Uniqueness of Reconstruction of Multiphase Morphologies from Two-Point Correlation Functions

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The restoration of the spatial structure of heterogeneous media, such as composites, porous materials, microemulsions, ceramics, or polymer blends from two-point correlation functions, is a problem of relevance to several areas of science. In this contribution we revisit the question of the uniqueness of the restoration problem. We present numerical evidence that periodic, piecewise uniform structures with smooth boundaries are completely specified by their two-point correlation functions, up to a translation and, in some cases, inversion. We discuss the physical relevance of the results.

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The restoration of the spatial structure of heterogeneous materials from two-point correlation functions is an inverse problem of fundamental importance, which has met with sustained interest over the past few decades [1–11]. Many experimental techniques used for the characterization of heterogeneous media, for example, small-angle x-ray and neutron scattering, time dynamics of energy transfer [12], or distant dipolar field nuclear magnetic resonance [13] yield structural information in the form of two-point correlation functions. It is generally assumed that there are many different structures compatible with a given two-point correlation function [5,9]. While this is the case for infinite random media, we provide strong evidence that continuous media with a periodic structure and smooth boundaries are completely specified by their two-point correlation functions. As will be discussed, this result corresponds well to the known unique character of a number of related problems.

In this paper we consider the case of *complete segregation* of the components into separate phases [14]. The microstructure can then completely be specified in terms of characteristic density functions $\rho_i(\mathbf{r})$:

$$\rho_i(\mathbf{r}) = \begin{cases} 1, & \text{when } \mathbf{r} \text{ is within phase } i, \\ 0, & \text{otherwise.} \end{cases} \quad (1)$$

In order to describe a heterogeneous medium numerically, a finite cell $\mathcal{V}$ subject to periodic boundary conditions is introduced. The two-point correlation functions $G_{ij}(\mathbf{r})$ are

$$G_{ij}(\mathbf{r}) = \langle \rho_i(0)\rho_j(\mathbf{r})\rangle = \frac{1}{V}\int_{\mathcal{V}} d\mathbf{r}'\rho_i(\mathbf{r}')\rho_j(\mathbf{r}+\mathbf{r}'), \quad (2)$$

where integration is over the volume of the periodic cell, $V = \int_{\mathcal{V}} d\mathbf{r}$. The discrepancy Δ between a configuration $\{\rho_i(\mathbf{r})\}$ and the target is defined as

$$\Delta = \sum_{ij} ||G_{ij}(\mathbf{r}) - G^t_{ij}(\mathbf{r})||, \quad (3)$$

where $||\dots||$ denotes the chosen norm, and $\{G^t_{ij}(\mathbf{r})\}$ is a set of target correlation functions. The restoration problem, laid out in Eqs. (1)–(3), is to find a set of density functions $\{\rho_i(\mathbf{r})\}$ that are consistent with a given set of correlation functions $\{G^t_{ij}(\mathbf{r})\}$.

For the case of a finite digitized system, this problem has recently been solved and its uniqueness has been proved [15]. For infinite systems, two fundamentally different approaches have been proposed. The first one [2–8] is based on a scheme originally proposed by Cahn [1], associating interfaces between two phases with isosurfaces of a correlated random Gaussian field.

The second approach, a stochastic reconstruction procedure proposed by Torquato and co-workers [9], is based on discretization of the spatial structure on a grid, each voxel of which is attributed to a single phase. The method departs from an arbitrary configuration that is consistent with the given volume fractions of phases, and then minimizes the discrepancy between the actual and target correlation functions by swapping pairs of voxels according to a Monte Carlo procedure. This discrete stochastic minimization approach is general and flexible, but computationally very demanding [9,10].

Recently we proposed [16] a highly efficient variant of the stochastic reconstruction algorithm, which uses only integer arithmetics. It therefore avoids any accumulation of roundoff errors, which is of particular importance in the present context.

Figures 1–4, present side by side a target configuration and the result of the reconstruction, obtained with the algorithm [16] using as input nothing but the correlation functions calculated from the target structures [19]. Full convergence (i.e., $\Delta \equiv 0$) is achieved in all cases.

The most striking feature of these results is the uniqueness of the reconstructions. Irrespective of whether a "random" structure (Figs. 1–4) [20] or a regular pattern of high symmetry (Figs. 2) are studied, and irrespective of the number of spatial dimensions, the result of the reconstruction process is always identical to the target structure up to a translation. This finding is in contradiction to conclusions drawn in earlier studies [5,9]. It also apparently contradicts the well-known fact that an

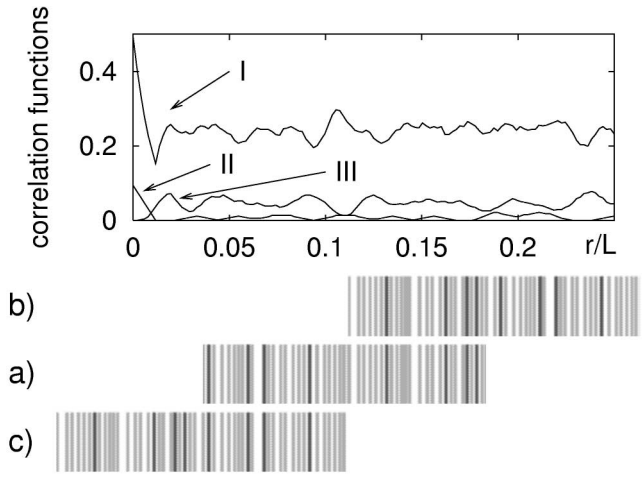

FIG. 1. Complete reconstruction of a one-dimensional system of nonoverlapping rods subject to periodic continuation conditions. Two types of rods, indicated in black and gray, are present in the structure. Top panel: correlation function used for the reconstruction: I and II—autocorrelation functions, III—cross correlation function. Bottom panel: (a) target configuration; (b) and (c) the same result of reconstruction shifted horizontally to demonstrate identity of target and reconstruction.

arbitrary function is specified only by the complete set of all n-point, $n = 1, \ldots, \infty$, spatial correlation functions (see, e.g., [14] and references cited therein), which has led to the notion that any reconstruction based only on two-point correlation function must produce nonunique results. Our findings show that the two-point correlation functions plus the constraints that characteristic density functions be positive and bounded everywhere are generally sufficient to warrant a unique solution.

This gives rise to the question of whether unique reconstruction is possible for all periodic structures, or whether there are exceptional cases. Examples of distinct structures with the same pair correlation functions are known [21–25]. In the context of crystallography, such homometric sets have been studied extensively [21,22].

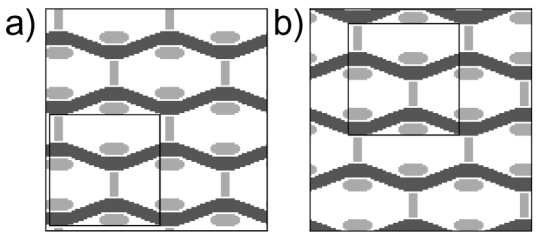

FIG. 2. Complete reconstruction of a triphase centrosymmetric knitted structure analogous to observed experimentally triblock copolymer system [17]. (a) target configuration; (b) reconstruction. Square boxes indicate elementary cells. The system was digitized on a 512×512 grid. The displacement of the reconstruction relative to the original structure is (40, 104) pixels.

Their existence is, however, unlikely to be relevant in the present context because the homometric property is not robust with respect to resolution enhancement. Homometric sets are invariably composed of well-separated points that are arranged in a highly symmetric manner. Digitized heterogeneous media, by contrast, always contain extended continuous regions of voxels representing the same phase. Homometric sets of this type are not known.

A systematic method to artificially construct different spatial structures that will give rise to the same autocorrelation function has been described [23,24]. The scheme is based on the fact that autocorrelation functions are insensitive to point inversion. However, as soon as cross correlation functions are used in addition to the autocorrelation function, all degeneracies of this type are lifted.

A theory that would unambiguously define the class of systems that can be uniquely reconstructed does not yet exist. We therefore have to limit our discussion to pointing out a number of closely related problems in physics and mathematical morphology whose unique character is well established.

The first example is the famous *phase problem* of crystallography. Generally, diffraction methods yield scattering intensities $I(\mathbf{q})$, which are proportional to the Fourier transform of the autocorrelation function $G(\mathbf{r})$ of the scattering length density $\rho(\mathbf{r})$, $I(\mathbf{q}) \sim \int d\mathbf{r} e^{i\mathbf{q}\mathbf{r}} G(\mathbf{r})$. The problem of finding $\rho(\mathbf{r})$ from the observed $I(\mathbf{q})$ is closely analogous to the reconstruction problem considered here. As has been shown by Karle and Hauptmann [26] the constraints that $\rho(\mathbf{r})$ be *positive* everywhere and representable by additive contributions from the atoms in the crystal structure are in general more than sufficient to

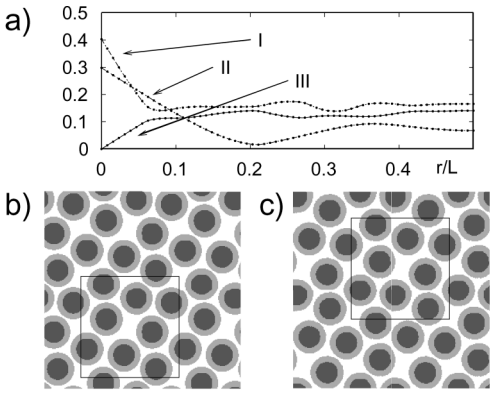

FIG. 3. Complete reconstruction of a disordered particulate system of disks of phase A enclosing a "nucleus" of phase B, interspersed in a matrix C. Structures of this type have been observed using scanning force microscopy in block-copolymer films [18]. Top panel: (a) linear sections of the 2D correlation functions used for the reconstruction process. Bottom panel: (b) target configuration; (c) reconstruction. Elementary cells of the periodic structure are represented by square boxes.

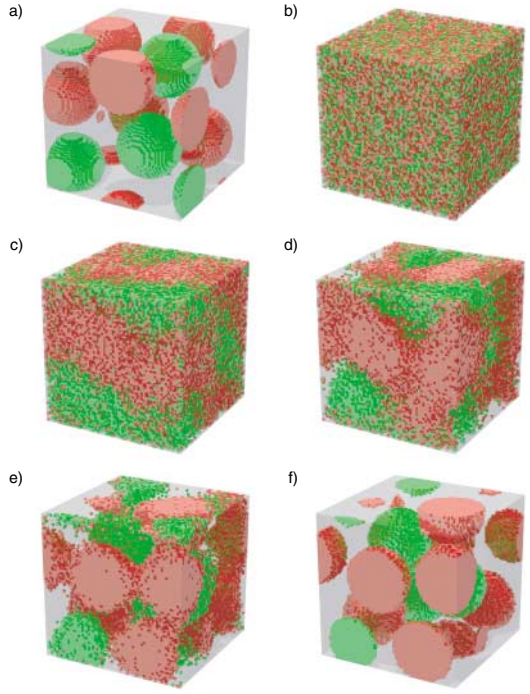

FIG. 4 (color online). Complete reconstruction of a disordered particulate system of spheres of phases A and B, interspersed in a matrix C. Three-dimensional structure is digitized on a $64 \times 64 \times 64$ grid; (a) target configuration; (b)–(f) intermediate steps of reconstruction. The size and location of the spheres in (f) are identical to (a) up to a translation. This was verified by calculating the correlation functions between (f) and (a).

warrant a unique solution. This observation forms the basis of the *direct methods* of structure determination. In the present context, the constraints on the characteristic density are even more severe than in case of x-ray crystallography. Indeed, the total number of unknowns, given by the number of nonzero voxels in the digitized unit cell, is less than the number of constraints produced by the conditions Eq. (1) and the system of nonlinear Eqs. (2).

The second class of related problems, namely, the characterization of random geometric structures by their correlation functions or their chord length distributions, has been a long-standing topic in integral geometry [27] and mathematical morphology [28]. Rigorous mathematical results from these efforts are consistent with our findings. In particular, it has been shown that convex polygons are uniquely determined by their correlation functions [29].

A third close relative to the reconstruction problem can be found in the domain of spin systems. In the discretized form, the reconstruction problem is equivalent to the search for a ground state of a spin system with long-range multi-spin interactions. Restricting the discussion to two-phase systems for simplicity, the characteristic density $\rho_{\mathbf{m}}$ Eq. (1) at voxel $\mathbf{m}$ can be written using the spin operator $S_{\mathbf{m}}$ as $\rho_{\mathbf{m}} = \frac{1}{2}(1 + S_{\mathbf{m}})$. Substituting this into Eq. (2) and using the 2-norm, the problem under study is reduced to the search for the ground state of the spin Hamiltonian

$$H = -\sum_{\mathbf{m},\mathbf{n}} G^t(\mathbf{m}-\mathbf{n}) S_{\mathbf{m}} S_{\mathbf{n}} + \sum_{\mathbf{m},\mathbf{n},\mathbf{l}} S_{\mathbf{m}} S_{\mathbf{m}+\mathbf{l}} S_{\mathbf{n}} S_{\mathbf{n}+\mathbf{l}}, \quad (4)$$

where $G^t(\mathbf{r})$ is the target two-point correlation function.

For the particular case of $G^t(\mathbf{r}) \equiv 0$ the ground state of the Hamiltonian Eq. (4) has been studied extensively [30–34] in connection with the problem of binary sequences with low autocorrelation, the so-called Bernasconi problem [35]. For select system sizes, the exact ground state of the Bernasconi problem is known [30,33]. It is always nondegenerate (up to translational degeneracy and inversion, if a system under periodic boundary conditions is considered).

It is important to note that we find the restoration to be unique only if complete convergence to the global minimum of the discrepancy can be achieved. If, due to technical difficulties, this criterion is not met, a variety of different structures may be found.

Uniqueness is lost as one goes from a periodic to an infinite aperiodic system. The numerical restoration becomes progressively more difficult as one uses a periodic cell of growing size relative to the decay length of the correlation functions, because the discrepancy Δ associated with structures differing in distant features becomes smaller and smaller. Finally, in the limit of an aperiodic system (infinite cell size) different structures become truly indistinguishable, and the system cannot be characterized completely by two-point correlations.

In conclusion, our results provide strong empirical evidence that the restoration of periodic, piecewise uniform structures with smooth boundaries from their two-point correlation functions is unique up to translation, and in some cases, inversion. "Smooth" in this context refers to the requirement that the length scales of the correlation functions have both an upper bound, given by the periodicity, and a lower bound, given by the shortest wavelength present in the structure. This excludes truly fractal morphologies, where no such lower bound exists.

Since the above conjecture is relevant to a number of different fields of physics, we hope that our empirical evidence will attract attention to the problem, leading to a rigorous mathematical analysis. Our own interest in this topic stems from the context of experimental structure determination by scattering methods. Once the structure factor has been measured, the correlation function is obtained by Fourier transformation, and a structural model can be constructed numerically. In order to estimate the sensitivity of such an approach to imperfections in the experimental data, it is important to know whether

or not it would lead to a unique result in the limit of an error-free measurement.

The uniqueness conjecture also applies to the problem of circular DNA sequencing by partial digestion with restriction enzymes [25].

Another important area of relevance is the kinetic theory of phase separation processes [36–39]. Numerical models of phase transformation processes are usually formulated on a periodic grid. The time-dependent phase morphology is commonly characterized in terms of two-point correlation functions and their moments [40]. The uniqueness conjecture presented here suggests that the two-point correlation functions contain *all* morphological information. At the same time, cases have been reported where different simulation conditions yield structures that appear different to visual inspection, even though their correlation functions are numerically indistinguishable [41]. It is important to note, however, that while uniqueness means that identical correlation functions must come from identical structures, it does not necessarily imply that *similar* correlation functions always arise from similar structures.

The uniqueness conjecture opens several interesting opportunities for future research in addition to the search for a precise formulation and formal proof: Is it possible to calculate higher order correlation functions from pair correlation functions directly, without going through the reconstruction process? How could variational bounds [14] on effective properties of heterogeneous media that use higher order correlation functions be improved? What are the possible implications for pattern recognition? Our results indicate that the class of geometrical objects completely characterized by their covariogramm is much broader than has been previously assumed [29].

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