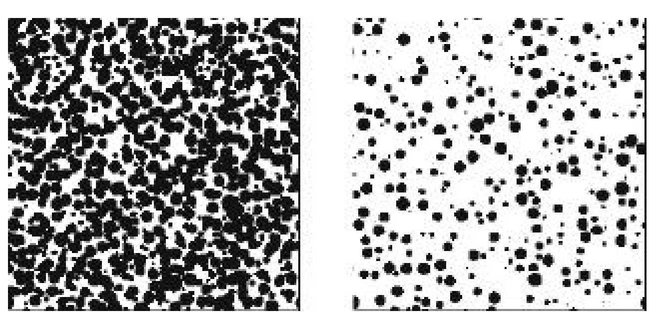

FIG. 4. Model M4 where the distribution of the sphere radii has resulted from the superposition of two Gaussian distributions with the parameters: mean radius R_1 = 10.0, standard deviation σ_1 = 0.25, relative number of the spheres 50%; mean radius R_2 = 20.0, standard deviation σ_2 = 0.10, relative numbers of the spheres 50%.

right) contains about 300 circles and we got for the ratio (6) the value $7.88 \cdot 10^{-2}$ from statistical analysis of their diameters. Repeating the computer experiments we get a set of values according Gaussian distribution *N(ì , ó)* with the mean *ì* = 8.075910 $^{-2}$ - compare to Eq. (6) - and with standard deviation *ó* = 9.06210 $^{-3}$.

In real cases, we usually have no possibility to repeat sectioning of the structure and we have to do our analysis from one section. Then is suitable to apply the well-known 'three sigma rule'. It says it is extremely improbable to have one particular measured value differing more than three sigma from the exact value, during measurement of any physical value with Gaussian distribution of magnitudes. In our situation, the value of sigma will depend on the number of objects in the section. Table 1 illustrates how many objects we have to take into consideration to get the result with desired precision.

We can conclude that the formula (6) is accepted with the uncertainty given in the Table 1 for the structures filled by identical sphere objects. Another results confirm the possibility to estimate the structure-filling factor from the coverage in a chosen section.

Influence of digitization error

The previous results were obtained for ideal case when each circle diameter *d* would be derived exactly, e.g. by analogue measurements on microphotograph at a big magnification. In fact, an obvious analysis is done with certain magnitude of each pixel on the digitized microphotograph. It brings the other error to the results, which is superimposed to the previous uncertainty.

In order to analyze it we used the data from the model M1. We have digitized this section area with various magnitudes of pixels, now. The results of the statistical analysis we see in Table 2. The first column gives how many pixels we can distinguish on the section area. In the second column is number of pixels for the biggest circle diameter D_{MAX} and third one gives relative error of the obtained result $s_A^2/(d_A)^2$.

The both types of the errors are superimposed in real cases when we analyze existing planar sections of composite films. Comparing both the Tables 1 and 2 we see that the influence of the limited number of objects is mostly more important.

Table 1. Reached precision of the results for different number of objects.

Number of objects in the section	3 σ [%]
30	98.6
300	30.7
3,000	10.6
30,000	3.04

Table 2. Statistical error caused by digitization.

Area of section [pixels]	Diameter D_{MAX} [pixels]	Error of digitization [%]
500×500	17	3.1
1,000×1,000	34	1.8
2,000×2,000	68	0.5
4,000×4,000	135	0.1
10,000×10,000	337	0.05

4. Size distribution of objects

When the structures studied consist of constant size objects, the analysis based on the statistical moments is sufficient. However, in many cases the structures are formed by objects - often again spherical - with certain size distribution. The goal of the analysis is then to derive this size distribution. As perspective seems to be, the method based on the integral transform [5].

It is very difficult to determine the distribution of object sizes experimentally. Nevertheless, the objects in the composite layer have spherical shape in many cases. In these cases the method based on the so-called Chord Length Distribution of Dark Segments (CHLD dark) [5] is applicable and seems to be very perspective.

This method was previously used for the reconstruction of the discontinuous circular metal island radii (2D problem) from so-called “dark chords” [5]. Dark chords are segments on the randomly oriented or parallel lines (in the anisotropic case) distributed over the digitized

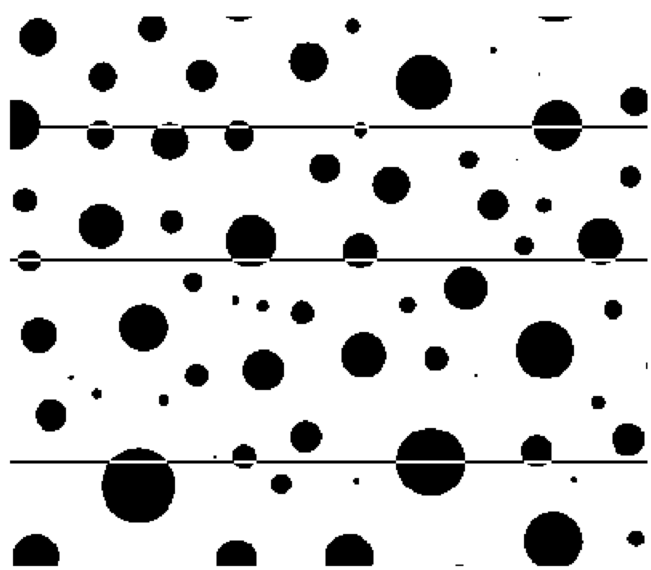

FIG. 5. Construction of the CHLD dark. Only segments that fully intersect an object are taken into account (white fragments)

2D microphotograph of the thin film intersecting the islands (Fig. 5). CHLD dark method uses the statistics (normalised histograms of lengths) of these chords as input data and produces distribution of the diameters on the output. If $P(D)$ is the distribution of the circular object diameters and $c(l)$ is the distribution of dark chords, a simple integral equation connects these two functions:

$$c(l) = \int_{D}^{\infty} P(D) \cdot \left[1 - \sqrt{1 - \frac{l^2}{D^2}} \right] \mathrm{d}D \qquad (7)$$

In three dimensions, instead of lines we shall have randomly oriented or parallel planes, and chord lengths correspond to areas of the circles that originate from intersection of the planes with objects. The integral equation remains valid.

In fact, this equation is classified as the Volterra integral equation of the first kind and this equation can be easily numerically solved by back-substitution. Its discrete equivalent is upper triangular matrix.

Because of the statistic nature of the input data and because of the their finiteness they seem to be “noisy”. During the solution some regularization must take place to avoid horrible oscillations of the finite solution [6]. Binomial filtering of both input and output data is suitable method that can accomplish this [7]. Unfortunately any regularization method causes the loss of the information. In this case discrete peaks (objects with the same radii) are widened into peaks with finite half-width (see Fig. 6). The input data for the reconstruction obtained from the planar section of model M1 are seen in Fig. 7.

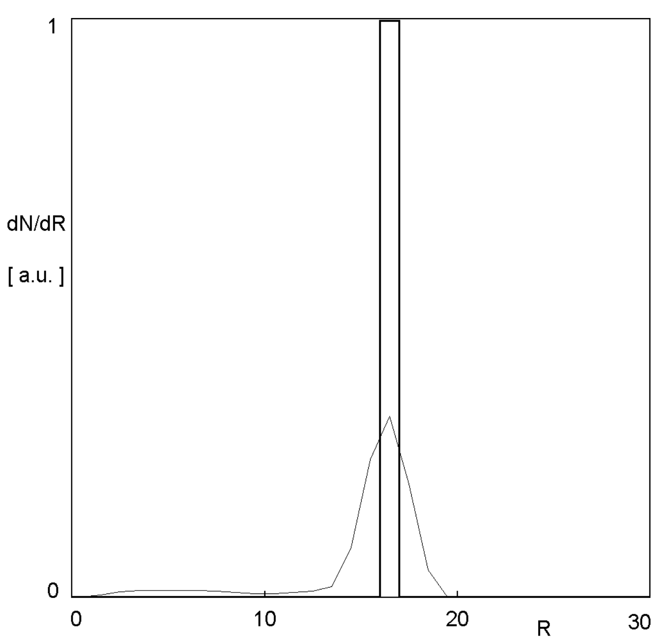

FIG. 6. Radius distribution in model M1 (Fig. 1), actual (histogram) and reconstructed (curve).

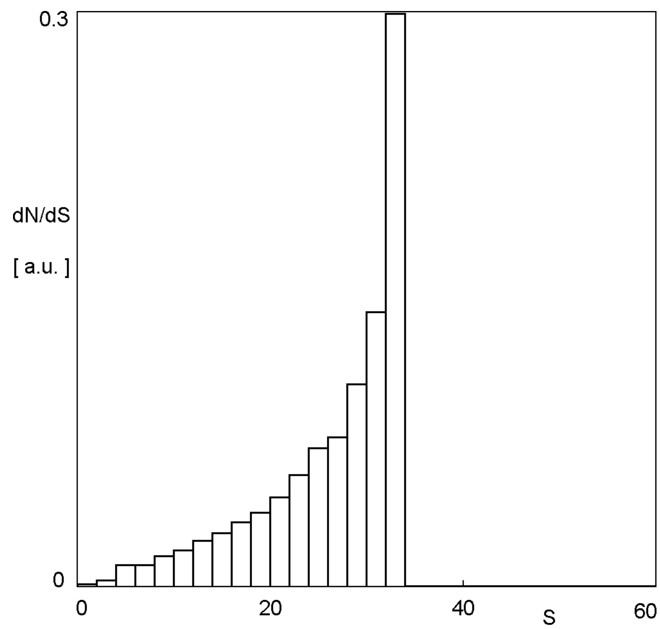

FIG.7. Size distribution of the circle areas in the planar section of the model M1 (Fig. 1).

The method reconstructs object sizes very well with two exceptions:

- Due to the regularization it cannot correctly reconstruct distribution of object sizes if the objects are of the same dimension. Nevertheless the position of the maximum of the reconstructed curve corresponds with the position of the discrete peak (see Fig. 6).
- Small dimensions are reconstructed badly. There are several reasons for it. The error during digitisation process manifests oneself mainly at small object sizes. Another error occurs during numerical solution. The back-substitution method computes first large sizes and these results are used for the computation of the smaller ones. Any error is propagated such way and distribution at small sizes is affected much more (see Figs. 8 and 9)

The use of this method can help us to find out the size distribution of objects in space. In the Figures 6, 8, and 9 are depicted both the actual size distributions by means of histograms and the size distributions reconstructed by the integral transform method from one section of the structure (continuous curve). These distributions are taken from our various models – with constant radii of spheres (Fig. 6) or with Gaussian distributions of sphere radii (Fig. 8 and 9).
It is evident that this method only approximates the right solution. Nevertheless, it provides valuable information about size distribution of objects although the primary information was not complete. It is probably impossible to gain it in other way.

5. Discussion

Our models are well useable to study the unfolding problems. Moreover, it is easy to transform them to study

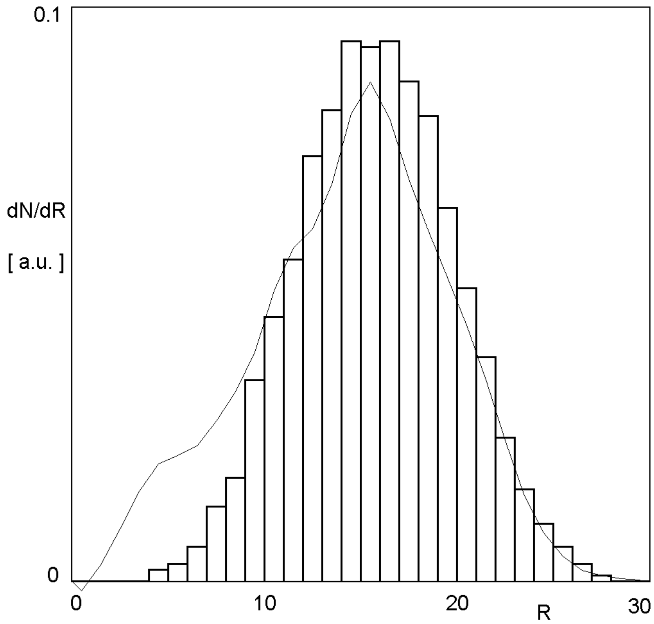

FIG. 8. Radius distribution in model M3 (Fig. 3), actual (histogram) and reconstructed (curve).structures with other forms of objects. We can conclude that the formula (6) is well acceptable to find out the structures with constant spheres but it is probably hard to say it for other forms of objects.

Even in case of spheres, the considerations about possible errors are important. The uncertainty, caused by limited number of objects or by digitisation, was shown. Both the types of errors are superimposed in real cases. We can say that the influence of the limited number of objects is mostly important for the possible error

The method of integral transform helps to reconstruct the size distribution of objects in 3D structures. We are convinced that the method is widely useable not only for spherical types of objects and that can be adapted for the determination of spatial distribution of objects, too. We are working on this problem to be able both to obtain the spatial distribution of objects from area occurrence in planar sections of composite films and to derive the parameters of composite films for other forms of objects.

Acknowledgments

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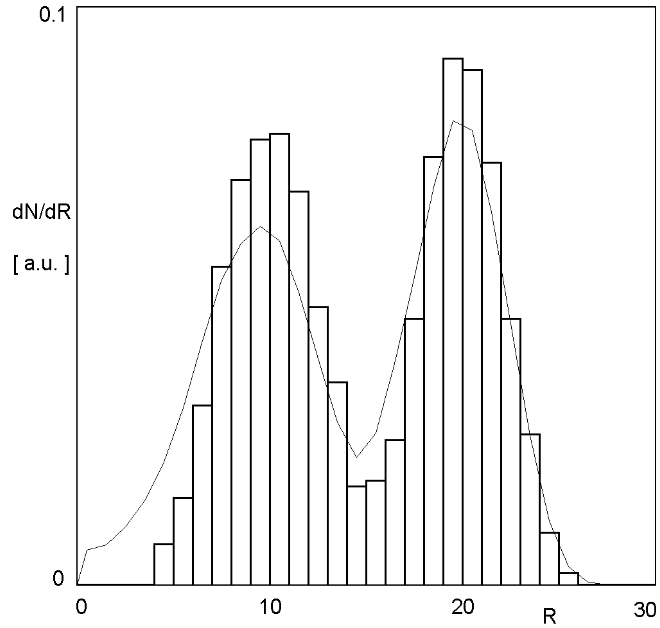

FIG. 9. Radius distribution in model M4 (Fig. 4), actual (histogram) and reconstructed (curve).

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