Image Analysis of Semicontinuous Metal Films

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In this paper the morphology of discontinuous metal films with irregular objects and semicontinuous films is studied by a combination of computer experimentation and image analysis of film micrographs. For the discussion of physical mechanisms which take part during thin film growth, an atomistic model based on direct Monte Carlo simulation technique was performed. For the analysis of experimental photographs, as well as simulated micrographs, the basic algorithms of mathematical morphology are used. Inadittion it is proposed a new set of features for thin film structure description, which are based on coefficients of Haar wavelet transform.

1. Introduction

In the initial stage of their growth, very thin metal films deposited on dielectric substrates consist of individual islands. In this case a Volmer-Weber mechanism of three-dimensional nucleation takes place. During further growth the islands increase, coalesce, create larger irregular objects and then form the semicontinuous metal layer. The basic information about film properties can be derived from the analysis of film photographs obtained from a transmission electron microscope.

In the study of thin film morphology, two kinds of problems arise. It is necessary to find convenient methods for quantitative characterisation of film micrographs, and to interpret the derived characteristics from the physical point of view. For the study of initial stages of growth the methods based on mathematical morphology [1] are commonly used. Both the size and spatial distribution of islands in very thin films can be characterised fully by standard morphological methods (e. g. [2]). However, the accuracy of these methods decreases with increasing film thickness and only a limited number of them can be used for the study of semicontinuous films.

In the present paper two tasks are solved: an attempt is made to extend the limits of some standard morphological methods to higher film thickness and to suggest new methods based on Haar wavelet transform. For both these tasks a computer experiment was suggested.

2. Models of film growth

For testing of various methods of image analysis of thin metal films, two models of three-dimensional film growth were prepared. The models are atomistic – they perform statistical operations with individual atoms. This approach is much more time consuming than the description of nucleation process by rate equations, however in addition to the time dependencies of macroscopic quantities as island concentration, the distribution functions of microscopic film properties can also be derived.

The only difference between the two models is in the coalescence process. The first model assumed the instantaneous coalescence of islands for all film thicknesses in the studied thickness range (a reasonable approximation for growth of low-melting metals as In at higher substrate temperatures), the second model included in a simplified form the formation of irregular objects and semicontinuous films.

Models are completely statistical; i.e. the migration of atoms is simulated by a Monte Carlo method without any artificial concepts as an introduction of Wigner-Seits cells [3], [4], etc. The main assumptions of the models are:

- The substrate is represented by a square lattice of 1024×1024 sites with cyclic boundary conditions (length unit).
- Atoms are deposited on the substrate randomly both in space and time with given rate of deposit.
- Single atoms migrate randomly with constant frequency (time unit).
- Two atoms occupying neighbouring sites form a nucleus.
- Migration of small clusters is possible, but its probability decreases rapidly with cluster magnitude.
- Only single atoms can be desorbed.
- Islands have a semi-spherical form.
- Coalescence:
  First model: Coalescence is liquid-like and very fast compared to other processes.
  Second model: The speed of coalescence depends on the object radii – for small objects it is instantaneous, for large objects it becomes slower. In further calculations, for the sake of simplicity the step form of this dependence is assumed.

The thickness of the modelled structures is measured in equivalent monolayers (mL).

The models were prepared in the FORTRAN-90 programming language and solved by a Pentium II/450 MHz microcomputer.

Typical data for the structure Au/dielectrics were used in the calculations – activation energy for surface diffusion 0.20 eV, energy of desorption 0.67 eV, substrate temperature 300 K and the deposition rate in limits $10^0 \div 10^6$ mL/s. The calculations were performed in thickness limits 0 ÷ 10 mL.

Duration of one run of simulation was several tens of hours; the exact value depended on the parameters of modelling. In the case of the second model the limit of coalescence process was fitted to experimental data.

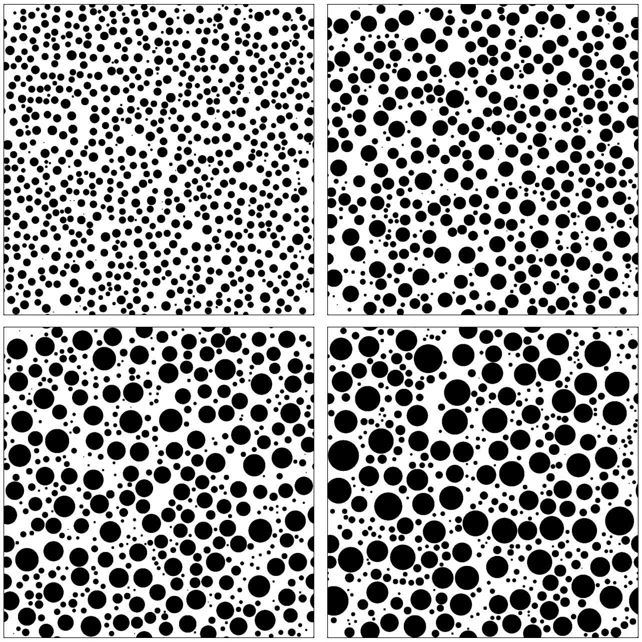

Fig. 1: Four simulated island structures with thicknesses 2.5, 5.0, 7.5 and 10.0 mL (from top left to bottom right). Model with unlimited coalescence of objects.

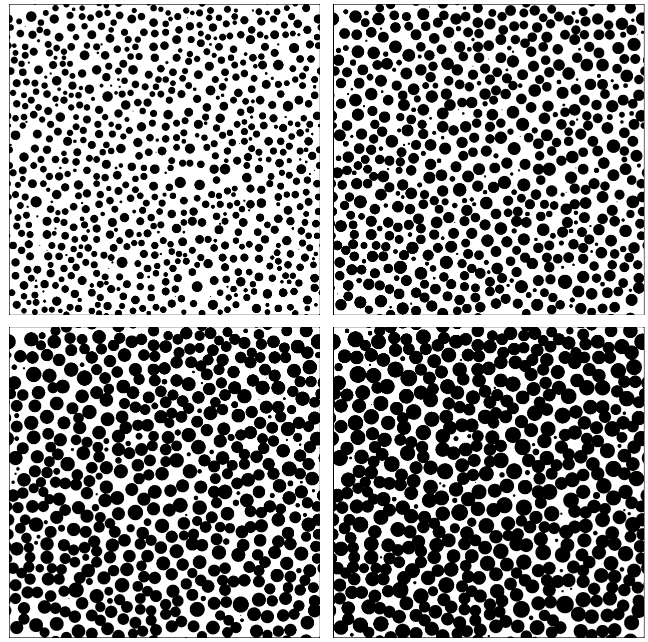

Fig. 2: Four simulated island structures with thicknesses 2.5, 5.0, 7.5 and 10.0 mL (from top left to bottom right). Model with limited coalescence of objects.

Examples of simulated structures are shown in Figs. 1 and 2. Figures represent only a limited part of working area (25 %) – typical maximum number of objects in studied structures was 4000 ÷ 5000.

3. Morphological methods

For the image analysis of discontinuous metal films the following morphological methods are typically used [2]:

a) Integral characteristics:
   Concentrations of objects; mean coverage of substrate.
b) Size distribution of individual objects:
   Distribution of object radii; form factors of objects.
c) 2D spatial distribution of objects:
   Radial distribution function; distribution of nearest-neighbours; covariance function; quadrat counts method; chord-length distribution; etc.

All these methods can be applied on data shown in Fig. 1. For the description of spatial distribution of irregular objects and for the characterisation of semicontinuous films, only methods not based on calculation of positions of object centres can be used. This criterion excludes the well-known radial distribution function, distribution of nearest neighbours and quadrat counts methods.

In Fig. 3 a distribution of object radii for model with instantaneous coalescence is shown. It can be seen that for larger film thicknesses the secondary nucleation process takes place – compare with Fig. 1.

As a powerful classical morphological method for the description of spatial distribution of objects the covariance was chosen. For discontinuous metal films the covariance function brings evidence about several morphological characteristics of the film in the same moment. By its analysis it is possible to derive values of coverage – either as *C(0)* or as *C( )* , to calculate the mean object radii – as a slope in the beginning *-dC(0)/dh* and even their distribution, and to characterise the regularity of the distribution of objects on the substrate – from the periodicity of the *C(h)* curve.

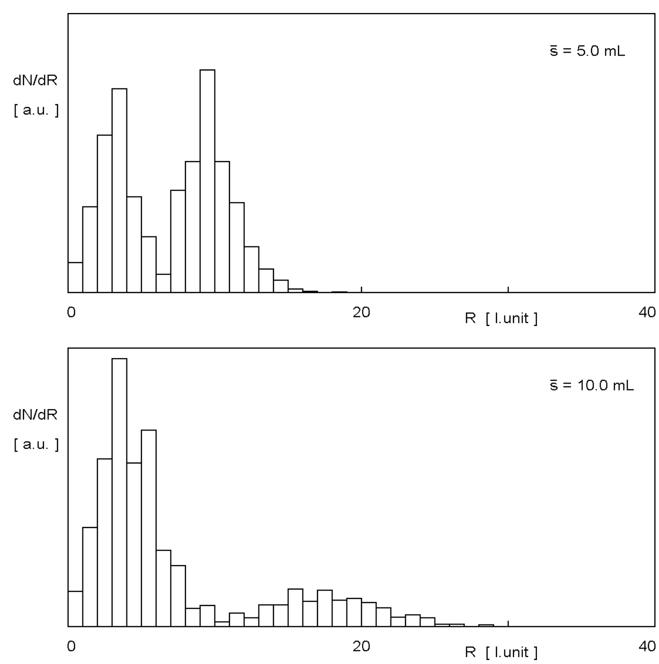

Fig. 3: Distribution of island radii for two film thicknesses. Model with unlimited coalescence of objects.

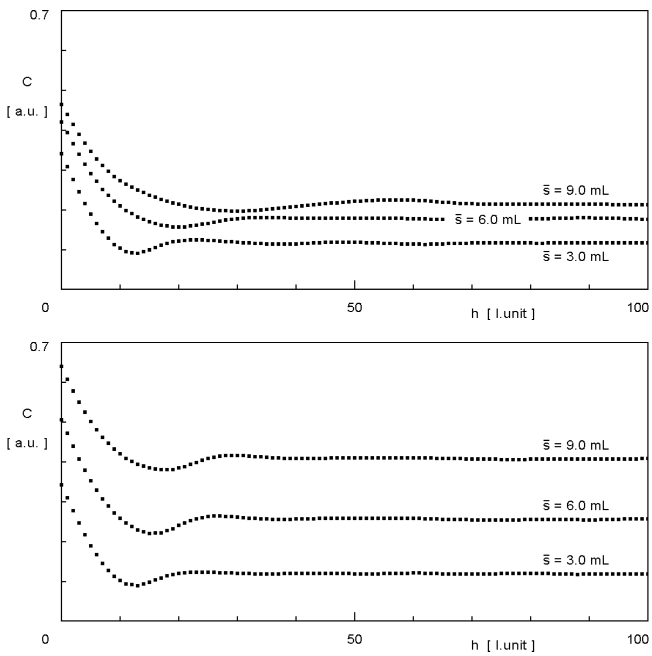

Fig. 4: The covariance function *C(h)* for various film thicknesses and two models – top: unlimited coalescence of objects, bottom: limited coalescence of objects.

In the top part of Fig. 4 this behaviour is demonstrated for discontinuous films with regular objects. Through a detailed analysis of the covariance functions for irregular objects it is discovered that the part of information is lost. In the case of continuous films the situation is even worse – only the mean coverage of the substrate can be derived from the covariance function.

From the analysis of classical methods based on the theory of mathematical morphology it follows that these methods either lose their accuracy for discontinuous films with irregular objects and for semicontinuous films or they cannot be applied at all. It is therefore necessary to search for new methods of image analysis based on other physical principles.

4. Features based on wavelet transform coefficients

In the next sections there is described a set of features able to characterise well each type of thin film structure, not only discontinuous one. The set of features is calculated from Haar wavelet transform coefficients of binarised thin film image. Therefore the Haar wavelet transform will be intuitively explained in this section. For a detailed description of wavelet transform, see for example [5, 6].

The transform of two-dimensional binary image is performed in the following way. The image is divided into blocks 2x2. Every block (which contains four pixels) is mapped onto another four 1x1 blocks. The four new obtained numbers correspond to the average and horizontal, vertical and diagonal details of original 2x2 block. The details coefficients are stored and the described step is repeated with an image from the average coefficients.

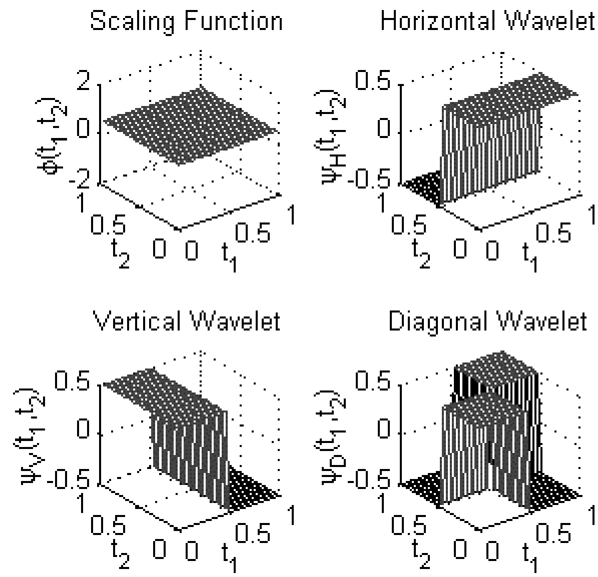

Fig. 5: The Haar scaling function and horizontal, vertical and diagonal wavelets.

The basis corresponding to the explained resolution is depicted in Fig. 5.It can be easily seen that the shapes of wavelets correspond with their adjectives, and that information hidden in wavelet transform coefficients can be thus easily interpreted.

Features which are able to describe well both the shapes of objects and their spatial distribution are powerful tools for thin film structure characterisation. We believe that a set of features proposed below fulfills these requirements. Let *MaxLev* be the maximal level of wavelet transform. Then at the ***i***-th level there are $2^{2i} + 1$ possible values of the coefficients.

These values are linearly distributed between 0 and 2^i for average coefficients and between -2^{i-1} and 2^{i-1} for detail coefficients. If the set of features would be histograms of detail coefficients at all levels then the number of bins $2NB(i)+1$, which would coincide with possible values of coefficients, would increase at the coarsest level, resulting in redundant information.

We solve this problem by choosing *KLev*-th level which determines the maximal possible number of bins. In other words *KLev* adjusts the accuracy of the obtained information (in the following calculations $KLev = 1$).

Now the new set of features characterising binary image can be presented. We propose to use the following set of numbers:

A_j^i, for $i = 1, \dots, MaxLev$, and $j = 1, \dots, 2NB(i)$;

H_j^i, V_j^i, D_j^i, for $i = 1, \dots, MaxLev$, and

$j = -NB(i), \dots, -1, 1, \dots, NB(i)$;

S;

where H_j^i is the number of horizontal detail coefficients of the ***i***-th level from the ***j***-th bin (analogically V_j^i and D_j^i for vertical and diagonal details and A_j^i for averages) and S is the total area of objects.

The set of features does not contain numbers of zero coefficients because they can be easily derived from other features. Other dependent feature is *S* but in further calculations it is easier to have it directly as one of the features.

The new features can be calculated for arbitrary binary images which contain sufficient numbers of objects or of studied phenomena to ensure satisfactory stability of the features. The proposed features can be used rather as "pre-features" to construct their forms which are more suitable to describe the structure of an image. In the next section the same progress is chosen.

5. Computer experiments

In order to test the new set of features they were calculated on the sequence of 20 binary images (0 and 1 correspond to the background and objects). The images depict the simulated island structures with different thicknesses, calculated with limited coalescence of objects, see Fig. 2 for some of them. Because the number of objects in all images is sufficient, the values of features are stable enough.

The following forms of features were calculated:

$$F_1^i = H_{-1}^i + H_1^i + V_{-1}^i + V_1^i$$
$$\text{for } i = 1, \ldots, 4;$$
$$F_2^i = (H_{-2}^i + H_{-1}^i + H_1^i + H_2^i) / (V_{-2}^i + V_{-1}^i + V_1^i + V_2^i)$$
$$\text{for } i = 1, \ldots, 4;$$
$$F_3^i = H_{-2}^i + H_2^i + V_{-2}^i + V_2^i$$
$$\text{for } i = 2, \ldots, 5.$$

The values of the feature forms are depicted in Figs. 6-8.

Features depicted in Fig. 6 correspond to the amount of pixels neighbouring with the edge of adjacent curvature. We believe that the second maximum on the curve corresponding to the first level arose because of the start of the second nucleation.

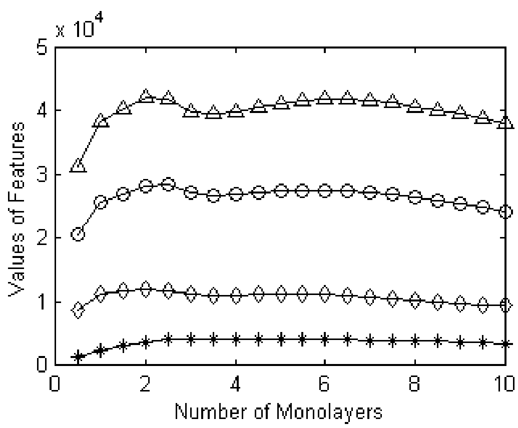

Fig. 6: The values of features F_1^1 (Δ), F_1^2( ), F_1^3 ( ) and F_1^4 (*) calculated for images of thin film structures with different thicknesses.

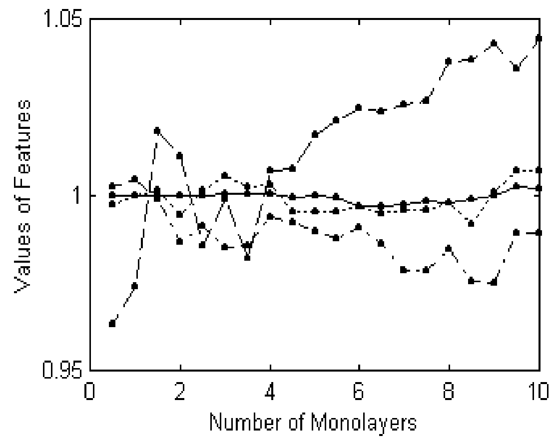

Fig. 7: The values of features F_2^1 (solid), F_2^2(dotted), F_2^3 (dashdot) and F_2^4 (dashed) calculated for images of thin film structures with different thicknesses.

In Fig. 7 the features stability can be seen. The depicted features correspond to the ratio of all horizontal and vertical details. Because in our models no direction of growth was preferred the values of the features should be close to one.

However, cases when the information hidden in the features can be easily interpreted are unfortunately rare. It is much more frequent that some thin film structure development does not influence the shape qualitatively but only quantitatively, see Fig. 8. The features depicted here are equal to the amount of the most significant horizontal and vertical details. It can be mentioned that the features at bigger levels are nonzero only for thicker film, i.e. when larger objects already exist. We also believe that the quantitative size of curves is significantly influenced by the fact that limited coalescence takes place, thus there are more lengthy resulting objects.

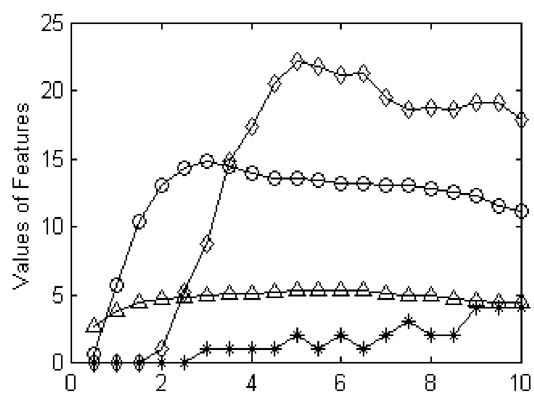

Fig. 8: The values of features F_3^2 (Δ), F_3^3( ), F_3^4 ( ) and F_3^5 (*) calculated for images of thin film structures with different thicknesses.

From the way of derivation of the new features, it can be seen that principally different binary images will have different values of features. This allows the possibility of describing not only discontinuous thin film structure, while classical morphological methods lose their accuracy. However, further interpretation of the new features should be investigated.

Acknowledgements

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