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Statistical description of systems on the basis of the Mandelbrot law: discontinuous metal films on dielectric substrates

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Abstract. The inverse power law has been used to describe the structures and the statistical distributions of discontinuous films of Au, Cu, and Mn on dielectric substrates. To this end, the rank of an island (k) was connected with its area (X) for the films with different coverage coefficients (metal contents) p. The dependencies of the island areas on the rank orders in a double-logarithmic plot are straight lines according to the Mandelbrot law.

The slope of the straight line, determined as $-1/\mu$, characterizes the distributions of the islands. One can distinguish three ranges of the critical index μ.

For $\mu > 2$, the moments m_1, m_2 are finite; if the number of islands is large, the distribution of island areas tends towards a Gaussian or log–normal one. The films structures exhibit Euclidean character.

When $1 < \mu < 2$, the moment m_1 is finite, m_2 is infinite; the islands become irregular in shape, and their areas may be described using Poisson distributions.

For $\mu < 1$, the 'hierarchization' of X is more pronounced, the moments m_1, m_2 are infinite, and the island areas are distributed according to a Lévy stable distribution (for example, Pareto's distribution).

At the percolation threshold (phase transition), the infinite cluster appears and the inverse power law is no longer valid.

1. Introduction

In this paper a brief survey of some results concerning power-law distributions and statistical descriptions of systems is given. The power-law distributions have been investigated during the past few decades in connection with fractal processes in physics and biology, and also in the field of social sciences and in analyses of the populations of cities [1–4].

Let as consider a system consisting of objects described by a random variable X; we can list the objects in the order of decreasing X-value. The object with the largest variable value is ranked first, the next smaller value is ranked second, and so on. In this way we obtain the rank order (k) of the object.

There are numerous inhomogeneous systems which may be described making use of the method presented above, examples of which include the following.

(a) The rank of a city (k) is connected with its population (X): the dependence of a city's population versus its rank on a double-logarithmic plot is roughly a straight line [1].

(b) If we count the frequency of occurrence of any word (X) in a piece of text and we list the words according to decreasing frequency of occurrence, we obtain a plot of the frequency of occurrence against the rank order. On a double-logarithmic plot, this dependence is also a straight line [3].

(c) Similarly, the turnover of Europe's 100 largest companies plotted as a function of rank on a double-logarithmic plot should give a straight line [5].

There are many other examples illustrating the inverse power law in various fields.

Mandelbrot presented the general case of the power law in the form [6]

$$X = \frac{1}{(k+\alpha)^{1+\beta}} \tag{1}$$

where α and β are constants.

When $\alpha = \beta = 0$, we obtain Zipf's law:

$$X = \frac{1}{k} \tag{2}$$

which is satisfied, for example, in the cases of city populations and frequencies of occurrence of words in a piece of text.

For other values of the constants α and β, the inverse power law takes different forms.

For $\alpha = 0$, $\beta = 1$, we obtain Lotka's law, which describes phenomena in the field of social sciences—for example, scientists producing X articles [7].

Bouchaud pointed out that there is a distinct correlation between the slope of the straight line representing the power law of Mandelbrot and the inhomogeneity of the system [5]. The slope of the straight line, determined as $-1/\mu$ (μ denotes the critical exponent), characterizes the distribution of the elements in the structure.

The main property of the variable is that all of its moments of order q are finite for $q \leqslant \mu$. Consequently, one can distinguish the following ranges for the critical exponent μ:

(i) For $\mu > 2$, the moment of first order (m_1) and the moment of second order (m_2) are finite. The ordinary moment m_1 is equal to the mean value; it is defined as

$$\bar{X} = \frac{1}{N}\sum_{i=N}^{N} X_i. \tag{3}$$

The central moment m_2 is a variance:

$$\sigma^2 = \frac{1}{N}\sum_{i=1}^{N} (X_i - \bar{X})^2. \tag{4}$$

For this range of μ, two parameters of the statistical distribution ($\bar{X}$ and σ^2) are finite, and for large N the usual central-limit theorem applies; therefore the statistical distribution of the system tends toward a Gaussian or log–normal one. The structure exhibits Euclidean character.

(ii) For $1 < \mu < 2$, inhomogeneity of the structure appears, and the moment of first order is finite. For a description of such a system, a statistical distribution with one parameter (the mean value) may be used—for example a Poisson distribution.

(iii) For $\mu < 1$, the 'hierarchization' of the structure is more pronounced, and the moments of first and second order are infinite. For large N the objects are distributed according to the Lévy stable distribution. The system may also be described making use of Pareto's statistical distribution; the structures become fractal.

For an appropriately greater slope of the straight line, a phase transition may occur. The increase of the angle between the straight line representing the inverse power law and the rank axis (in a logarithmic plot) determines the increase of the inhomogeneity of the system, and determines also the direction of evolution of the structure toward a phase transition.

In this paper, the inverse power law is used to describe the structures and the statistical distributions of discontinuous metal films on dielectric substrates. Theoretical considerations as well as experimental examinations have led to an understanding of the nucleation, initial stage, and growth of films vacuum deposited onto dielectric substrates. Under appropriate evaporation conditions (substrate temperature, evaporation rate, evaporation time), clusters consisting of different numbers of adatoms appear on the substrates. During evaporation, due to the flux of metal atoms and coalescence processes, the clusters become islands which are different as regards shape and size [8–11]. The inhomogeneity of the (areas of the) islands increases with metal content increase; that is why these structures are suitable (and interesting) for use in verification of the suitability of the above-presented ranges for the statistical description.

To this end, the rank of an island (k) is connected with its area (X) for the films with different metal contents (coverage coefficients). The ranges of the exponent μ determined correspond to different statistical descriptions of islands in discontinuous metal films. The calculated statistics is compared with that determined on the basis of microscopic examination.

2. Experimental procedure and results

The methods used to obtain the discontinuous Cu and Mn films on quartz-glass substrates were described in papers by Dobierzewska-Mozrzymas *et al* [12, 13]. The microstructures of the films were examined with an electron microscope, using a carbon replica for the Cu films. Mn and thinner Cu films for structural examinations were evaporated onto copper grids covered with Formvar and an amorphous carbon layer. To determine the structural parameters of the films, such as the coverage coefficient (the coverage coefficient is defined as the ratio of the metal area to the total area of the film) and island areas, we used the following image analysis of the electron micrographs. In the first step, the micrographs were digitized using the HP Jet Scan II+ camera at 300 DPI resolution. Making use of the method described in [14], a non-ambiguous digitization level was determined. Next, a binarization process enabled us to separate the conductor (island) pixels from the insulator (substrate) ones. Using a special computer program the islands were ranged in order of decreasing island area. The results for the metal films with different coverage coefficients are presented in figure 1. The dependencies of the island area (X) on its rank order (k) in a double-logarithmic plot, representing the power law of Mandelbrot (1), are straight lines. Their slopes depend on the inhomogeneity of the structure, which is connected with the metal content. For coverage coefficients $p \leqslant 0.3$ (figure 2(a)), the islands are regular in shape; their areas differ insignificantly. The slopes of these straight lines correspond to values of $\mu > 2$ (case (i)). The parameters of the distribution, such as the mean value and the variance, are finite. These structures may be described by a statistical distribution using two parameters—for example, a Gaussian or log–normal distribution. Figure 2(b) shows an experimental and a calculated log–normal distribution.

On the basis of structural examination, the parameters of the distribution, such as the mean value $\bar{X}$ and the variance σ^2, were determined. For these parameters, the log–normal distribution was calculated. The experimental distribution obtained from the histograms of island areas is also presented, in figure 2(b). It is seen that for more homogeneous structure (figure 2(a)), when the islands are more regular in shape, their areas do not differ much; the agreement between the theoretical and experimental description is good. The statistics with

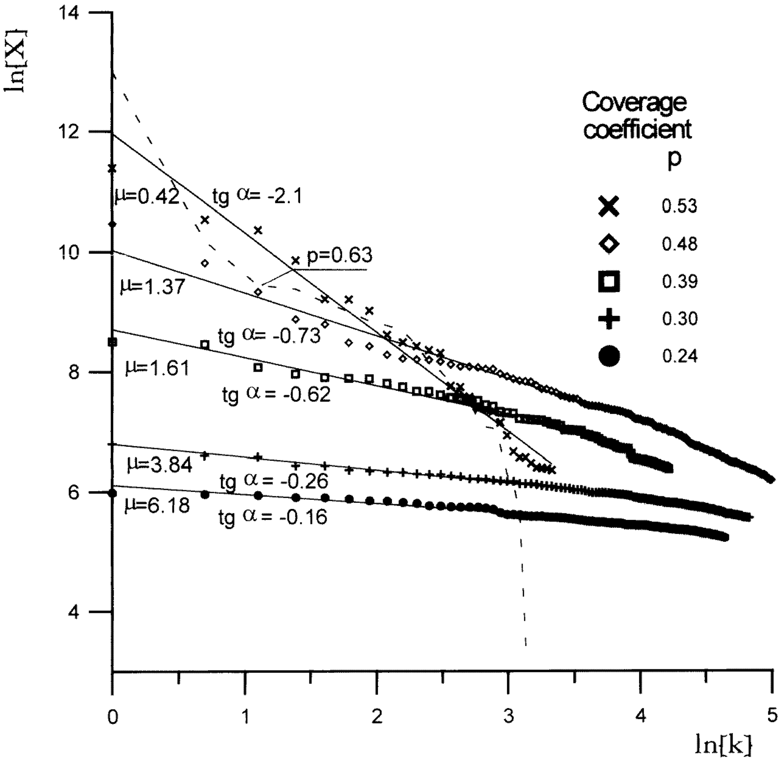


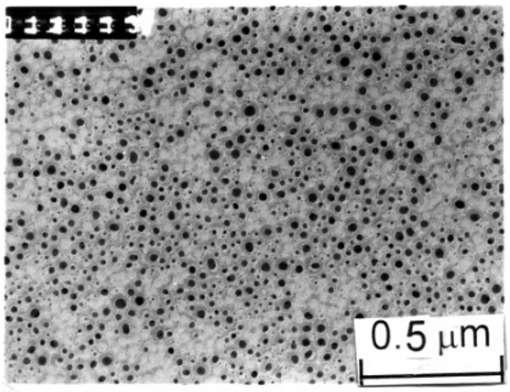
Figure 1. Island area X versus rank order k for films with different coverage coefficients in a double-logarithmic plot; the straight lines were fitted using the least-squares regression method.

two parameters may describe such structures.

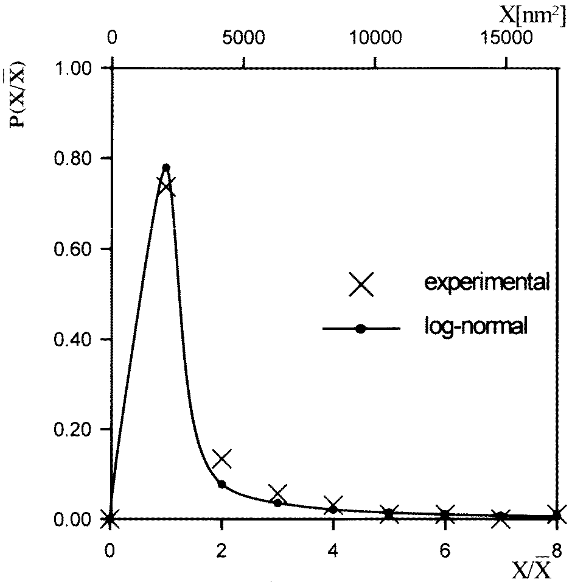
With increasing metal content, the islands become more irregular; they change shape from spherical to ellipsoidal and then to long paths along the film, as is seen in figures 3(a) and 4(a). Also, the areas of the islands are different. The experimental histograms presented in figures 3(b) and 4(b) show that, in addition to the islands with small areas, there are also islands with very large areas. When $1 < \mu < 2$, the Poisson distribution describes the metal film structure. Experimental and theoretical results are compared in figure 3(b). The probability of the appearance of large islands is small, but they do occur in the discontinuous films. In this case, the angle between the straight line representing the Mandelbrot law and the rank axis (in the logarithmic plot) increases, and the critical index is in the range $1 < \mu < 2$ or $\mu < 1$ (cases (ii) and (iii)). It is interesting that for $\mu \approx 1$ the entire sum is, in a first approximation, given by its largest term:

$$X_1 \Big/ \sum_{i=2}^{N} X_i \approx 1.$$

In such a case, the mean value $\bar{X}$ in fact loses its meaning. This is a key feature of the Lévy distribution. These structures exhibit self-similar properties; they become fractal. An experimental histogram of such a structure is presented in figure 4(b). The islands with large areas occur in the discontinuous Mn film with the coverage coefficient $p = 0.62$. To describe



(a)



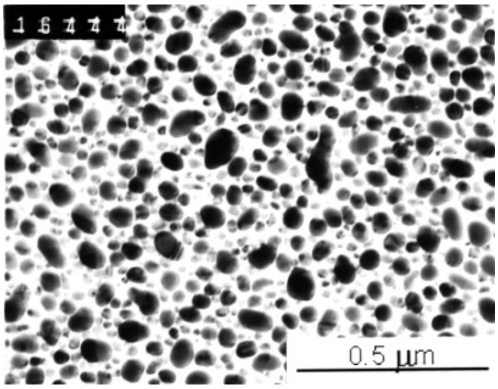
(b)

Figure 2. (a) The microstructure of a Cu film with coverage coefficient $p = 0.30$. (b) The log–normal distribution of island areas for this film, for a log–normal distribution with standard deviation $\sigma_\Lambda = 1.8$.

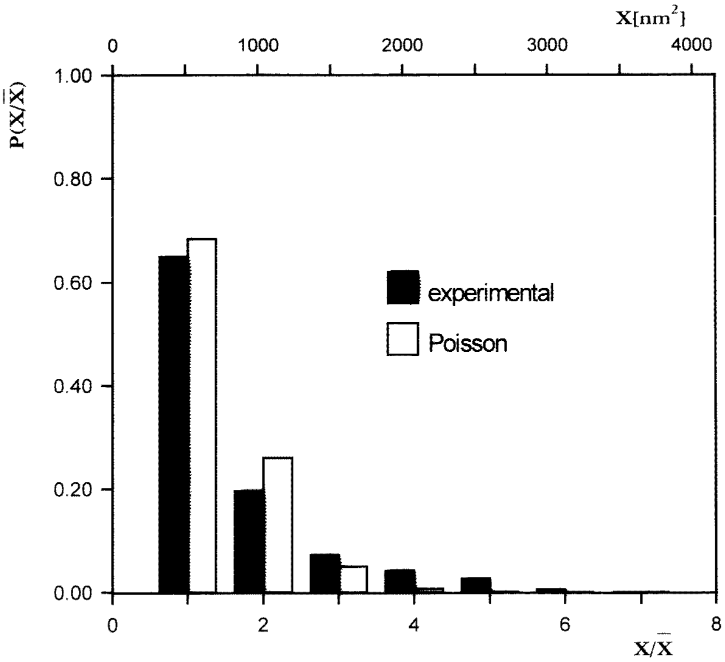
this structure, Pareto's distribution was used. Comparison of the experimental results with calculated ones (figure 4(b)) allows us to confirm that there is satisfactory agreement.

The values of the structural parameters for the different metal films are presented in table 1. With increasing metal content, inhomogeneity of the structure appears, and the critical index decreases. The contribution of the first term (the largest island) to the sum significantly increases when the structure becomes more irregular. For lower metal content, the mean value $\bar{X}$ increases with increasing coverage coefficient.

In the phase transition (at the percolation threshold), the infinite cluster appears, and the



(a)



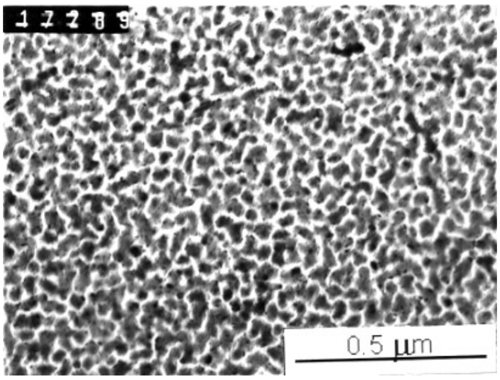
(b)

Figure 3. (a) The microstructure of a Cu film with coverage coefficient $p = 0.39$. (b) The Poisson distribution of island areas for this film.

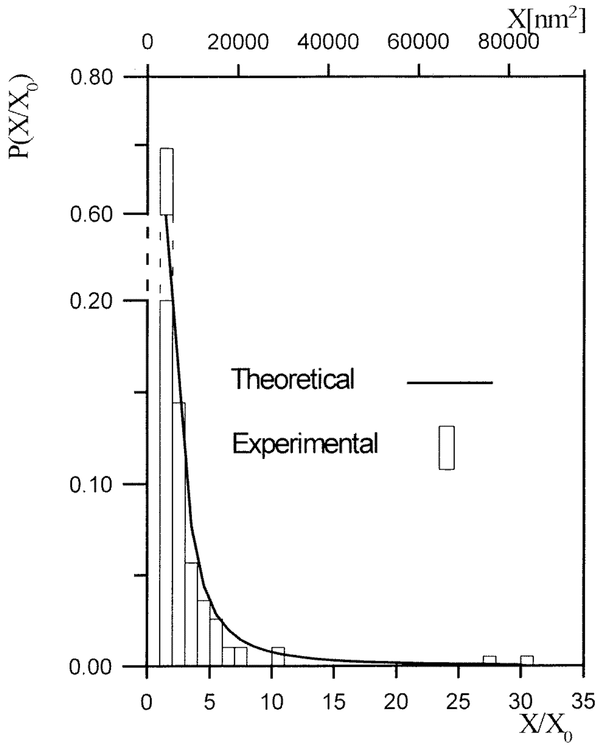
inverse law loses its validity. For the discontinuous Cu film, $p_c = 0.63$ (figure 1); for this value, the optical and electrical properties of Cu films change from dielectric to metallic ones.

3. Conclusions

The structures of discontinuous metal films deposited onto dielectric substrates depend distinctly on the metal content. With increasing coverage coefficient, the metal islands become



(a)



(b)

Figure 4. (a) The microstructure of a Mn film with coverage coefficient $p = 0.62$. (b) The experimental distribution of island areas and Pareto's distribution of island areas for this film. Pareto's distribution is defined as follows: $\rho(X) = (\mu/X_0)(X_0/X)^{\mu+1}$ for $X > X_0$ where X_0 denotes a typical scale, and the smallest variable is of order X_0 [5].

more and more irregular in shape; their areas are different. That is why these systems are suitable for application of the different statistics determined on the basis of the Mandelbrot law.

Table 1. Structural parameters for discontinuous Au, Cu, and Mn films.

No	Metal	Coverage coefficient p	μ	$X_1 / \sum_{i=2}^{N} X_i$
1	Au [15]	0.24	6.18	0.0085
2	Cu	0.30	3.84	0.044
3	Cu	0.39	1.61	0.046
4	Cu	0.48	1.37	0.23
5	Mn	0.62	1.01	1.17
6	Mn	0.64	0.44	11.35

The slope of the straight line representing the inverse power law is connected with the inhomogeneity of the system. According to Bouchaud, there are three ranges of the slope determined by the critical index μ.

For $\mu > 2$, the structures are homogeneous; the system may be described by the statistics of two parameters: the mean value and the variance.

In the middle range, $1 < \mu < 2$, inhomogeneity of the structure appears, and the statistics of one parameter (the mean value) describes these systems.

When the 'hierarchization' is more distinct ($\mu < 1$), the contribution of the first object to the total sum is significant, and the objects are described according to the Lévy stable distribution.

In this paper the ranges of the statistical descriptions presented above were verified experimentally for discontinuous metal films on dielectric substrates.

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