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ON THE "TRIANGULAR" INEQUALITY IN THE THEORY OF TWO-PHASE RANDOM MEDIA

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A necessary condition on the two-point correlation function of binary random media, noticed by Matheron [1] and called by him "triangular" inequality, is studied in this note. An appropriate result, due to Achiezer and Glazman [2], is first recalled. Simple consequences of this inequality are given, as well as a necessary condition for its validity in a statistically isotropic medium. It is shown that it represents a requirement, independent of that of the familiar positive definiteness, that should be additionally imposed on the two-point correlation function of any realistic binary medium.

Key words. random materials, two-phase media, correlation functions

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Consider a random and statistically uniform medium that occupies d-dimensional space $\mathbb{R}^d$. The medium is "binary", i.e., it consists of two phases labelled 1 and 2. Phase 1 (which needs not to be connected) occupies Ω_1 and phase 2 occupies its complement Ω_2. The characteristic function of Ω_1 is f_1. Thus,

$$f_1(x) = \begin{cases} 1, & \text{if} \quad x \in \Omega_1, \\ 0, & \text{otherwise.} \end{cases}$$

As it is well-known, the statistical properties of the medium follow from the set of multipoint probabilities or moments of f_1:

$$\eta_1 = \langle f_1(0) \rangle\,, \quad \langle f_1(0) f_1(z_1) \rangle\,, \ldots, \tag{1}$$

where each $z_k \in \mathbb{R}^d$, see for instance [3]. The angled brackets signify ensemble averaging. Such multipoint probabilities are symmetric in their arguments. One point could be taken at the origin, because of the assumed statistical uniformity.

It is, in fact, convenient to work with η_1 and the multipoint moments

$$M_p(z_1, z_2, \ldots, z_{p-1}) = \langle f_1'(0) f_1'(z_1) f_1'(z_2) \ldots f_1'(z_{p-1}) \rangle, \quad p = 2, 3, \ldots, \qquad (2)$$

where

$$f_1'(z) = f_1(z) - \eta_1 \qquad (3)$$

is the fluctuating part of the field $f_1(z)$.

Of course, not any infinite hierarchy of functions M_p can represent moments derived from a random medium and, moreover, from a two-phase one. The reason, well recognized and very clearly explained by Frisch [4], is that the function M_p should satisfy, in particular, certain compatibility conditions. The real problem in this connection arises when modelling a random constitution of practical interest. In such cases the first few moments (as a rule the two-point and, more rarely, the three-point ones) are prescribed using certain, very often heuristic and not very rigorous arguments. Though the form of the prescribed moments can, in principle, be checked experimentally, the question remains as to whether these moments can be inserted into the infinite hierarchy of multipoint moments (2), i.e. whether they pertain to a *real* random medium. The problem is even tougher when the two-phase media are dealt with, having in mind that the latter very often appear in application. Frisch [4], for example, presented examples of two-point probability densities that look plausible but *cannot* belong to any real random medium. Another more recent example is connected with the often used "well-stirred" approximation for random dispersion of spheres, for which, as far as the two-point moment is only concerned, overlapping is forbidden and the sphere location is not statistically interconnected otherwise. This approximation turns out to be realistic only at sphere fraction $\eta_1 \leq 1/8$ in 3D, as shown in [5, 6].

For any statistically homogeneous medium one restriction that is generally known is that its two-point correlation function should be positive definite, so that its Fourier transform must be positive. The converse is also true, namely, for any positive-definite function there exists a random medium for which this function represents its two-point correlation (the Bochner or Bochner-Khinchine theorem, see, e.g., [3]). Further restrictions are known if the medium is also statistically isotropic [3]. For two-phase media, as introduced above, it *ought* to be possible to find more restrictions but none are known; a conjecture on how to recognize realistic two-point correlation functions for such media was recently made by Matheron [1]. As a matter of fact, a method for deriving relations of such a type has been proposed in the recent work [6] on the basis of a certain variational reasoning.

Here we shall study in more detail a requirement, specific for the correlation of a two-phase medium. This is an inequality first noticed, to the best of the author's knowledge, by Matheron [1] and called by him "triangular" due to obvious geometrical reasons. It appears that this inequality closely resembles a certain property of the positive definite functions, first pointed out by Achiezer and Glazman [2] almost forty years ago. That is why we shall first recall the appropriate result of Achiezer and Glazman.

Following these authors, introduce the class $\mathcal{G}$ of real and even functions $g(x)$, $x \in \mathbb{R}^d$, for which the kernel

$$\Gamma(x,y) = g(x) + g(y) - g(x-y) \tag{4}$$

is positive definite, i.e.

$$\sum_{i,j=1}^{k} \Big[g(x_i) + g(x_j) - g(x_i - x_j)\Big] a_i a_j \geq 0, \quad \forall x_i \in \mathbb{R}^d,\ a_i \in R. \tag{5}$$

Proposition 1. Let $\gamma_2(x)$ be a real positive definite and even function on $\mathbb{R}^d$. Then $1 - \gamma_2(x) \in \mathcal{G}$ (and thus $\lambda(1-\gamma_2(x)) \in \mathcal{G}$ as well, $\forall \lambda \geq 0$).

Proof. Due to the definition (5), $1 - \gamma_2(x) \in \mathcal{G}$ if the kernel

$$T(x,y) = 1 + \gamma_2(x-y) - \gamma_2(x) - \gamma_2(y) \tag{6}$$

is positive definite. To prove this, consider the identity

$$\sum_{i,j=1}^{2k} \gamma_2(y_i - y_j) b_i b_j = \sum_{i,j=1}^{2k} \gamma_2(y_{2i} - y_{2j-1}) b_{2i} b_{2j-1}$$

$$+ \sum_{i,j=1}^{2k} \gamma_2(y_{2i} - y_{2j}) b_{2i} b_{2j} + \sum_{i,j=1}^{2k} \gamma_2(y_{2i-1} - y_{2j-1}) b_{2i-1} b_{2j-1}$$

$$+ \sum_{i,j=1}^{2k} \gamma_2(y_{2i-1} - y_{2j}) b_{2i-1} b_{2j}.$$

Choose now $y_{2i} = 0$, $y_{2i-1} = x_i$, $b_{2i} = -a_i$, $b_{2i-1} = a_i$, $i = 1, \dots, k$. Then

$$0 \leq \sum_{i,j=1}^{2k} \gamma_2(y_i - y_j) b_i b_j = \sum_{i,j=1}^{k} \Big[1 + \gamma_2(x_i - x_j) - \gamma_2(x_i) - \gamma_2(x_j)\Big] a_i a_j.$$

Hence the kernel $T(x,y)$, see (6), is indeed positive definite, which proves the proposition.

Remark 1. The Proposition 1 and its simple proof, given here for the sake of completeness, belong to Achiezer and Glazman [2], see also [7, p. 265].

Let the medium be two-phase and let

$$\gamma(x', x'') = \gamma(x' - x'') = \frac{1}{2}\Big\langle |f_1(x') - f_1(x'')|^2 \Big\rangle \tag{7}$$

denote the so-called variogramme of the field $f_1(x)$. Using the definition of the two-point correlation, it is easily seen that

$$\gamma(x) = \eta_1\eta_2(1-\gamma_2(x)), \tag{8}$$

where

$$\gamma_2(x) = \frac{M_2(x)}{M_2(0)} = \frac{\langle f_1'(0)f_1'(x)\rangle}{\langle f_1'^2(0)\rangle}, \quad M_2(0) = \big\langle f_1'^2(0)\big\rangle = \eta_1\eta_2,$$

so that $\gamma_2(x)$ is the most often used two-point correlation for which $\gamma_2(0) = 1$.

According to Proposition 1, $\gamma \in \mathcal{G}$, since $\gamma_2(x)$ is positive definite. Hence the field $\Gamma(x, y)$, generated by $\gamma(x)$, see (4), is positive definite. The following proposition shows, however, that for a two-phase medium an additional fact holds.

Proposition 2. The variogramme of any two-phase random medium generates a field $\Gamma(x, y)$ which is not only positive definite, but which is nonnegative itself. In other words, the so-called *triangular inequality* of Matheron [1] holds:

$$\gamma(x-y) \le \gamma(x) + \gamma(y), \quad \forall x, y \in \mathbb{R}^d. \tag{9}$$

Proof. Obviously,

$$\begin{aligned}\gamma(x,y) &= \frac{1}{2}\Big\langle |f_1(x)-f_1(y)|^2\Big\rangle = \frac{1}{2}\Big\langle |f_1(x)-f_1(0)+f_1(0)+f_1(y)|^2\Big\rangle \\ &= \frac{1}{2}\Big\langle |f_1(x)-f_1(0)|^2\Big\rangle + \frac{1}{2}\Big\langle |f_1(0)-f_1(y)|^2\Big\rangle - \alpha(x,y) \\ &= \gamma(x)+\gamma(y)-\alpha(x,y),\end{aligned}$$

where

$$\alpha(x,y) = \Big\langle (f_1(0)-f_1(x))(f_1(0)-f_1(y))\Big\rangle.$$

To prove (9) it suffices to show that $\alpha(x, y) \ge 0$. But, if the origin 0 lies in the constituent '2', then $f_1(0) = 0$ and $\alpha(x, y) = \langle f_1(x)f_1(y)\rangle \ge 0$. Similarly, if 0 lies in the constituent '1', then $f_1(0) = 1$ and again $\alpha(x, y) = \langle (1 - f_1(x))(1 - f_1(y))\rangle \ge 0$.

Combining (8) and (9) yields

$$\gamma_2(x) + \gamma_2(y) - \gamma_2(x-y) \le 1 \tag{10}$$

or

$$1 + \gamma_2(r' + \theta r'') \ge \gamma_2(r') + \gamma_2(r''), \quad \forall\theta \in [-1, 1], \tag{11}$$

having chosen $|x| = r'$, $r'' = |y|$. This inequality should thus be satisfied by the two-point correlation of *any* realistic statistically homogeneous and two-phase random medium.

Corollary 1. Let the medium be statistically isotropic as well, so that $\gamma_2(x) = \gamma_2(r)$, $r = |x|$. Then

$$\gamma_2'(0) \le \pm\gamma_2'(r), \quad \forall r \in (0, \infty). \tag{12}$$

Indeed, choose the vectors x, y colinear, once with the same directions and then with the opposite directions; $|y| = \Delta r$, $|x| = r$, $r > \Delta r > 0$. Then

$$\gamma_2(\Delta r) + \gamma_2(r) \leq 1 + \gamma_2(r \pm \Delta r)$$

which, at $\Delta r \ll 1$, implies (12).

Since $\gamma_2(0) = 1$ and $\gamma_2(r) \leq 1$, we have obviously $\gamma_2'(0) \leq 0$. The inequality (12) is then equivalent to

$$|\gamma_2'(r)| \leq |\gamma_2'(0)|, \quad \forall r \in (0, \infty), \tag{13}$$

which means, in particular, that the steepest decrease of the two-point correlation function $\gamma_2(r)$ of an isotropic two-phase medium is at the origin $r = 0$.

Corollary 2. A positive definite function $\gamma_2(r)$ may serve as a two-point correlation of a two-phase statistically homogeneous and isotropic medium, only if $\gamma_2'(0) < 0$.

Indeed, (13) immediately shows that $\gamma_2'(0) = 0$ yields $\gamma_2'(r) = 0$, $\forall r \in (0, \infty)$, i.e. $\gamma_2(r) \equiv 1$, which is impossible.

The inequality $\gamma_2'(0) < 0$ for a two-phase medium follows also from the fact that $-\gamma_2'(0)$ is proportional to S/V, where S is the specific surface (i.e. phase boundary) within the small volume V, see [8] and especially [9, p. 177] for details and a proof. More precisely, $S/V = -4\eta_1(1 - \eta_1)\gamma_2'(0)$, which obviously implies $\gamma_2'(0) < 0$ for such media.

Remark 2. As it is well-known, not every real and positive function is positive definite and vice versa. Hence the triangular inequality represents a necessary condition that should be imposed on the two-point correlations of random media in *addition* to their positive definiteness, if the modelled medium is two-phase. To illustrate this consider as an example first the function

$$\gamma_2(r) = \frac{1}{(1 + (r/\beta)^2)^2}.$$

It is positive definite (since its Fourier transform is positive) and hence it represents, according to the Bochner-Khinchine theorem, a two-point correlation of a certain statistically homogeneous and isotropic random medium. On the other hand, $\gamma_2'(0) = 0$, so that the triangular inequality fails for this medium and the latter therefore *cannot* be two-phase.

Conversely, consider again the above mentioned “well-stirred” dispersion of spheres. Its two-point correlation satisfies the triangular inequality for all values of the sphere fraction $\eta_1 \in (0, 1)$ (since the field $f_1(x)$ is binary). On the other hand, the Fourier transform of this correlation is positive definite only at $\eta_1 \leq 1/8$, as it can be directly shown. Hence the positive definiteness and triangular inequality are indeed two mutually independent necessary conditions that should be satisfied by the two-point correlation of binary random media.

In general, it seems hard to give a complete description of the functions that satisfy the triangular inequality (10). (The variogrammes under study cannot obviously be homogeneous of degree 1, i.e. $\gamma(\lambda x) \neq \lambda\gamma(x)$, and thus they are not semi-norms on $\mathbb{R}^d$.) A simple and rich class of such function can be easily described though. To this end note that (11) implies $\gamma_2''(0) \geq 0$ for such a function and thus $\gamma_2(r)$ is convex and monotonically increasing in a certain vicinity of the origin. If the latter properties hold for all $r \in [0, \infty)$, it suffices to claim that the respective function is an admissible two-point correlation. More precisely, recall that in 1D a bounded even function which is convex on the right half-axis is positive definite [10, p. 187]. A radially symmetric function $\gamma_2(r)$ in 3D with these properties is not obliged to be of that kind.[1] However, for such functions the following result holds:

Proposition 3. If $\gamma_2(r)$ is monotonically decreasing, nonnegative and convex, then it satisfies the triangular inequality (10).

Proof. Since $\gamma_2(r)$ is monotonically decreasing, in order to prove (10) it suffices to show that

$$1 + \gamma_2(r' + r'') \geq \gamma_2(r') + \gamma_2(r''),$$

having taken the vectors x, y colinear, with the same direction; $r' = |x|$, $r'' = |y|$. Let $r' > r''$ for definiteness. Then

$$1 - \gamma_2(r'') = \gamma_2(0) - \gamma_2(r'') = -\gamma_2'(\xi')r'', \quad \xi' \in (0, r''),$$

$$\gamma_2(r') - \gamma_2(r' + r'') = -\gamma_2'(\xi'')r'', \quad \xi'' \in (r', r' + r'').$$

The convexity of $\gamma_2(r)$ means that $\gamma_2''(r) \geq 0$, so that $\gamma_2'(\xi'') \geq \gamma_2'(\xi')$, because $\xi'' > \xi'$. Hence

$$1 - \gamma_2(r'') \geq \gamma_2(r') - \gamma_2(r' + r''),$$

which proves the proposition.

A simple example of an admissible and physically reasonable two-point correlation is

$$\gamma_2(r) = e^{-\mu r}, \tag{14}$$

proposed by Debye *et al.* [8]. This is the so-called exponential correlation, discussed, for instance, in the book of Stoyan *et al.* [9] (where a planar random set with this correlation is explicitly constructed in Sec. 10.5.1). Being convex, positive and monotonically decreasing, the function (14) satisfies the triangular inequality (10), as it follows from Proposition 3. Its Fourier transform is positive. Hence this function may represent a two-point correlation for a two-phase statistically homogeneous and isotropic medium in $\mathbb{R}^d$ for any d. A more general class of similarly admissible correlations is obviously given by

$$\gamma_2(r) = \int_0^\infty e^{-rt}\, d\sigma(t); \tag{15}$$

[1] The function in 3D

$$\gamma_2(r) = \begin{cases} 1 - r/a, & \text{if } r \leq a, \\ 0, & \text{if } r > a, \end{cases}$$

$r = |x|$, is bounded and convex, but its Fourier transform $\widehat{\gamma_2}(k)$ is proportional to $2(1 - \cos ak) - ak \sin ak$ and hence it is not positive everywhere.

here $\sigma(t)$ is an arbitrary bounded and non-increasing function on $(0,\infty)$ such that $\int_0^\infty d\sigma(t) = 1$. (If $\sigma(t) = H(t-\mu)$, Debye's function (14) is recovered from (16), $H(t)$ being the Heaviside function.) In other words, the class (16) gathers the Laplace transforms of all nonnegative functions on $(0,\infty)$ (more precisely, of all bounded measures there).

Note finally that the class (15) coincides with the class of the so-called completely monotonic functions, according to the well-known Bernstein theorem, see, for instance, [11] or [7]. It is curious, however, that such completely monotonic functions (15) may represent correlations only for dispersions of overlapping or touching particles. The reason is that non-overlapping always implies the condition $\gamma_2''(0) = 0$, as it follows from the results of Kirste and Porod [12], see also [13] and [5]. This condition, however, never holds for the functions (15).

Another example of an admissible two-point correlation is the function

$$\gamma_2(r) = \begin{cases} 1 - \dfrac{3r}{4a} + \dfrac{r^3}{16a^3}, & \text{if } r \le 2a, \\ 0, & \text{if } r > 2a, \end{cases} \tag{16}$$

since it is obviously positive definite, nonnegative and convex. Hence it satisfies the triangular inequality (10) as well. Note that (16) is the two-point correlation of the so-called Miller's cell material [14] in the simplest case when the cells are spherical, see also [15].

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