Exact conditions on physically realizable correlation functions of random media

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Algorithms have been developed recently to construct realizations of random media with specified statistical correlation functions. There is a need for the formulation of exact conditions on the correlation functions in order to ensure that hypothetical correlation functions are physically realizable. Here we obtain positivity conditions on certain integrals of the autocorrelation function of d-dimensional statistically homogeneous media and of statistically isotropic media. These integral conditions are then applied to test various classes of autocorrelation functions. Finally, we note some integral conditions on the three-point correlation function. [S0021-9606(99)51343-3]

I. INTRODUCTION

An intriguing inverse problem is the reconstruction of realizations of random heterogeneous media, such as porous and composite media, with specified statistical correlation functions.[1–6] Typically the algorithms are used to ''reconstruct'' an actual random medium (experimental or theoretical) using correlation functions that are determined experimentally or theoretically. Recently, it has been proposed that the same algorithms be used to ''construct'' realizations of random media that have specified model or hypothetical correlation functions.[4,5] Such a program may ultimately lead to a systematic means of classifying the microstructure of random media.

In light of these developments, there is a need for the formulation of exact conditions on the correlation functions in order to ensure that hypothetical correlation functions are physically realizable. In this article, we utilize the spectral representation of the autocorrelation function (two-point correlation function) $S_2(\mathbf{r})$ to obtain positivity conditions on certain integrals of S_2 for random media in any space dimension d. Moreover, we show that if the random medium is also statistically isotropic, there are d different positivity conditions that can be exploited. These integral conditions are then employed to test various classes of correlation functions that have recently been proposed. Finally, we note some integral conditions on the three-point correlation function. We begin with a basic review of the definitions and properties of the n-point correlation functions.

II. *n*-POINT CORRELATION FUNCTIONS

For completeness, we review definitions and basic properties of the n-point correlation functions. Much of this discussion follows that of Torquato and Stell.[7]

A. Definitions

The two-phase random medium is a domain of space $\mathcal{V}(\omega)\in\mathcal{R}^d$ of volume V which is composed of two regions or phases: phase 1, the region $\mathcal{V}_1$ of volume fraction ϕ_1 and phase 2, the region $\mathcal{V}_2$ of volume fraction ϕ_2. Let $\partial\mathcal{V}$ denote the surface or interface between $\mathcal{V}_1$ and $\mathcal{V}_2$. For a given realization ω, the characteristic function $\mathcal{I}^{(i)}(\mathbf{x})$ of phase i is defined by

$$\mathcal{I}^{(i)}(\mathbf{x})=\begin{cases}1, & \text{if } \mathbf{x}\in\mathcal{V}_i,\\ 0, & \text{if } \mathbf{x}\notin\mathcal{V}_i.\end{cases} \tag{1}$$

It is natural to consider multipoint statistics that are based on expectations of products of the characteristic function $\mathcal{I}^{(i)}(\mathbf{x})$. The simplest multipoint statistic is the one-point correlation function $S_1^{(i)}(\mathbf{x}_1)$ defined to be

$$S_1^{(i)}(\mathbf{x}_1)=\langle\mathcal{I}^{(i)}(\mathbf{x}_1)\rangle, \tag{2}$$

where angular brackets denote an ensemble average. More generally, the n-point correlation function $S_n^{(i)}(\mathbf{x}_1,\mathbf{x}_2,\ldots,\mathbf{x}_n)$ is defined by

$$S_n^{(i)}(\mathbf{x}_1,\mathbf{x}_2,\ldots,\mathbf{x}_n)=\langle\mathcal{I}^{(i)}(\mathbf{x}_1)\mathcal{I}^{(i)}(\mathbf{x}_2)\cdots\mathcal{I}^{(i)}(\mathbf{x}_n)\rangle. \tag{3}$$

The function $S_n^{(i)}$ is equal to the *probability of finding n points in phase i at positions* $\mathbf{x}_1,\mathbf{x}_2,\ldots,\mathbf{x}_n$ and hence is also referred to as the *n-point probability function* for phase i.

B. Symmetries and ergodicity

If the n-point probability function $S_n^{(i)}$ depends on the absolute positions $\mathbf{x}_1$, $\mathbf{x}_2$, ..., $\mathbf{x}_n$, then we say that the medium is *statistically inhomogeneous*. The medium is *statistically homogeneous* if $S_n^{(i)}$ is *translationally invariant*, i.e., invariant under translation of the spatial coordinates. This means that for some constant vector $\mathbf{y}$

$$\begin{aligned}S_n^{(i)}(\mathbf{x}_1,\mathbf{x}_2,\ldots,\mathbf{x}_n)&=S_n^{(i)}(\mathbf{x}_1+\mathbf{y},\mathbf{x}_2+\mathbf{y},\ldots,\mathbf{x}_n+\mathbf{y})\\&=S_n^{(i)}(\mathbf{x}_{12},\ldots,\mathbf{x}_{1n}),\end{aligned} \tag{4}$$

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where $\mathbf{x}_{jk}=\mathbf{x}_k-\mathbf{x}_j$. We see that for statistically homogeneous media, the n-point probability function depends not on the absolute positions but on their relative displacements. Thus, there is no *preferred origin* in the system, which in relation (4) we have chosen to be the point $\mathbf{x}_1$. The one-point probability function, in this instance, is a constant *everywhere*, namely, the volume fraction ϕ_i of phase i, i.e.,

$$S_1^{(i)}=\phi_i. \tag{5}$$

When the system is statistically homogeneous, the *ergodic hypothesis* enables one to replace ensemble averaging with volume averaging in the limit that the volume tends to infinity, i.e.,

$$S_n^{(i)}(\mathbf{x}_{12},\ldots,\mathbf{x}_{1n})=\lim_{V\to\infty}\frac{1}{V}\int_{\mathcal{V}}\mathcal{I}^{(i)}(\mathbf{y})\mathcal{I}^{(i)}(\mathbf{y}+\mathbf{x}_{12})\cdots\mathcal{I}^{(i)}(\mathbf{y}+\mathbf{x}_{1n})d\mathbf{y}. \tag{6}$$

The medium is statistically homogeneous but *anisotropic* if $S_n^{(i)}$ depends on both the orientations and magnitudes of the vectors $\mathbf{x_{12}},\mathbf{x_{13}},\ldots,\mathbf{x_{1n}}$. The medium is said to be *statistically isotropic* if the multipoint probability function of interest is *rotationally invariant*, i.e., invariant under rigid-body rotation of the spatial coordinates. For such media, this implies $S_n^{(i)}$ depends only on the distances $x_{jk}=|\mathbf{x}_{jk}|$, $1\leqslant j<k\leqslant n$. For example, for isotropic media, the two- and three-point functions have the form

$$\begin{aligned}S_2^{(i)}(\mathbf{x}_1,\mathbf{x}_2)&=S_2^{(i)}(x_{12}),\\ S_3^{(i)}(\mathbf{x}_1,\mathbf{x}_2,\mathbf{x}_3)&=S_3^{(i)}(x_{12},x_{13},x_{23}).\end{aligned} \tag{7}$$

Both $S_2^{(i)}$ and $S_3^{(i)}$ can be obtained from *any planar cut* through the medium when it is isotropic. Moreover, $S_2^{(i)}$ can also be found from a *lineal* cut through an isotropic medium.

C. Known exact conditions

The n-point function for phase 2, $S_n^{(2)}$, can be expressed in terms of the set of phase 1 functions $S_1^{(1)},S_2^{(1)},\ldots,S_n^{(1)}$.[7] For example, for $n=2$, we have

$$S^{(2)}(\mathbf{r})-\phi_2^2=S^{(1)}(\mathbf{r})-\phi_1^2, \tag{8}$$

where $\mathbf{r}=\mathbf{x}_{12}$. Accordingly, we will consider the n-point function for phase 1 only and, for brevity, denote it by $S_n\equiv S_n^{(1)}$. Exact conditions (asymptotic properties and bounds) have been given for S_n for any n.[7] In what follows, we will state such known exact conditions on the two- and three-point functions, in particular.

1. Two-point function

For statistically homogeneous media, the two-point function (or *autocorrelation function*) $S_2(\mathbf{r})$ obeys the following conditions at the extreme values of its argument:

$$\lim_{\mathbf{r}\to 0}S_2(\mathbf{r})=\phi_1,\quad \lim_{\mathbf{r}\to\infty}S_2(\mathbf{r})=\phi_1^2. \tag{9}$$

The second condition assumes no long-range order. Elementary bounds are given by

$$0\leqslant S_2(\mathbf{r})\leqslant\phi_1. \tag{10}$$

Debye, Andersen, and Brumberger[8] showed that the slope of S_2 at the origin is equal to $-s/4$ for three-dimensional *isotropic* media, where s is the specific surface (interface area per unit volume), which is always positive. In any spatial dimension d, the slope of S_2 at the origin is proportional to the negative of s and therefore

$$\left.\frac{dS_2}{dr}\right|_{r=0}\leqslant 0. \tag{11}$$

For the first three space dimensions, this derivative is explicitly given by

$$\left.\frac{dS_2}{dr}\right|_{r=0}=\begin{cases}-s/2, & d=1,\\ -s/\pi, & d=2,\\ -s/4, & d=3.\end{cases} \tag{12}$$

We note that Berryman[9] showed that the derivative of the angular average of the *anisotropic* correlation function of three-dimensional media has the same relationship to s as the isotropic result.

2. Three-point function

For statistically homogeneous media without long-range order, the three-point function S_3, under permutations of the distances x_{12}, x_{13}, and x_{23}, has the following asymptotic properties:

$$\begin{aligned}&\lim_{x_{12}\to 0,x_{13}\to 0}S_3(\mathbf{x}_{12},\mathbf{x}_{13})=\phi_1,\\ &\lim_{x_{23}\to 0}S_3(\mathbf{x}_{12},\mathbf{x}_{13})=S_2(\mathbf{x}_{12}),\end{aligned} \tag{13}$$

$$\begin{aligned}&\lim_{\substack{x_{13}\to\infty\\ x_{12}\ \text{fixed}}}S_3(\mathbf{x}_{12},\mathbf{x}_{13})=\phi_1 S_2(\mathbf{x}_{12}),\\ &\lim_{\text{all }x_{ij}\to\infty}S_3(\mathbf{x}_{12},\mathbf{x}_{13})=\phi_1^3.\end{aligned} \tag{14}$$

Elementary bounds are given by

$$0\leqslant S_3(\mathbf{x}_{12},\mathbf{x}_{13})\leqslant\min[S_2(\mathbf{x}_{12}),S_2(\mathbf{x}_{13}),S_2(\mathbf{x}_{23})]. \tag{15}$$

III. EXACT INTEGRAL CONDITIONS ON S_2

In the study of the spectral representation of time series (one-dimensional random processes), it is well known that the Fourier representation of the autocorrelation function S_2 leads to certain positivity conditions on integrals of S_2.[10] Such conditions are also well-known in the theory of turbulence.[11] Here we obtain such results for statistically homogeneous random media in any space dimension d. Moreover, we show that if the random medium is also statistically isotropic, there are d different positivity conditions that one can utilize.

The Fourier transform of some arbitrary function $f(\mathbf{r})$ in d dimensions is given by

$$\tilde{f}(\mathbf{k})=\int f(\mathbf{r})e^{-i\mathbf{k}\cdot\mathbf{r}}d\mathbf{r}, \tag{16}$$

and the associated inverse operation is defined by

$$f(\mathbf{r}) = \frac{1}{(2\pi)^d} \int \tilde{f}(\mathbf{k}) e^{i\mathbf{k}\cdot\mathbf{r}} d\mathbf{k}. \tag{17}$$

When the function just depends on the modulus $r = |\mathbf{r}|$, then we have the following simpler expressions for the first three space dimensions:

$$\begin{aligned} \tilde{f}(k) &= 2\int_0^\infty dr f(r)\cos kr, \\ f(r) &= \frac{1}{\pi}\int_0^\infty dk \tilde{f}(k)\cos kr, \quad d=1, \end{aligned} \tag{18}$$

$$\begin{aligned} \tilde{f}(k) &= 2\pi\int_0^\infty dr f(r) r J_0(kr), \\ f(r) &= \frac{1}{2\pi}\int_0^\infty dk \tilde{f}(k) k J_0(kr), \quad d=2, \end{aligned} \tag{19}$$

$$\begin{aligned} \tilde{f}(k) &= \frac{4\pi}{k}\int_0^\infty dr f(r) r \sin kr, \\ f(r) &= \frac{1}{2\pi^2 r}\int_0^\infty dk \tilde{f}(k) k \sin kr, \quad d=3, \end{aligned} \tag{20}$$

where $k = |\mathbf{k}|$ and $J_0(x)$ is the zeroth-order Bessel function of the first kind.

We consider statistically homogeneous media and define the covariance function $\gamma(\mathbf{r}) = S_2(\mathbf{r}) - \phi_1^2$. Invoking the ergodic hypothesis, we can express $\gamma(\mathbf{r})$ as the volume integral

$$\gamma(\mathbf{r}) = \lim_{V\to\infty} \frac{1}{V} \int J(\mathbf{x}) J(\mathbf{x}+\mathbf{r}) d\mathbf{x}, \tag{21}$$

where $J(\mathbf{x}) \equiv I^{(1)}(\mathbf{x}) - \phi_1$ is a random variable with zero mean and $\mathcal{I}^{(1)}(\mathbf{x})$ is the characteristic function of phase 1 given by Eq. (1). According to the Weiner–Khinchtine theorem,[10] a necessary and sufficient condition for the existence of a statistically homogeneous covariance function $\gamma(\mathbf{r})$ is that it has the spectral representation

$$\gamma(\mathbf{r}) = \frac{1}{(2\pi)^d} \int \tilde{\gamma}(\mathbf{k}) e^{i\mathbf{k}\cdot\mathbf{r}} d\mathbf{k}. \tag{22}$$

Relation (22) assumes that the Fourier transform or *spectral density* $\tilde{\gamma}(\mathbf{k})$ exists for all $\mathbf{k}$, which implies that $\gamma(\mathbf{r})$ is absolutely integrable, i.e., $\int|\gamma(\mathbf{r})|d\mathbf{r} < \infty$.[12] (Note that Debye *et al.*[8] showed that $\tilde{\gamma}(\mathbf{k})$ can be obtained directly for a porous medium via scattering of radiation.) Now we show that the spectral density of $\gamma(\mathbf{r})$ is positive, i.e.,

$$\tilde{\gamma}(\mathbf{k}) = \int \gamma(\mathbf{r}) e^{-i\mathbf{k}\cdot\mathbf{r}} d\mathbf{r} \geqslant 0. \tag{23}$$

To prove this positivity property, we use the definition (21) and the Fourier representations of $J(\mathbf{x})$ and $J(\mathbf{x}+\mathbf{r})$ in order to rewrite Eq. (23) as follows:

$$\begin{aligned} \tilde{\gamma}(\mathbf{k}) &= \int \left[\lim_{V\to\infty} \frac{1}{V} \int J(\mathbf{x}) J(\mathbf{x}+\mathbf{r}) d\mathbf{x}\right] e^{-i\mathbf{k}\cdot\mathbf{x}} d\mathbf{r} \\ &= \lim_{V\to\infty} \frac{1}{V} \int \tilde{J}(\mathbf{k}) J(\mathbf{x}) e^{i\mathbf{k}\cdot\mathbf{x}} d\mathbf{x} \\ &= \lim_{V\to\infty} \frac{1}{V} \tilde{J}(\mathbf{k}) \tilde{J}^*(\mathbf{k}) = \lim_{V\to\infty} \frac{1}{V} |\tilde{J}(\mathbf{k})|^2 \geqslant 0, \end{aligned} \tag{24}$$

where the complex conjugate of $\tilde{J}(\mathbf{k})$, denoted by $\tilde{J}^*(\mathbf{k})$, arises in the third line of Eq. (24) since $J(\mathbf{x})$ is real.

The existence of a positive spectral density $\tilde{\gamma}(\mathbf{k})$ implies the real-space asymptotic properties (9), the upper bound of Eq. (10), and the condition that the slope at the origin is negative as specified by Eq. (11). However, $\tilde{\gamma}(\mathbf{k}) \geqslant 0$ does not imply the lower bound of Eq. (10) (pointwise positivity of S_2).

The positivity property Eq. (23) holds for any wavenumber $\mathbf{k}$. In particular, it holds for $\mathbf{k} = \mathbf{0}$, i.e., the real-space volume integral of $\gamma(\mathbf{r})$ must be positive or

$$\int [S_2(\mathbf{r}) - \phi_1^2] d\mathbf{r} \geqslant 0. \tag{25}$$

The integral condition (25) holds for statistically homogeneous but anisotropic media. This positivity condition could also have been obtained immediately from the work of Lu and Torquato[13] on the so-called *coarseness* or standard deviation of the local volume fraction. In particular, it can be obtained from the asymptotic expression given for large window sizes and the fact that the coarseness is always positive.

If the medium is also statistically isotropic, then the two-point correlation function depends on the magnitude $r \equiv |\mathbf{r}|$ and Eq. (25) simplifies as

$$\int [S_2(r) - \phi_1^2] r^{d-1} dr \geqslant 0. \tag{26}$$

Here we have used the fact that $d\mathbf{r} = \Omega r^{d-1} dr$ in a d-dimensional spherical coordinate system, where Ω is the positive d-dimensional solid angle. If we let

$$\langle r^n \rangle = \int_0^\infty [S_2(r) - \phi_1^2] r^n dr, \tag{27}$$

denote the nth moment of the scalar $S_2(r) - \phi_1^2$, then Eq. (26) states that the moment $\langle r^{d-1} \rangle \geqslant 0$ for isotropic two-phase random media in d spatial dimensions.

Furthermore, we now show that all lower-order positive moments $(\langle r^0 \rangle, \langle r^1 \rangle, \cdots, \langle r^{d-2} \rangle)$ must also be positive for isotropic media. For concreteness, consider first the case $d = 3$. From Eq. (26), we have that the second moment $\langle r^2 \rangle$ must be positive; however, we also show that $\langle r^1 \rangle \geqslant 0$ and $\langle r^0 \rangle \geqslant 0$. We have already noted the well-known fact that $S_2(r)$ can be determined from a planar cut through the three-dimensional isotropic random medium. Therefore, one can create a two-dimensional random medium from this planar cut and since the two-point function $S_2(r)$ remains invariant, then condition (26) applies with $d = 2$, i.e., $\langle r^1 \rangle \geqslant 0$. (Interestingly, this first moment condition can also be obtained from rigorous bounds on the fluid permeability[14] and trapping constant[15] of three-dimensional isotropic media and the fact that these transport properties must be positive.) Similarly, since a one-dimensional random medium can be produced from a lineal cut through the three-dimensional medium and $S_2(r)$ again remains invariant, then condition (26) applies with $d=1$, i.e., $\langle r^0\rangle \geqslant 0$. To summarize, for $d=3$, the zeroth, first, and second moments of $S_2(r)-\phi_1^2$ must be positive.

In general, for d-dimensional isotropic media, $S_2(r)$ can be extracted from a cut of the d-dimensional medium with an m-dimensional subspace $(m=1,2,\ldots,d-1)$ and therefore formula (26) also applies for $d=m$ for any m. Thus, we have the following d positivity conditions for d-dimensional isotropic media:

$$\langle r^n\rangle \geqslant 0, \quad n=0,1,\cdots,d-1. \tag{28}$$

We see that for two-dimensional isotropic random media, the zeroth and first moments must be positive, whereas for one-dimensional media, only the zeroth moment need be positive.

For exactly the same reasons, a statement corresponding to Eq. (28) can be made for the entire spectral density $\tilde{\gamma}(k)$, i.e., the one-, two-, $\cdots$ and d-dimensional Fourier transforms of $\gamma(r)$ must all be positive for d-dimensional isotropic media. Therefore, whenever we refer to the positivity condition (23) in the case of isotropic media, it will be implicit that we mean all s-dimensional Fourier transforms, $s=1,2,\ldots,d$.

The real-space integral conditions (25) and (28) are special cases of the more general integral condition (23) and thus the former are necessary but not sufficient conditions that physically realizable correlation functions must meet. In the next section, we apply the integral conditions to test various classes of correlation functions.

IV. APPLICATIONS OF INTEGRAL CONDITIONS ON S_2

Any physically realizable two-point correlation function $S_2(\mathbf{r})$ of statistically homogeneous media must be pointwise positive and satisfy the positivity condition (23). The zero-wavenumber integral conditions (25) and (28) may first be checked as they are easier to compute than the full Fourier transform. Moreover, if they are negative, then there is no need to compute the Fourier transform.

In the ensuing discussion, we will consider several hypothetical *isotropic* correlation functions that have been proposed for use in random-media construction algorithms.[4,5] All of these functions satisfy the conditions (9), (10) and (11). We will check to see if they obey the positivity conditions (23) and (28) in various spatial dimensions.

Using simple probabilistic arguments, Debye, Andersen, and Brumberger[8] showed that the two-point correlation function of three-dimensional isotropic porous media consisting of voids of ''random shape and size'' is given by

$$\gamma(r)=S_2(r)-\phi_1^2=\phi_1\phi_2 e^{-r/a}, \tag{29}$$

where a is a *positive* correlation length. Recently, Yeong and Torquato[4] found a realization of a two-dimensional microstructure that possesses the correlation function (29). They referred to such a system as a *Debye random medium*. It is easily verified (by direct calculation) that all of the positive moments of $\gamma(r)$ are positive, indicating that a Debye random medium may be realizable in any dimension. Indeed, a Debye random medium is realizable for any d, since the general positivity condition (23) is obeyed for any d.

More interesting and potentially problematic behavior arises when one wants to introduce short-range order into the correlation function. An example of such a function, proposed by Yeong and Torquato,[4] has the form[16]

$$\gamma(r)=S_2(r)-\phi_1^2=\phi_1\phi_2 e^{-r/a}\cos(qr), \tag{30}$$

where q is a parameter that controls the short-range order. Observe that such an S_2 is positive [lower bound of Eq. (10)] provided that $e^{-r_0/a}\cos(qr_0)\geqslant -\phi_1/\phi_2$, where r_0 is the first positive root of the equation $\tan(qr)-(qa)^{-1}=0$ that specifies the first minimum.

Now let us examine the positivity conditions for Eq. (30). First, we see that the zeroth moment is always positive since

$$\langle r^0\rangle = \frac{a}{1+q^2a^2}\geqslant 0. \tag{31}$$

Moreover, the one-dimensional Fourier transform is given by

$$\tilde{\gamma}(k) = \frac{2a[1+(q^2+k^2)a^2]}{[1+(q^2+k^2)a^2-2qka^2][1+(q^2+k^2)a^2+2qka^2]}. \tag{32}$$

Now since $q^2+k^2\geqslant 2qk$, then $\tilde{\gamma}(k)$ is also positive for any a and q. Therefore, we conclude that there are realizable one-dimensional random media that have the correlation function given by Eq. (30), provided that $S_2(r)$ is positive.

The first moment of Eq. (30) is given by

$$\langle r^1\rangle = \frac{a^2(1-q^2a^2)}{(1+q^2a^2)^2}, \tag{33}$$

and therefore is positive provided that $(qa)^2\leqslant 1$. This condition is clearly more restrictive than the zeroth-moment condition (31), which applies as well to both $d=2$ and $d=3$. Thus, we immediately ascertain that random media having the correlation function (30) cannot be realized in *both two and three dimensions* for $(qa)^2\geqslant 1$. The fact that $\langle r^1\rangle \geqslant 0$ if $(qa)^2\leqslant 1$ is insufficient to ensure that there are not values of $(qa)^2$ smaller than one that are impermissible. To check this possibility, one must compute the Fourier transform. A numerical evaluation of the first integral (19) with $f=\gamma$ reveals that $\tilde{\gamma}(k)$ is positive for all k if $(qa)^2\geqslant 1$, i.e., the first-moment condition is sufficient in this case. Thus, there are realizable two-dimensional random media that have the correlation function given by Eq. (30), provided that $S_2(r)$ is positive and $(qa)^2\leqslant 1$.

The analysis given above for the function (30) can be used to explain the results of two numerical construction experiments of digitized media carried out in two dimensions. Yeong and Torquato[4] found two-dimensional realizations of random media having the function (30) with the parameters $\phi_1=\phi_2=0.5$, $a=8$ pixels, and $q=1$ (pixel)$^{-1}$. It is important to emphasize that they sampled for S_2 only in two orthogonal directions. Cule and Torquato[5] and Manwart and Hilfer[6] independently repeated the aforementioned construction but they sampled for S_2 in all directions and found that they could not realize Eq. (30) for all values of r. The reasons for these seemingly contradictory results is now clear. In the first experiment,[4] orthogonal sampling made the problem effectively one-dimensional and, as we have seen, for $d=1$, the function (30) with the above parameters is realizable. In the second set of experiments,[5,6] sampling was truly two-dimensional and for $d=2$ the function (30) with the above parameters is *not* realizable.

The second moment of Eq. (30) is given by

$$\langle r^2\rangle=\frac{2a^3(1-3q^2a^2)}{(1+q^2a^2)^3}, \tag{34}$$

and therefore is positive provided that $(qa)^2\leqslant 1/3$. Therefore, this condition for three-dimensional media is more restrictive than Eq. (33), which applies to $d=3$ as well. The three-dimensional Fourier transform of Eq. (30) is given by

$$\tilde{\gamma}(k)=\frac{8\pi a^3[1+2k^2a^2+2k^4a^4+2q^2k^2a^4-2q^2a^2-3q^4a^4]}{[1+(q^2+k^2)a^2-2qka^2]^2[1+(q^2+k^2)a^2+2qka^2]^2}. \tag{35}$$

It is seen that $\tilde{\gamma}(k)\geqslant 0$ for all k provided that the second moment condition is satisfied. We conclude that there are realizable three-dimensional random media that have the correlation function given by Eq. (30), provided that $S_2(r)$ is positive and $(qa)^2\leqslant 1/3$.

Another example of a correlation function that possesses short-range order is

$$\gamma(r)=S_2(r)-\phi_1^2=\phi_1\phi_2 e^{-r/a}\frac{\sin(qr)}{qr}. \tag{36}$$

This function was proposed by Cule and Torquato[5] to mimic random media comprised of nonoverlapping particles. The autocorrelation $S_2(r)$ of Eq. (36) is positive provided that $e^{-r_0/a}\sin(qr_0)\geqslant -\phi_1/\phi_2$, where r_0 is the first positive root of the equation $\cot(qr)-(qr)^{-1}-(qa)^{-1}=0$ that specifies the first minimum.

The zeroth, first, and second moments of the covariance Eq. (36) are positive for all a and q. Indeed, since the one-, two-, and three-dimensional spectral densities are also positive, we can conclude that there are realizable one-, two-, and three-dimensional random media that have the correlation function given by Eq. (36), provided that $S_2(r)$ is positive.

V. EXACT INTEGRAL CONDITIONS ON S_3

Integral conditions on higher-order correlation functions can be found by generalizing the aforementioned spectral-representation procedure. This will be the subject of a future article. Here we note in passing integral conditions on the three-point function $S_3(r,s,t)$ of isotropic media that are already known in the study of the effective properties of random media.

Certain rigorous estimates of the effective conductivity of isotropic two-phase composites depend on a functional ζ of the three-point function $S_3(r,s,t)$ defined below.[17–20] This functional is positive and bounded from above by unity, i.e.,

$$0\leqslant\zeta\leqslant 1. \tag{37}$$

In any dimension $d\geqslant 2$, ζ is given by

$$\zeta=\frac{d^2}{(d-1)\phi_1\phi_2\Omega^2}\int\int\frac{d\mathbf{r}}{r^d}\frac{d\mathbf{s}}{s^d}[d(\mathbf{n}\cdot\mathbf{m})^2-1]\times\left[S_3(r,s,t)-\frac{S_2(r)S_2(s)}{\phi_1}\right], \tag{38}$$

where $\mathbf{n}=\mathbf{r}/|\mathbf{r}|$ and $\mathbf{m}=\mathbf{s}/|\mathbf{s}|$ are unit vectors and Ω is the total solid angle contained in a d-dimensional sphere. We see that for $d=2$

$$\zeta=\frac{4}{\pi\phi_1\phi_2}\int_0^\infty\frac{dr}{r}\int_0^\infty\frac{ds}{s}\int_0^\pi d\theta\cos(2\theta)\times\left[S_3(r,s,\theta)-\frac{S_2(r)S_2(s)}{\phi_1}\right], \tag{39}$$

and for $d=3$

$$\zeta=\frac{9}{2\phi_1\phi_2}\int_0^\infty\frac{dr}{r}\int_0^\infty\frac{ds}{s}\int_{-1}^1 d(\cos\theta)P_2(\cos\theta)\times\left[S_3(r,s,\theta)-\frac{S_2(r)S_2(s)}{\phi_1}\right], \tag{40}$$

where P_2 is the Legendre polynomial of order 2 and θ is the angle opposite the side of the triangle of length t.

Moreover, certain rigorous estimates of the effective shear modulus of isotropic two-phase composites depend on a functional η of the three-point function $S_3(r,s,t)$ defined below.[18–20] This functional is also positive and bounded from above by unity, i.e.,

$$0\leqslant\eta\leqslant 1. \tag{41}$$

In any dimension $d\geqslant 2$, η is given by

$$\eta=-\frac{(d+2)(5d+6)}{d^2}\zeta+\frac{(d+2)^2}{(d-1)\phi_1\phi_2\Omega^2}\times\int\int\frac{d\mathbf{r}}{r^d}\frac{d\mathbf{s}}{s^d}[d(d+2)(\mathbf{n}\cdot\mathbf{m})^4-3]\times\left[S_3(r,s,t)-\frac{S_2(r)S_2(s)}{\phi_1}\right]. \tag{42}$$

For $d=2$, Eq. (42) reduces to

$$\eta=\frac{16}{\pi\phi_1\phi_1}\int_0^\infty\frac{dr}{r}\int_0^\infty\frac{ds}{s}\int_0^\pi d\theta\cos(4\theta)\times\left[S_3(r,s,t)-\frac{S_2(r)S_2(s)}{\phi_1}\right], \tag{43}$$

and for $d=3$, Eq. (42) reduces to

$$\eta=\frac{5\zeta_2}{21}+\frac{150}{7\phi_1\phi_2}\int_0^\infty\frac{dr}{r}\int_0^\infty\frac{ds}{s}\int_{-1}^1 d(\cos\theta)P_4(\cos\theta)\times\left[S_3(r,s,t)-\frac{S_2(r)S_2(s)}{\phi_1}\right], \tag{44}$$

where P_4 is the Legendre polynomial of order four.

The asymptotic properties Eqs. (13) and (14) on S_3, the pointwise bounds (10), and the integral constraints (37) and (41) constitute the known exact conditions on S_3. The forthcoming analysis mentioned above will yield more general integral conditions on S_3.

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