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K. Mecke D. Stoyan (Eds.)

Morphology of Condensed Matter

Physics and Geometry of Spatially Complex Systems



Springer

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Cover Picture: 3D Image of a block copolymer (SBS) microdomain structure reconstructed from a series of TappingMode scanning force microscopy phase and height images. The enclosed volume can be interpreted as PS cylinders within a $200 \times 160 \times 45 \text{ nm}^3$ large portion of the SBS film. For size comparison and illustration of the molecular structure of the material, a SBS molecule bridging two PS domains is sketched (Reprinted from: R.Magerle, 'Nanotomography', *Physical Review Letters* **85**, 2749 (2000); ©2000 by The American Physical Society)

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Preface

What does a typical foam cell look like? How do we measure spatially structured phases of composite materials? What patterns can be observed in thin films, emulsions, or polymer blends? Is it possible to describe and compare structures of complex, often disordered, materials quantitatively and relate the observed structure to macroscopic physical properties? Does the liquid flow through porous rocks, for instance, depend on the shape of the pores? These are only a few questions which are of enormous interest to both the scientific and the industrial communities.

The rapid growth of high resolution three-dimensional imaging technology is likely to impact on numerous experimental areas in physics, chemistry, geology and biology as well as on technical disciplines, including materials science, chemical engineering, or diagnostic medicine. Nowadays, experimental techniques such as high-resolution X-ray computed tomography or confocal microscopy are available to measure the morphology of complex materials and visualise, for instance, multiple fluid phases in porous materials. Such experimental facilities will provide researchers in the near future with a rich source of experimental data to further the goals of their research. The development of facilities which can acquire images, perform geometric analysis, visualise and calculate physical properties will have a major impact on the goal to catalogue material structure and infer physical properties from structural characteristics. Therefore, the development of expertise in computational image analysis, spatial statistics and materials modelling using typical three-dimensional data sets will become an essential requirement in these fields. Morphological and spatial statistics techniques are therefore certain to be more generally useful for a range of researchers.

The objective behind this volume is to bring the tools of physics, spatial statistics and mathematics to bear on these problems, which concern not only workers in those fields but also, for instance, engineers, geologists and forestry researchers. While the previous volume ‘Statistical Physics and Spatial Statistics – The Art of Analysing and Modelling Spatial Structures and Pattern Formation’, edited by Klaus Mecke and Dietrich Stoyan (Lecture Notes in Physics, Vol. 554, Springer, July 2000) focuses on the statistical characterisation of random spatial configurations, in particular point processes and Gibbsian particle models, we try to emphasise here applications of morphology on real physical systems ranging from porous and composite materials to microemulsions and foams. Additional techniques of spatial statistics and integral geometry, such as shape analysis and mark correlations, as well as measures such as contact distributions, topological Betti numbers and tensor valuations are also considered.

In an attempt to guide the reader and categorise the diverse contributions we have divided the articles into two parts: the first focusing on physical systems such as spatially structured solids and complex fluids. The second part comprises contributions with a background in spatial statistics and morphology. Of course, a strict distinction is not possible since each article provides some insight in each category but nevertheless the book tries to group together main features, such as solids, fluids, shape measures and spatial distributions.

Part I. Complex Structured Condensed Matter

Porous media, foams, microemulsions and liquid crystals are examples of spatially structured materials. Since they are nowadays experimentally accessible, these systems are particularly interesting for spatial statisticians and mathematicians in order to apply their techniques to characterise and model random spatial configurations.

Porous materials such as sandstones or papers are considered as standard examples in nature for a complex spatial structure. Worldwide, the petroleum industry spends an excess of a billion dollars annually on the characterisation of reservoirs and on laboratory measurements of core materials. These measurements are critical in the development of oil and gas fields because they play a major role in predicting hydrocarbon recovery. Despite the large sums spent on measurements and the ability to image and visualise, there is little basic science to support the interpretation of data and to improve the quantitative morphological description and modelling of laboratory core measurements. Modest improvements in relating physical properties to geometric shapes of pores would significantly reduce the economic risk associated with new oil and gas developments and have a major impact on the petroleum industry. Such a structure–property relationship is also essential in crustal geophysics. Within the Earth’s crust, fluid flow in fractured rock masses influences a range of important geological processes, particularly the genesis of many types of precious metal and base metal ore deposits and hydrocarbon migration in some reservoir types. A rigorous understanding of fracture-controlled fluid migration based on statistical measures of structure and distribution of fractures is also required to develop more effective strategies for the sustainable development of geothermal energy resources and for the analysis of risk associated with toxic waste - both nuclear and chemical - containment in deep underground repositories. Another important application of morphological techniques to describe the shape of solids is in paper manufacture: the development of improved printing mediums and technologies depends on an understanding of how a complex material made up of cellulose fibre matrix, often coated with a consolidated mass of pigment and binder, can be modified to improve printability, durability and appearance. To understand the flow, optical and mechanical properties of paper products, one must develop a realistic structural description of the pore space coupled with an ability to simulate flow and mechanical properties.

A major shortcoming in the understanding of processes involving complex porous and composite materials has been the inability to accurately characterise the statistical distribution of shape and structure of disordered microstructures. Successful predictive modelling of the properties of ‘real world’ materials is reliant on accurate statistical characterisation, such as for the generation of equivalent network structures required

for multiphase flow studies or fracture prediction. Dominique Jeulin shows in ‘Spatial Statistics and Micromechanics of Materials’ how useful bounds of elastic properties can be determined from the three-point statistics of random media and how fracture statistics models based on random functions allow us to predict the probability of fracture of materials.

The availability of direct measurements of the three-dimensional microstructure of sedimentary rocks, for example, requires the development of computational tools to gauge the stochastic nature of the void space and to construct realistic model representations of the complex space. We need to generate stochastic model networks with topological properties representative of real sedimentary rocks and measure and quantify pore geometry and correlations that occur at the pore scale. This task is approached in the contribution ‘Characterising the Morphology of Disordered Materials’ by Christoph Arns, Mark Knackstedt and Klaus Mecke, where advanced methods of image analysis and integral geometry (see Part II) are applied to reconstruct and predict physical properties of porous media based on the measurement of pore space. A major aim is to understand the structure–property correlation since the relationship between microstructure and macroscopic properties is a central issue in materials science. To date, engineers have relied on simple empirical models that ignore actual microstructural information, to correlate macroscopic properties of disordered materials to their microstructure. The bulk of experimental and theoretical work has been devoted to establishing the empirical coefficients for each class of material. Imaging materials via high-resolution X-ray CT, for instance, and subsequent laboratory measurement of material properties may help us form a more accurate and comprehensive picture of the role of the shape of disordered materials in governing the mechanical and transport properties. The development of theoretical models describing the relationships between disordered media and fluid transport properties is of specific interest to geologists, both in the laboratory and the field. Hans-Jörg Vogel demonstrates in ‘Topological Characterisation of Porous Media’ the significance of the Euler characteristic for the prediction of transport in natural soil where it is used as a function of pore diameter to generate network models of the porous structure. Other applications of the Euler characteristic are studied in Part II of this volume, for instance, in the contributions by Vanessa Robins on Betti numbers, Joachim Ohser et al. on discretized sets, and Claus Beisbart et al. on vector and tensor-valued descriptors, as well as in Part II of the previous Lecture Notes in Physics, Vol. 554.

Most modern materials have complex three-dimensional structures on the micron and nanometer scale. Unfortunately, most microscopy techniques available today are limited to two-dimensional imaging and the choices for obtaining nanometer-resolution three-dimensional volume images are rather limited, at least in materials science. This considerably hinders a detailed study of the structure–property relationship and renders a direct comparison between simulation results, the materials microstructure and its physical properties difficult, if not impossible. In particular, quantitative experimental data is needed as input for reliable computer simulations and for benchmarking of models. To this end, Robert Magerle presents nanotomography as a general procedure for high-resolution three-dimensional volume imaging of composite materials based on scanning probe microscopy. The specimen under study is eroded gradually and the remaining material is imaged with scanning probe microscopy at each freshly exposed surface.

From the resulting series of images, the specimen's three-dimensional microstructure can be reconstructed with methods adopted from computed tomography. This approach is expected to be a simple and versatile means for the imaging of various materials and physical properties with micron, nanometer and even atomic resolution. This effort is of immediate benefit to modelling scientists, spatial statisticians and physicists to interpret and compare model assumptions to reliable data of real-space volume images in three dimensions.

In contrast to solid structures, soft condensed matter, such as foams and fluids, have a typical energy scale which is comparable with thermal energies at room temperature. Thus, thermally activated movement of its molecular components are sufficient to induce configurational changes, which may eventually lead to complex spatial structure at room temperatures. Such thermally induced patterns are of particular importance for biological systems, such as living organisms, which are based on self-organisation of molecules in membranes and cells. However, the understanding of the underlying mechanisms is also essential for the application of many technologies in everyday life, including colloidal dispersions (paints, inks, food, creams, lotions), foams (beverages, see the contribution by Francois Graner), liquid crystals (displays, see the contribution by Friederike Schmid and Nguyen Phuong), polyelectrolyte gels used in diapers, as well as soaps for washing and cleaning. Most biomaterials, for example blood or cartilage, belong to the material class of soft condensed matter. One particularly important biomaterial is the biomembrane, that is, the protein carrying lipid bilayer which surrounds each cell and its organelles and which form spontaneously in mixtures of water and lipids. A good model system for the structural properties of biomembranes are self-assembling amphiphilic systems studied by Ulrich Schwarz and Gerhard Gompper in this volume, but also Langmuir monolayers studied by Mathias Lösche und Peter Krüger.

Soft condensed matter has received much attention in the past mainly because of its spatial structure and relevance for biological studies. Bicontinuous phases in microemulsions (see U. Schwarz and G. Gompper), complex domain patterns in fluid layers on substrates (see M. Lösche und P. Krüger), orientational order in liquid crystals (see F. Schmid and N. H. Phuong), or cellular shapes in foams (see F. Graner) are just a few spatial features observable in soft condensed matter. As for solid structures, the shape and connectivity of the constituents are essential for physical properties and biological function, so that techniques from spatial statistics and morphology become useful.

Ulrich Schwarz and Gerhard Gompper demonstrate in 'Bicontinuous Surfaces in Selfassembling Amphiphilic Systems' the richness of emerging structures in the case of self-organising amphiphiles. These are molecules with both hydrophilic and hydrophobic parts and which are particularly relevant for biological studies. In water, they self-assemble into extended sheet-like structures due to the hydrophobic effect. Many different interfacial structures emerge such as random surfaces, bicontinuous labyrinths, triply periodic minimal surfaces, their constant mean curvature and parallel surface companions. The free energy of an amphiphilic system can be written as a functional of its geometry and phase diagrams can be calculated by comparing the free energies following from different geometries. Due to the richness of its spatial structures, self-organising amphiphiles may serve as model systems for the study of the relationship of shape and energy in soft condensed matter.

Related systems are Langmuir monolayers which are commonly used as model systems for biological membranes. Biomembranes are complex, quasi-two-dimensional lipid-protein structures that self-assemble in aqueous media. For a quantitative understanding of the physics that underlies the functioning of biological cells, an assessment of the structural aspects of the molecular interactions within the membrane is necessary. These interactions may lead to complex two-dimensional domain patterns which are studied and characterised by Mathias Lösche and Peter Krüger in ‘Morphology of Langmuir Monolayer Phases’ using geometric tools such as Minkowski functionals (see Part II in this volume as well as in the previous Lecture Notes in Physics, Vol. 554).

Oriental order in liquid crystals is another important example of complex structures in condensed matter. Randomly distributed hard spheres in three dimensions form two types of structures, depending on their density: a disordered fluid phase and an ordered crystalline phase. For randomly distributed anisotropic particles, the situation is different: several so-called ‘mesophases’ exist at intermediate densities between the fluid and the solid state. For instance, the particles may be oriented in one common preferred direction, but do not have crystalline translational order (nematic phase). Alternatively, the particles may be arranged in layers of two dimensional fluids (smectic phases). Since these mesophases are neither crystalline nor truly liquid, they are commonly referred to as ‘liquid crystal phases’. Friederike Schmid and Nguyen Phuong present computer simulations of simple model systems with special emphasis on systems of hard ellipsoids. The occurring orientational order of the molecules can be described by mark correlation functions which are introduced and studied in detail in Part II.

Whereas in liquid crystals geometric order is induced by molecular interactions, in foams the reverse effect occurs: geometrical constraints determine the energy landscape of the configurations. Liquid foams consist of gas bubbles separated by a continuous film of liquid which exhibit a complex spatial structure. The morphology of the vapour cells is a fascinating topic as shown by Graner in ‘Two-Dimensional Fluid Foams at Equilibrium’. Fluid foams try to minimise their surface energy by decreasing the area of the bubbles. This mathematical ‘minimum perimeter’ problem has previously been approached by Gompper and Schwarz dealing with amphiphilic systems, but the minimal surfaces in foams are subjected to complex topological constraints which are very common in nature. Such foam-like structures occur also as grain boundaries in crystals, as magnetic domains in solids, as honeycombs, as biological epithelia, or as retina patterns.

In all these examples of structured condensed matter, the use of techniques stemming from spatial statistics and morphology are indispensable. The methods are described in detail in Part II, although the tools of mathematical morphology will have already been introduced and applied in the previous sections: for instance, mark correlations by F. Schmid and N. H. Phuong, Minkowski functionals and parallel surfaces by C. Arns et al., by U. Schwarz and G. Gompper, and by M. Lösche and P. Krüger, or the topology of minimal surfaces and perimeters by F. Graner. These techniques and other methods of spatial statistics to characterise the morphology of structures are considered in detail in the second part of this volume.

Part II. Spatial Statistics and Morphology

The imaging of complex topologies and geometries, typical of all real world materials, is now feasible, due to advances in three-dimensional imaging technology and computer processing. These advances are of importance to a variety of scientists, both academic and industrial, leading to an increasing interest not only to capture images but also to model and characterise them, that is, to quantify disordered morphologies. The ability to generate non-destructively three-dimensional images will allow the cataloguing of the detailed microstructure of a range of complex materials. The question remains: How can one describe disordered forms of arbitrary shape? We need to quantify random morphologies obtained experimentally utilising tools from integral, statistical and differential geometry and topology, and generate a complete inventory of forms using these techniques. This is of fundamental interest to scientists grappling with the quantification and comparison of disorder in general. Therefore, in Part II, stochastic techniques and geometric measures are introduced to characterise spatial structures occurring, for instance, in the physical systems introduced in Part I. Its first part focuses on the morphology, i.e., on the shape and connectivity of spatial domains, whereas in the second part the spatial arrangement, i.e., correlation functions and distance distributions, play a more important role.

Various statistical methods for the characterisation of textures, shapes and topological properties of geometrical structures are described. In contrast to the papers in Part I, the texts here are method-oriented. Pierre Soille describes in ‘Morphological Texture Analysis: An Introduction’ texture analysis methods based on ideas from mathematical morphology. Texture is usually defined as ‘the characteristic physical structure given to an object by the size, shape, arrangement, and proportions of its parts’. It is shown that granulometries, certain families of transformations of images constructed by means of structuring elements, are very helpful for describing textures. A particular case of such a transformation is the construction of the parallel set for various radii. Soille shows that it is useful to evaluate the transformed images by geometrical measures such as area or volume. A project for the near future is to use other geometrical measures as well and, in particular, to use Minkowski functionals of dimensions smaller than d if the object of interest lives in R^d . Soille studies in detail two of the possible structuring elements: discrete lines and discs, and then describes the application of granulometries for the characterisation of anisotropy.

Claus Beisbart, Robert Dahlke, Klaus Mecke and Herbert Wagner demonstrate in ‘Vector- and Tensor-Valued Descriptors for Spatial Patterns’ the successful application of an advanced concept of integral geometry: additive tensor valuation. This is an important generalisation of the well-known Minkowski functionals and enable novel characterisations of orientations in structures of physical interest. Applications to density functional theory for fluids (see Part I) are possible, as are applications to the galaxy distribution in the universe and the geometry of the electric charge distribution in molecules, which may be relevant in chemistry.

The next two papers describe methods of computational topology. As already emphasised in Part I, one important problem in the analysis of three-dimensional image data is the estimation of topological characteristics such as the Euler characteristic of a porous medium (cf. Arns et al. and Vogel). The Euler characteristic is an appropriate

parameter for the quantification of the connectivity of a phase and of its complement. If the structure is on the scale of the resolution of the grid of observation points, then the crucial point for the study of topological properties of a discretized set is the choice of an adjacency system in a point lattice. The theoretical foundation of the concepts of adjacency is still under development. In the paper ‘The Euler Number of Discretized Sets – On the Choice of Adjacency in Homogeneous Lattices’ by Joachim Ohser, Werner Nagel, and Katja Schladitz, the commonly used graph theoretic approach is related to convex and integral geometry. This allows an improved mathematical approach and a proof of the consistency of certain estimators of the Euler characteristics of a set and of its complement (‘foreground’ and ‘background’ of an image, respectively). Sufficient conditions for the unbiasedness of the estimators are also formulated.

Vanessa Robins extends in ‘Computational Topology for Point Data: Betti Numbers of α -Shapes’ the topic of Minkowski functionals by considering Betti numbers. These numbers, which were formulated in algebraic topology, yield more detailed information on the topology of structures in physics than the Euler characteristic. In fact, the Euler characteristic is simply a linear combination of Betti numbers. In this paper, the Betti numbers are determined for α -shapes, which are planar sets of the form $S \oplus b(o, \alpha)$, where S is the data set, a finite point set approximating some target set X , and $b(o, \alpha)$ is the disc of radius α centred at the origin o . In other words, the α -shape is the parallel set of S with radius α . The aim of these calculations is to obtain topological information on X by using various values of α .

Dietrich Stoyan, Ashot Davtyan and Daulet Turetayeve present in ‘Shape Statistics for Random Domains and Particles’ a short survey on the methods developed by statisticians for the description of the shape of planar sets such as images of particles, grains or vesicles. This includes the simple shape indices like area/perimeter ratios, but offers more sophisticated approaches using various functions and models. Additionally, a family of models of planar ‘lattice animals’ is introduced. These models have some similarity to the smoother objects studied in Part I by Schwarz et al. and Lösche et al.

The rest of Part II is closer still to classical methods of spatial statistics. Daniel Hug, Günter Last and Wolfgang Weil discuss in their long paper ‘A Survey on Contact Distributions’ contact distribution functions systematically. The classical spherical contact distribution $H_s(r)$ is the probability distribution function of the random distance from a random test point outside a structure X to the boundary of X . This function is closely related to the d -dimensional Minkowski measure intensity of $X \oplus b(o, r)$, which is the simplest of Mecke’s morphological functions. For the Boolean model with isotropic grains there are beautiful and well-known formulae for $H_s(r)$. This contribution gives formulae for contact distribution functions corresponding to structuring elements different to the sphere or disc used in $H_s(r)$, also for anisotropic Boolean models. Furthermore, Poisson cluster point processes and grain models constructed by means of such point processes are considered. Finally, contact distribution functions of inhomogeneous cluster processes and Boolean models are presented.

Claus Beisbart, Martin Kerscher and Klaus Mecke apply in ‘Mark Correlations: Relating Physical Properties to Spatial Distributions’ ideas of the second-order theory of marked point processes to physical data. In one of their applications, the ‘points’ are positions of galaxies and the ‘marks’ are characteristics of the galaxies such as luminosity.

They use known types of mark correlation functions and introduce some new concepts which are particularly powerful for the case where the marks are directions. Their aim is to interpret physically the information obtained.

Kasper Berthelsen and Jesper Møller study in ‘Spatial Jump Processes and Perfect Simulation’ finite spatial jump processes. These are Markov processes with piecewise constant sample paths which play an important role in the simulation of Gibbs point processes. Known particular cases are spatial birth-and-death processes. It is shown how perfect simulation enables an efficient form of simulation of these processes. As a particular case, their application to the case of planar hard core Gibbs processes is demonstrated.

Gunter Döge and Dietrich Stoyan describe in ‘Statistics for Non-Sparse Spatially Homogeneous Gibbs Point Processes’ the application of the Takacs–Fiksel method to point data. The idea is to interpret a given pattern as a sample of a homogeneous Gibbs point process with a pair potential and to estimate parameters of the potential function by means of a least squares technique. The new problem here is that the patterns analysed are dense, while in the statistical literature typically sparse patterns have been analysed.

Turbulence is ubiquitous in nature and is not only observed in fluid flow. Although responsible for many interesting disordered structure formation, its detailed mechanisms are still not understood. Martin Greiner introduces in ‘Spatial Statistics of a Turbulent Random Multiplicative Branching Process’ random multiplicative branching processes as empirical models for fully developed turbulence. In order to compare these geometric cascade models with experimental data, it is necessary to discuss spatial statistics in terms of a generating functional. This application of a statistical standard technique closes the second part of this volume.

It is a pleasure to thank all participants of the second international Wuppertal workshop on ‘Statistical Physics and Spatial Statistics’ for their valuable contributions, their openness to share their experience and knowledge, and for the numerous discussions which made the workshop so lively and fruitful. The workshop took place at the University of Wuppertal from 5 to 9 March 2001. Its aim was to provide a forum for the exchange of fundamental ideas between physicists and spatial statisticians, both working in a wide spectrum of science related to stochastic geometry. This volume comprises the majority of the papers presented orally at the workshop as plenary lectures. The editors are in particular grateful to all authors of this volume for their additional work.

Stuttgart and Freiberg,
September 2002

Klaus Mecke
Dietrich Stoyan

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