Z. Phys. D – Atoms, Molecules and Clusters 20, 317–319 (1991)

Atoms, Molecules
Zeitschrift für Physik D and Clusters

Multifractal analysis of gas evaporated metal particle aggregates

J. Håkansson* and G.A. Niklasson

Department of Theoretical Physics and Physics Department, Chalmers University of Technology, S-41296 Göteborg, Sweden

Received 10 September 1990

Abstract. We have produced aggregates of small cobalt particles by evaporation in a few hundred Pa of inert gas. The structure of the aggregates was studied by transmission electron microscopy. We have used a box counting algorithm in order to determine the generalized dimensions and the multifractal $f(\alpha)$ spectrum of the aggregates. Box sizes were varied within wide limits in order to study the convergence of the spectra to optimum values. We have found that the structure of gas evaporated aggregates cannot be described by a single fractal dimension. A multifractal description is necessary with generalized dimensions spanning a range of 1.60 to 1.82.

PACS: 02.40; 61.16D; 61.90

1. Introduction

The concept of fractal geometry [1] has been found to be useful for the characterisation of a large class of disordered structures. Recently, however, it has been realized that in many cases a more detailed structural characterisation can be obtained by the formalism of multifractals [2, 3]. In this case the structure is described by a continuum of generalized dimensions, the so called Renyi dimensions [4], D_q, or alternatively by a spectrum of scaling indices, α, and their densities $f(\alpha)$ [5]. The characterisation of deterministic geometrical multifractals has been studied theoretically in several recent papers [6–10], but applications to experimental structures have so far been relatively rare [10, 11].

In this paper we present preliminary results from a study of the multifractal structure of aggregates of metal particles produced by evaporation in an inert gas. These aggregates cannot be described by a single fractal dimension, instead a continuous spectrum of dimensions are necessary. This is in contrast to earlier studies of electrodeposited clusters [10, 11], which could be described by a single dimensionality exponent.

2. Experiment and calculations

Ultrafine Co particles were produced by inert-gas evaporation [12] from a heated tungsten spiral in a conventional bell-jar system. Evaporation took place in 1.3 kPa of argon gas and resulted in particles of about 100 Å radius that were clustered into large connected aggregates as seen in the inset of Fig. 2. The deposits were collected onto carbon covered copper grids for electron microscopy. The micrographs were subsequently digitized by an image analyser. It has been shown previously that gas evaporated aggregates display a fractal structure over a range of length scales [13, 14], with dimensionality dependent on the magnetic character of the particles [15].

The generalized dimensions and the $f(\alpha)$ spectra of the aggregates were obtained by the box counting method. We divided the micrographs into boxes, all with the same side length L, and determined the projected area of the particles in each box. Let M be the total projected particle area, N the number of non-empty boxes and N_i the area measure in the ith box. Then the generalized dimensions can be directly obtained from [8]

$$D_q = \left(\ln \sum_{i=1}^{N} (N_i/M)^q \right) \Big/ ((q-1)\ln L) = Z_q/\ln L. \qquad (1)$$

Here D_0 is the usual fractal dimension, while D_2 for example denotes the correlation dimension. The spectrum of scaling indices is related to the generalized dimensions by the Legendre transformation [5]

$$f(\alpha(q)) = q\alpha(q) - (q-1)D_q, \qquad (2)$$

where

$$\alpha(q) = (d/dq)((q-1)D_q). \qquad (3)$$

* *Present address*: ABB Corporate Research, S-72178 Västerås, Sweden

We have systematically varied the box size in order to study the convergence of the generalized dimensions and the $f(\alpha)$ spectrum. The convergence has been studied in detail before on deterministic fractals [7–9] and certain general features have emerged. The best approximation to D_q for large positive q is obtained with box sizes on the order of the largest particle in an aggregate. For negative q the convergence is very slow and one should use boxes that are smaller than the larger of the particles. Indeed, the value of $D_{-\infty}$ can be obtained exactly from calculations of D_q with a box size much smaller than the smallest particle in the sample [8].

3. Results and discussion

In Fig. 1 we display $f(\alpha)$ spectra using various box sizes for an aggregate of cobalt particles that is shown in the inset of Fig. 2. It is seen that we have a rather good convergence as the box size is varied for points to the left of the maximum of the curve, i.e. for positive q. For points to the right of the maximum corresponding to negative q convergence is slow and the $f(\alpha)$ spectrum cannot be accurately determined. The best approximation to $f(\alpha)$ for a given α is given by the highest value for positive q and the lowest value for negative q [9]. This assertion is based on studies of a deterministic fractal aggregate, where the box counting result could be compared to the exact analytical solution [9].

We consider the case of positive q in more detail in Fig. 2, where we display the generalized dimensions D_q obtained from calculations with various box sizes. It is

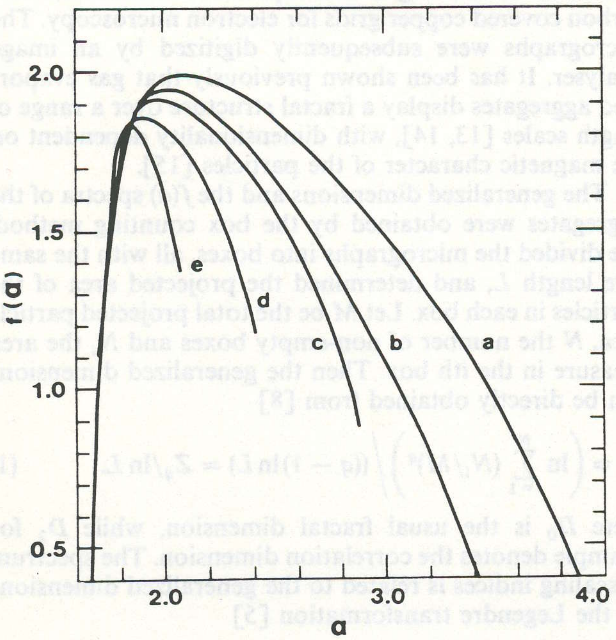

Fig. 1. Spectrum of scaling indices, $f(\alpha)$, for a cobalt aggregate produced by evaporation in 1.3 kPa of argon. The aggregate is shown in the inset of Fig. 2. The curves are given for box side lengths of (a) 1/9, (b) 1/18, (c) 1/36, (d) 1/72 and (e) 1/144 of the micrograph width

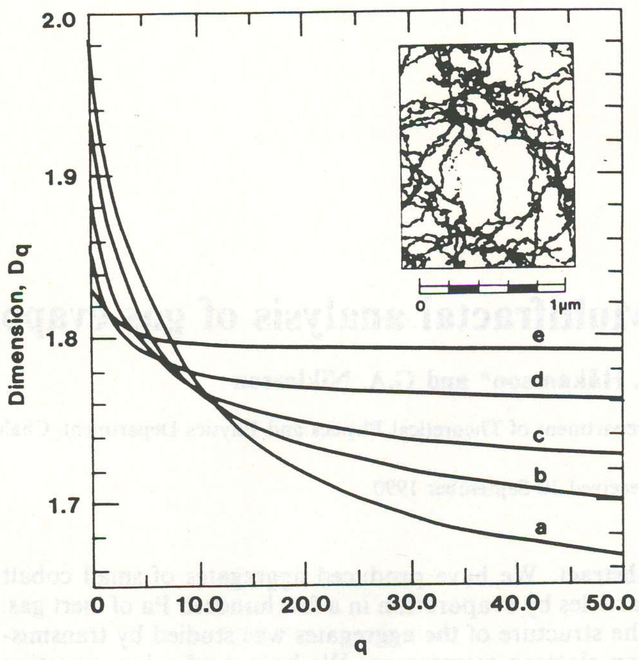

Fig. 2. Generalized dimensions, D_q, for the cobalt aggregate shown in the inset. The box side lengths were (a) 1/9, (b) 1/18, (c) 1/36, (d) 1/72 and (e) 1/144 of the micrograph width

realized from (2) and the remarks in the previous paragraph that the smallest D_q for a given q is the best approximation to the actual value. The box size that gives the best result varies with the parameter q. We expect that the real values of D_q are somewhat lower than in the curves in Fig. 2. This is indicated by the value of $D_{-\infty} = 1.817 \pm 0.004$ and by the previously determined pair correlation function [15], which gives $D_2 \approx 1.70$. In addition, plots of Z_q as a function of ln L (see (1)) [10, 11] yield lower generalized dimensions than in Fig. 2. For example we obtain $D_0 = 1.703 \pm 0.006$ and $D_\infty = 1.598 \pm 0.006$. It seems that in Fig. 2 the convergence towards the real values of D_q is not yet complete. We infer that the spectrum of generalized dimensions for the gas evaporated cobalt particles is fairly narrow and lies between about 1.60 and 1.82. The value of D_2 falls between the values previously found for aggregates consisting of paramagnetic and ferromagnetic aggregates, respectively [15]. This is because the present sample consists of an agglomerate of ferromagnetic chainlike aggregates, which should have a higher dimensionality than a single magnetic aggregate.

We now discuss the possibility that the multifractal structure of the cobalt aggregates may be due to the well-known [12] log-normal size distribution of the individual particles. Mandelbrot [16] has shown that log-normal distributions are examples of multifractal measures. We have generalized the derivations of Mandelbrot [16] to the case of a log-normal measure on a fractal structure. We obtain the result

$$D_q = D_0 - \nu q, \quad (4)$$

and

$$f(\alpha) = D_0 - ((\alpha - \nu - D_0)^2/4\nu), \quad (5)$$

where ν is a parameter characterising the width of the log-normal distribution. The optimal $f(\alpha)$ spectrum in Fig. 1 is qualitatively similar to the parabolic shape implied by (5). However, the generalized dimensions depicted in Fig. 2 do not seem to depend linearly on q as predicted by (4). Better convergence of the experimental data is needed in order to carry out a strict comparison.

In conclusion we have determined the generalised dimensions D_q and the $f(\alpha)$ spectrum for an aggregate of gas evaporated cobalt particles. To our knowledge, this was the first time that geometrical multifractality has been demonstrated in an experimental specimen. A more detailed study on the convergence of the D_q's as the box size is varied is in progress and may lead to improved values of the generalized dimensions.

This work was partially supported by grants from the Swedish Natural Science Research Council and the National Swedish Board for Technical Development. We are very grateful to A. Torebring and C. Larsson for valuable experimental assistance.

References

1. Mandelbrot, B.B.: The fractal geometry of nature. New York, San Francisco: W.H. Freeman 1983
2. Mandelbrot, B.B.: Pure Appl. Geophys. **131,** 5 (1989)
3. Tél, T.: Z. Naturforsch. **43a,** 1154 (1988)
4. Renyi, A.: Probability theory. Amsterdam: North Holland 1970
5. Halsey, T.C., Jensen, M.H., Kadanoff, L.P., Procaccia, I., Shraiman, B.I.: Phys. Rev. A **33,** 1141 (1986)
6. Tél, T. and Vicsek, T.: J. Phys. A **20,** L835 (1987)
7. Tél, T., Fülöp, A. and Vicsek, T.: Physica A **159,** 155 (1989)
8. Håkansson, J. and Russberg, G.: Phys. Rev. A **41,** 1855 (1990)
9. Håkansson, J.: (to be published)
10. Argoul, F., Arneodo, A., Grasseau, G., Swinney, H.L.: Phys. Rev. Lett. **61,** 2558 (1988)
11. Sagués, F., Mas, F., Vilarrasa, M., Costa, J.M.: J. Electroanal. Chem. **278,** 351 (1990)
12. Granqvist, C.G., Buhrman, R.A.: J. Appl. Phys. **47,** 2200 (1976)
13. Forrest, S.R., Witten, Jr., T.A.: J. Phys. A **12,** L109 (1979)
14. Farestam, T., Niklasson, G.A.: J. Phys. Cond. Matter **1,** 2451 (1989)
15. Niklasson, G.A., Torebring, A., Larsson, C., Granqvist, C.G., Farestam, T.: Phys. Rev. Lett. **60,** 1735 (1988)
16. Mandelbrot, B.B.: In: Frontiers of physics: Landau memorial conference. Gotsman, E. (ed.). New York: Pergamon 1989