ISSN 1330–0008
CODEN FIZAE4

MORPHOLOGICAL DESCRIPTION OF COMPOSITE FILMS

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Received 7 April 1995

Revised manuscript received 1 August 1995

UDC 538.975

PACS 68.55.Jk

The initial stages of growth of thin metal films, in two-dimensions (discontinuous metal films on dielectric substrates) and in three-dimensions (plasma-chemically prepared composite films), are studied by a combination of direct measurement and modelling. For the analysis of photographs of films from the transmission electron microscope and the description of spatial distribution of objects, various methods of mathematical morphology were used. The interpretation of the derived characteristics and the extension of the analysis for composite films were performed by the computer experiment. As a result, morphological methods sensitive for the characterisation of distribution of objects in the third dimension were found.

1. Introduction

Very thin metal films deposited on dielectric substrates in the initial stage of their growth consist of individual islands. The basic information about these objects can be derived from the analysis of film micrographs obtained with a transmission electron microscope. As the typical micrographs consist of at least $10^3 - 10^4$ objects, a quantitative characterization of film morphology must be performed by methods of computational physics.

Conventional methods of thin film physics, such as thermal evaporation in vacuum, can be used for the preparation of discontinuous metal films in two dimensions (2D), while the plasma-chemical methods can produce composite films, where metal islands are located in the dielectric or polymer matrix and arranged in three dimensions (3D films). For the study of composite films, the micrographs are the main source of morphological information, too. However, in this case we have only two-dimensional projections of 3D objects. A part of information is lost and a description of film morphology is much more complicated.

In the study of thin film morphology, two kinds of problems arise. It is necessary to find convenient methods for quantitative characterization of the film micrographs and to interpret the derived characteristics, i.e. to establish the connection between morphological data and the physical processes which take part during film nucleation and growth. For the study od 2D films, methods based on mathematical morphology (see Ref. 1) are regularly used for the first task and main attention at present is devoted to the second problem (e.g. Schmeisser [2] or Blacher et al. [3]). For the characterization of composite-film morphology, the corresponding methods must first be created.

2. *Morphology of 2D films*

The theory of mathematical morphology deals with properties and relations of objects distributed generally in n dimensions (n = 1,2,3,...). In thin film physics, the following methods are most popular for the description of discontinuous metal films (2D):

1. Integral characteristics (information about the complete set of objects):

- concentration (the total number of objects in given sample);
- coverage (the relative area of micrograph covered by objects).

2. Size distribution (information about the individual objects):

- distribution of (equivalent) object radii;
- distribution of form factors of objects (form factor, FF, being the measure of deviation of object form from an exactly circular one).

3. Spatial distribution (information about the distribution of objects on the substrate):

- radial distribution function (or pair distribution function) RDF;
- distribution of nearest neighbours;
- covariance C;

- distribution of Wigner-Seitz cells of objects (or Voronoi tessellations);
- quadrat counts method, etc.

For 2D films, mathematical morphology was successfully applied and the correlation between experimental data and corresponding morphological characteristics was established. It was found: the nucleation process can be described best either with the help of integral characteristics or by means of RDF; the physical mechanisms of growth of small islands can be analysed by the size distributions of objects and by the distribution of Wigner-Seitz cells; the "quadrat counts" method and RDF are influenced substantially by the coalescence process and by the degree of randomness of spatial distribution of islands; the covariance function, C, brings simultaneously information about many physical processes which take part during film nucleation and growth; the radial distribution function, RDF, can be reconstucted from the set of distributions of N-th nearest neighbours (Hrach et al. [4]), etc.

To establish these facts, it was necessary to analyse many micrographs of discontinuous metal films prepared under various experimental conditions and to study them with the help of computer simulations. As this methodology is very promising, we shall apply it to the study of 3D films.

3. *Computer experiment*

For the analysis of the composite film morphology, a computer experiment was made. We can generate model "micrographs" of composite films with known properties, analyse them with various morphological algorithms and compare the results with known film parameters. This method can help us to establish which method is sensitive enough to reconstruct at least partially the distribution of islands in 3D.

During the projection of 3D structure on a plane, part of information is lost and this missing information must be replaced by some realistic assumptions. From the analysis of real micrographs made by Biederman and Martinů [5], it follows that in the low metal volume fraction composites, the islands are nearly spherical and often have the same diameters. In that case, the 3D composite film can be simulated by simple "hard sphere" model.

The model consists of an active area representing substrate (dimensions 1.0 $\times$ 1.0 relative units - in these units all lengths will be measured) surrounded by a larger part (total dimensions 1.75 $\times$ 1.50) in order to exclude the edge effects during generation of island structures and during evaluation of morphological characteristics. The third dimension, the thickness of the sample d, was variable. The distances on the substrate plane are denoted by h. The number of objects, N, is given relative to the active central part of the substrate.

The objects were represented by hard spheres with constant radii R_0. Their radius was derived from the given total number of objects N and from the expected coverage Θ. As a further parameter of the model, a "diffusion zone", DZ, was introduced (DZ represents the minimum distance between edges of objects and with its help, it is possible to influence the randomness of spatial distribution of objects).

4. *Morphology of 3D films*

We prepared several sets of model composite films, where in one set only the film thickness d varied. We processed these data by various morphological algorithms. Some results of our computer experiment are shown in Figs. 1 and 2 (in these model structures, the DZ was chosen to obtain the most regular structure of islands).

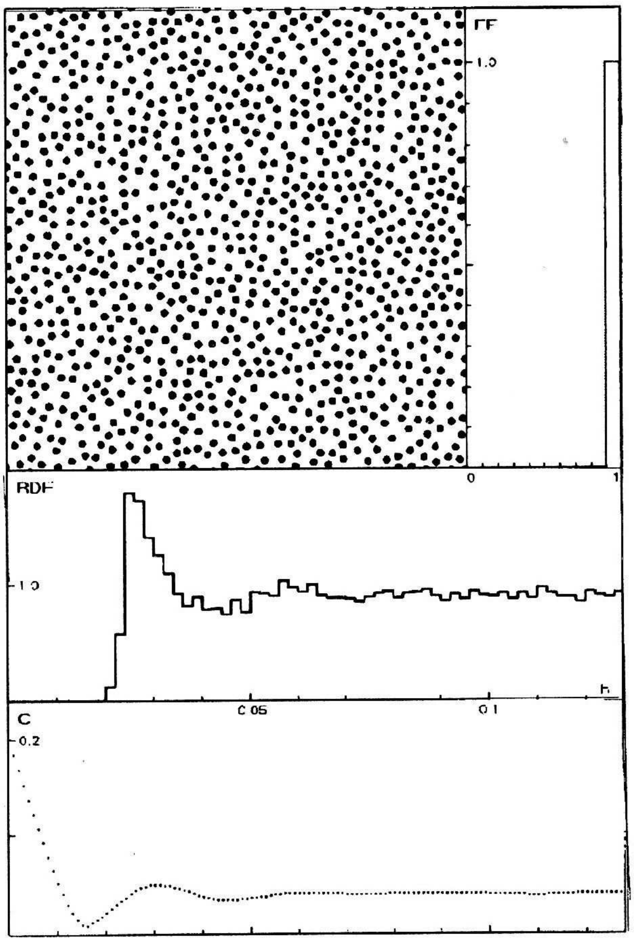

Fig. 1. Results of the model with parameters: $N = 1000$, $\Theta = 0.2$, DZ = *maximum for the given* N *and* R_0 *in 2D structure, and* $d = 4R_0$.

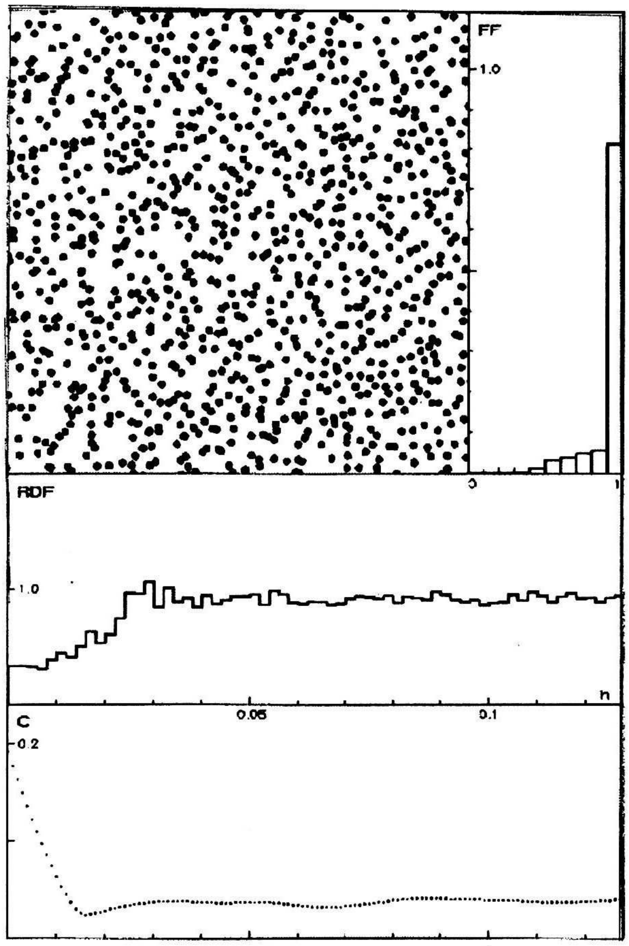

Fig. 2. Results of the model with parameters : $N = 1000$, $\Theta = 0.2$, DZ *= the same as in Fig. 1, and* $d = 10R_0$.

From the discussion of model structures, it was found that three morphological methods are the most sensitive to the film thickness and to the distribution of islands in 3D: the form factor FF, the radial distribution function RDF and the covariance C.

If the form of objects in composite film is known (as in our model), the form of projections of objects on the substrate plane described by form factor FF can be predicted (or calculated from the composite film prepared under the same experimetal conditions, but with lower thickness) and the deviations can be explained as the formation of multiple projections. From the change of FF for a given thickness compared to FF for low thickness (approximately 2D), the distribution of objects in 3D can be evaluated in probability terms.

The radial distribution function RDF has two impotant features: the onset of its nonzero part and the presence and/or magnitude of its maximum. From Figs. 1 and 2 it can be seen that the increase of film thickness influences both these features.

The covariance C is the most complex characteristic of film morphology, as it is influenced both by spatial and size distributions of objects. In 3D the information about film properties can be derived from the slope of covariance curve at the beginning (the increase of mean dimension of object projections) and from the positions and magnitudes of minima and peaks of the covariance curve (the change of the degree of randomness of objects).

5. *Conclusion*

The sensitivity of distribution of form factors FF to the distribution of objects in 3D is expected (e.g., Novák and Hrach [6]), while the possibilities of RDF and C in this aspect are fairly surprising. For the discussion of experimental micrographs of composite films, it is necessary to use data from several samples of various thicknesses, because only in this way it is possible to compensate the loss of information caused by use of 2D micrographs instead of real computer tomography, which is not acceptable in thin film physics.

Some experimental structures fulfil the first of our two assumptions only - the objects are spherical but they have not the same diameter. For this case, a more sophisticated model with variable both island radii and corresponding diffusion zones is under consideration.

Acknowledgements

The first author gratefully acknowledges the support of the Charles University Prague under Grant No. GAUK-14/94.

References

1) J. Serra, *Image Analysis and Mathematical Morphology*, Academic Press, London, 1982;

2) H. Schmeisser, Thin Solid Films **22** (1974) 83 and 99;

3) S. Blacher, F. Brouers, P. Gadenne and J. Lafait, J. Appl. Phys. **74** (1993) 207;

4) R. Hrach, D. Novotný, S. Novák, J. Pavlík and V. Hrachová, Proc. 6th Joint EPS-APS Intern. Conf. "Physics Computing '94", Lugano 1994, Switzerland, Editor R. Gruber and M. Tomassini, European Phys. Soc., p 333;

5) H. Biederman and L. Martinů, *Plasma Deposition, Treatment and Etching of Polymers*, Academic Press, New York, 1990;

6) S Novák and R. Hrach, Int. J. Electronics **73** (1992) 969.

MORFOLOŠKI OPIS KOMPOZITNIH SLOJEVA

Izravna mjerenja i modeliranje primjenjuju se za proučavanje početnih stadija rasta tankih metalnih slojeva u dvije dimenzije (nespojene nakupine na dielektričnim podlogama) i u tri dimenzije (plazma - kemijski načinjeni kompozitni slojevi). Razne se metode matematičke morfologije upotrebljavaju za analize snimaka iz transmisijskih elektronskih mikroskopa i za opis čestica raspodijeljenih u prostoru. Primjenom računala razvijene su morfološke metode koje su osjetljive za raspodjelu objekata i u trećoj dimenziji.