Light scattering in ceramics

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Abstract

A framework for systematically treating various aspects of optical propagation in ceramics and similar composite materials. The point of view is a compromise which attempts to deal with some of the difficulties of a completely rigorous treatment of the subject while providing a convenient means for evaluating the optical properties of real materials and designing processing strategies for new materials.

Introduction

For a variety of practical reasons, there is increasing use of ceramic materials in optical applications. In fabricating or evaluating such materials for particular applications it is helpful to have a some means of predicting how a particular stoichiometric or microstructural characteristic influences a given optical property. Some time ago [1], we employed an extremely simple scalar wave model to estimate the relative importance of porosity and grain size in determining the transparency of ceramics. Recently, Schroeder and Rosolowski [2] have made effective use of this model in an extensive study of diffuse scattering. The particular value of our model lies in the use of simple "doubly random telegraph wave" statistics as a means of independently parameterizing the effects of ceramic density and consituent particle size. However, it is a scalar theory and gives no insight into important electromagnetic characteristics such as birefringence, depolarization, and finite geometry effects. We attempt here to broaden the utility of the original model by developing it within the context of an electromagnetic framework. We caution the reader that the goal is a self-contained and, hopefully, reliable formulation of the problem not an exhausive review of the relevant theoretical and experimental literature.

Over the last twenty years or so a great deal has been written about the propagation of light in randomly inhomogeneous media, yet the problem remains a difficult one. Most of the work treats scalar wave propagation and relates to problems in atmospheric propagation. Condensed composite materials, such as ceramics, have received little specific attention. Rigorous formulations of the subject, in most instances, are essentially multiple scattering calculations which lead to an infinite hierachy of equations for the statistical moments of the wave field in terms of multi-point correlations of the local scattering strength of the medium. To make any sensible use of these elegant formulations one must be able to rationalize some way of truncating the infinite hierarchy. Long ago, Keller [3], calling it the "dishonest" method, and Bourret [4,5] calling it "local independence", provided arguments for a truncation which makes the most effective use of two-point correlations. Tatarskii [6], building on the work of Bourret, developed an electromagnetic multiple scattering formulation which has been the guiding inspiration for the work presented here. We begin with a vector formulation of electromagnet scattering and a discussion of some of the difficulties inherent in a general, rigorous solution to the problem. Then, for some practically useful cases, we exhibit solutions for the mean coherent field and mean coherence matrix in terms of general two-point correlation functions associated with scattering by polarizability fluctuations from a self-consistent mean. Finally, we show how, with suitable stereological interpretation, simple exponential correlation functions usefully approximate important aspects of ceramic microstructure.

Random scattering of a vector field

Our task here is to establish an electromagnetic model of scattering in a ceramic medium. Because of the high density of scattering centers in ceramics our essential problem is to cope with fluctuations in a hugh background of multiple scattering. A useful model must be some kind of "mean field" approach wherein the background is normalized out and scattering is associated with deviations from a mean which best approximates the homogeneous properties of the real ceramic. However, there are several possible ways of approaching this goal each of which involves cumbersome mathematical details. In order to best delineate the options and to avoid becoming entangled in details, it is useful to first outline the "mean field" method in somewhat abstract terms. To this end we set down a compact, symbolic representation of a general scattering process and exhibit formal solutions for the mean coherent field and mean coherence matrix.

We start with a general, symbolic self-consistent scattering equation for a monochromatic, vector field $\bar{F}(1)$ at the observation point (1) which arises from an elastic scattering process at the source points (2)

$$\bar{F}(1) = \bar{F}^{o}(1) + \bar{\bar{\Gamma}}^{o}(12)\cdot\bar{\bar{\chi}}(2)\cdot\bar{F}(2) \tag{1}$$

where $\bar{F}^{o}(1)$ is the field in the absence of the scattering process, $\bar{\bar{\chi}}(2)$ is the dyadic local scattering strength, and $\bar{\bar{\Gamma}}^{o}(12)$ is a dyadic integro-differential operator which symbolizes the propagation of the field from point (2) to point (1). Later we discuss in considerable detail, possible specific forms for this propagator (see Table 1). By iterating Equation 1 an infinite number of times we may write a formal, symbolic solution for the field as

$$\bar{F}(1) = \bar{\bar{T}}(12)\cdot\bar{F}^{o}(2) \tag{2}$$

where

$$\bar{\bar{T}}(12) = \left(\bar{\bar{I}}(12) - \bar{\bar{\Gamma}}^{o}(12)\cdot\bar{\bar{\chi}}(2)\right)^{-1} \tag{3a}$$

$$= \bar{\bar{I}}(12) + \bar{\bar{\Gamma}}^{o}(13)\cdot\bar{\bar{\chi}}(3)\cdot\bar{\bar{T}}(32) \tag{3b}$$

To establish a means for normalizing out the effects of the background scattering we divide $\bar{\bar{\chi}}(2)$ into a homogeneous part $\bar{\bar{\chi}}^{h}$ and an inhomogeneous part $\Delta\bar{\bar{\chi}}(2)$. With such a division Equation 3 may be recast in the form

$$\bar{F}(1) = \bar{F}^{h}(1) + \bar{\bar{\Gamma}}^{h}(12)\cdot\Delta\bar{\bar{\chi}}(2)\cdot\bar{F}(2) \tag{4a}$$

$$\bar{\bar{T}}(12) = \bar{\bar{T}}^{h}(12) + \bar{\bar{\Gamma}}^{h}(13)\cdot\Delta\bar{\bar{\chi}}(3)\cdot\bar{\bar{T}}(32) \tag{4b}$$

where

$$\bar{\bar{T}}^{h}(12) = \left(\bar{\bar{I}}(12) - \bar{\bar{\Gamma}}^{o}(12)\cdot\bar{\bar{\chi}}^{h}\right)^{-1} \tag{5}$$

determines the solution to the homogeneous problem and

$$\bar{\bar{\Gamma}}^{h}(13) = \bar{\bar{T}}^{h}(14)\cdot\bar{\bar{\Gamma}}^{o}(43) \tag{6}$$

is the propagator associated with the homogeneous medium.

These formal solutions to the deterministic problem may be used to generate formal solutions of the stochastic problem. Solutions are expressed as a hierarchy of equations for ensemble averages of products of the scattered field in terms of coupled moments of products of the field and the local scattering strength. For example, the first two equations of the hierarchy are

$$\langle\bar{F}(1)\rangle = \bar{F}^{h}(1) + \bar{\bar{\Gamma}}^{h}(12)\cdot\langle\Delta\bar{\bar{\chi}}(2)\cdot\bar{F}(2)\rangle \tag{7a}$$

$$\langle\Delta\bar{\bar{\chi}}(1)\cdot\bar{F}(1)\rangle = \langle\Delta\bar{\bar{\chi}}(1)\rangle\cdot\bar{F}^{h}(1) + \langle\Delta\bar{\bar{\chi}}(1)\cdot\bar{\bar{\Gamma}}^{h}(12)\cdot\Delta\bar{\bar{\chi}}(2)\cdot\bar{F}(2)\rangle \tag{7b}$$

Since ceramic microstructure is exceedingly complex and, generally, poorly characterized, it is pointless to push the statistical analysis beyond a decent limit. Yet, we would like to be able to model at least the gross microstructural features with some fidelity. We compromise by truncating the hierarchy of moment equations in a way which seems to make the most effective use of two-point correlation functions. To accomplish this, we follow Bourret[4,5] and decouple the the moment equations by assuming that mean values of the moments of the field are "locally independent" of the mean values of the moments of the scattering strength. This physically reasonable assumption has been shown by diagrammatic techniques to be equivalent to a complete summation of the first order "strongly connected" diagrams. If we take $\langle\Delta\bar{\bar{\chi}}\rangle = 0$ (an important and by no means trivial step in the development), we may write, in the Bourret approximation, a "Dyson equation" for the mean coherent field as

$$\langle\bar{F}(1)\rangle = \bar{F}^{h}(1) + \bar{\bar{\Gamma}}^{h}(13)\bar{\bar{\Gamma}}^{h}(32)\cdot\underline{\underline{\bar{\bar{B}}}}(32)\cdot\langle\bar{F}(2)\rangle \tag{8a}$$

$$\langle\bar{\bar{T}}(12)\rangle = \bar{\bar{T}}^{h}(12) + \bar{\bar{\Gamma}}^{h}(14)\bar{\bar{\Gamma}}^{h}(43)\cdot\underline{\underline{\bar{\bar{B}}}}(43)\cdot\langle\bar{\bar{T}}(32)\rangle \tag{8b}$$

Here the natural products of two dyadic operators are usefully represented as "tetradic" operators where, in particular, the tetradic

$$\underline{\underline{\bar{\bar{B}}}}(21) = \langle\Delta\bar{\bar{\chi}}(2)\Delta\bar{\bar{\chi}}(1)\rangle \tag{9}$$

symbolizes the two-point correlations of the dyadic scattering strength. For application here and later, we define in coordinate notation two different types of tetradic products - viz.

$$(\bar{\bar{U}}\bar{\bar{V}}\cdot\bar{\bar{W}})_{im} = (\bar{\bar{U}})_{ij}(\bar{\bar{V}})_{kl}(\bar{\bar{W}})_{jklm} \quad (10a)$$

$$(\bar{\bar{U}}\bar{\bar{V}}:\bar{\bar{W}})_{ijmn} = (\bar{\bar{U}})_{ik}(\bar{\bar{V}})_{jl}(\bar{\bar{W}})_{kmln} \quad (10b)$$

where summation over repeated indices is implied. Continuing in the spirit of the "local independence" assumption we may write the "Bourret-ladder"[6] approximation to the "Bethe-Saltpeter equation" for the coherence matrix or second moment of the field as

$$\langle\bar{F}(1)\bar{F}^*(2)\rangle = \langle\bar{F}(1)\rangle\langle\bar{F}^*(2)\rangle + \langle\bar{\bar{T}}(13)\rangle\langle\bar{\bar{T}}^*(24)\rangle:\bar{\bar{B}}'(34):\langle\bar{F}(3)\bar{F}^*(4)\rangle \quad (11)$$

where

$$\bar{\bar{B}}'(34) = \langle\Delta\bar{\bar{X}}(3)\Delta\bar{\bar{X}}^*(4)\rangle \quad (12)$$

Random scattering of electromagnetic field

Building on the abstact overview of the previous section, let us now explore specific representations and applications. In what is, perhaps, the most precise application of this operator formalism, we start with vacuum and interpret Equation 1 as the self-consistent equation for the dipolar field associated with an induced polarization density $\bar{P}(2)$ - assumming that $\bar{F}^h(1) = \bar{E}^v(1)$ is the field in the absence of the polarizable medium and due to remote sources. However, in interpreting the symbols we must distinquish two distinct possibilities which are summarized in the following table (along with a scalar wave interpretation of Equation 1).

Table 1. Interpretations of Equation 1

Version	Scalar	Vector(S)	Vector(NS)
$\bar{F}$	ψ	$\bar{E}' = \bar{\bar{\alpha}}^{-1}\cdot\bar{P}$	$\bar{E}'' = \bar{\bar{\beta}}^{-1}\cdot\bar{P}$
$\bar{\bar{X}}$	$(n^2 - 1)/4\pi$	$\bar{\bar{\alpha}}$	$\bar{\bar{\beta}} = \bar{\bar{\alpha}}\cdot\left(\bar{\bar{1}} - 4\pi/3\ \bar{\bar{\alpha}}\right)^{-1}$
$\bar{\bar{\Gamma}}^o$	Γ^v	$\bar{\bar{F}}^v$	$\bar{\bar{\Gamma}}^v$

In the version labelled "S", $\bar{E}'(1)$ is the self-consistent local "effective polarizing field", $\bar{\bar{\alpha}}(2)$ is the local, microscopic dyadic polarizability, and

$$\bar{\bar{F}}^v(12) = ⨍ \bar{\bar{G}}^v(12)\ dV_2 = ⨍ \{q_v^{-2}\ \bar{\nabla}_1\bar{\nabla}_1 + \bar{\bar{1}}\}\ G^v(12)\ dV_2 \quad (13)$$

with

$$G^v(12) = q_v^2\ \exp(iq_v\ |r_1 - r_2|\ )/\ |r_1 - r_2| \quad (14)$$

is the vacuum dyadic propagator with $q_v = \omega/c$. The integral sign and dyadic propagator symbol is cross-hatched to remind us that the kernel of the integral is singular when source and field points coincide so that the integration must exclude a small volume centered on the singularity. Coping with this singularity has been the source of much confusion in electromagnetic problems, but the "famous lemma" of Born and Wolf[7] is extremely useful in many instances and in this case leads to the alternative interpretation which is labelled as "NS" in Table 1. The lemma may be stated

$$\bar{\bar{F}}^v(12) = \bar{\bar{\Gamma}}^v(12) + (4\pi/3)\ \bar{\bar{1}}(12) \quad (15)$$

where $\bar{\bar{1}}(12)$ is the unit Dirac delta dyadic and

$$\bar{\bar{\Gamma}}^v(12) = \{q_v^{-2}\ \bar{\nabla}_1\bar{\nabla}_1 + \bar{\bar{1}}\}\ \Gamma^v(12) = \{q_v^{-2}\ \bar{\nabla}_1\bar{\nabla}_1 + \bar{\bar{1}}\}\int G^v(12)\ dV_2 \quad (16)$$

is the vacuum propagator with a non-singular kernel.

In spite of the more direct physical insight afforded by interpretation S, Born and Wolf instruct us in the computational utility of interpretation NS. For our purposes, the important point is that NS allows us to use normal mode expansions without equivocation. If $\bar{\bar{\alpha}}(2)$ is divided into a homogeneous and inhomogeneous part, the key to the analysis is expansion of Equation 5 in terms of an appropriate set of normal modes. Specifically, the plane wave expansion may be written

$$\bar{\bar{t}}^{h}(\bar{q}) = \bar{\bar{1}} + \bar{\bar{\gamma}}^{v}(\bar{q})\cdot\bar{\bar{\beta}}^{h}\cdot\bar{\bar{t}}^{h}(\bar{q}) \tag{17}$$

where the lower case symbols denote the plane wave amplitudes of the corresponding upper case spatial representations. Two important (and very nearly obvious) points emerge from an arduous, but straight forward solution of Equation 17 - viz.

1. The microscopic wave scattering view, embodied in Equation 1, is consistent with the more traditional macroscopic picture of wave propagation in an anisotropic medium, if we interpret $\bar{\bar{1}} + 4\pi\bar{\bar{\beta}}$ as the macroscopic dielectric dyadic $\bar{\bar{\epsilon}}$. Thus, a dyadic generalization of the Lorentz-Lorenz relation[8]

$$(4\pi/3)\bar{\bar{\alpha}} = \left(\bar{\bar{\epsilon}} - \bar{\bar{1}}\right)\cdot\left(\bar{\bar{\epsilon}} + 2\,\bar{\bar{1}}\right)^{-1} \tag{18}$$

is the the critical link between micro-and macroscopic view points.

2. The dyadic propagator $\bar{\bar{\Gamma}}^{h}(12)$, in Equation 6, is rigorously the propagator for the macroscopic electric field of the macroscopic Maxwell equations specified by Equation 18.

Equations 4-6 then represent a framework consistent with macroscopic Maxwell equation. The enormous background of multiple scattering at the microscopic level is accounted for in the macroscopic field which may be determined quite precisely by boundary value methods. However, in making a macroscopic interpretation of these equations, we find, once again, that the symbols may be given two distinct interpretations which are summarized in the following table.

Table 2. Interpretations of Equations 4

Version	Macroscopic(NS)	Macroscopic(S)
$\bar{\bar{F}}$	$\bar{\bar{E}}$	$(\bar{\bar{\alpha}}^{eff})^{-1}\cdot(\bar{\bar{P}} - \bar{\bar{P}}^{h})$
$\bar{\bar{F}}^{h}$	$\bar{\bar{E}}^{h}$	$\bar{\bar{D}}^{h}$
$\Delta\bar{\bar{\chi}}$	$(\bar{\bar{\epsilon}} - \bar{\bar{\epsilon}}^{h})/4\pi$	$\bar{\bar{\alpha}}^{eff}$
$\bar{\bar{\Gamma}}^{h}$	$\bar{\bar{\Gamma}}^{h}$	$\bar{\bar{\Gamma}}^{h} = \bar{\bar{\epsilon}}^{h}\cdot\bar{\bar{\Gamma}}^{h} + 4\pi/3\;\bar{\bar{1}}$

The "NS" interpretation is, potentially, the most widely applicable since, as mentioned earier, it uses the propagator appropriate to a given specification of homogeneous background and boundary conditions which, in principle, can always be expressed in terms of the normal modes of the homogeneous problem. In the NS version one can directly use well established solutions of macroscopic Maxwell equations for anisotropic materials[9] and finite samples[10,11]. The "S" interpretation is based on an obvious extension of Equation 15 and gives, perhaps, more direct physical insight by associating the excess scattering with a "local effective polarizability dyadic" of the form

$$(4\pi/3)\bar{\bar{\alpha}}^{eff} = \left(\bar{\bar{\epsilon}} - \bar{\bar{\epsilon}}^{h}\right)\cdot\left(\bar{\bar{\epsilon}} + 2\,\bar{\bar{\epsilon}}^{h}\right)^{-1} \tag{19}$$

Such a polarizability would be appropriate to a tiny particle of material with a dielectric dyadic $\bar{\bar{\epsilon}}$ embedded in a matrix characterized by a dielectric dyadic $\bar{\bar{\epsilon}}^{h}$. The choice between these interpretations is largely a matter of taste, but it is crucial to keep various aspects of the interpretation consistent. In order to make contact with earlier work[6,12] we follow version S of Table 2 in the remainder of this paper.

We turn now to some specific results for stochastically inhomogeneous materials. If we can be content with neglecting microstructural details we need not proceed further since the dyadic generalization of the Bruggeman relation[13]

$$\langle\bar{\bar{\alpha}}^{eff}(\bar{r})\rangle \equiv 0 \tag{20}$$

is an accurate specification of a "mean effective medium" in which excess scattering is minimized. The scalar version has been use with some success in treating, for example, the reflectivity of "cermet" materials[14]. Equation 20, of course, also defines the best homogeneous background for the calculation of microstructural effects, but it defines in the general case an anisotropic background and we are required to use propagators appropriate to anisotropic media. Although feasible, using such propagators would greatly complicate an already complicated problem and would, in fact, be inconsistent with interpretation S. As a compromise we use the "diagonal sum" of Equation 20 to define a scalar ϵ^h. The appropriate propagator then is the solution for an isotropic medium and has the form of $\mathcal{F}$ in Equation 13 with the subscript "v" replaced everywhere by "h" with $q_h = \epsilon^h\omega/c$. Using this propagator, we may write a plane wave expansion of Equation 8b in the form

$$\langle\bar{\bar{t}}(\bar{q})\rangle = \bar{\bar{t}}^h(\bar{q}) + \bar{\bar{\gamma}}^h(\bar{q})\cdot\bar{\bar{\theta}}(\bar{q})\cdot\langle\bar{\bar{t}}(\bar{q})\rangle \tag{21}$$

where

$$\bar{\bar{\gamma}}^h(\bar{q}) = \int \bar{\bar{G}}^h(\bar{R}) \exp(i\bar{q}\cdot\bar{R})\, dR^3 \tag{22}$$

and

$$\bar{\bar{\theta}}(\bar{q}) = \int \bar{\bar{G}}^h(\bar{R})\cdot\underline{\bar{\bar{B}}}(\bar{R}) \exp(i\bar{q}\cdot\bar{R})\, dR^3 \tag{23}$$

Stationary random statistics have been assumed so that $\underline{\bar{\bar{B}}}$ is taken to be a function of coordinate differences only. Since Equation 17 and 21 are identical in form, we can draw on previous analysis and immediately set down an important and useful result. Namely, we find that

$$\bar{\bar{\epsilon}}^{eff}(\bar{q}) = \epsilon^h\left(\bar{\bar{1}} + 4\pi\,\bar{\bar{\theta}}(\bar{q})\right) \tag{24}$$

effectively represents the dielectric dyadic to be associated with plane waves of wave vector $\bar{q}$. For example, in complete analogy with homogeneous anisotropic media, the two propagation modes associated with given a wave vector have polarizations and indices of refraction determined by

$$\hat{u}^{(i)}(\bar{q})\cdot\left(\bar{\bar{\epsilon}}^{eff}(\bar{q})\right)^{-1}\cdot\hat{u}^{(j)}(\bar{q}) = \left(n^{(i)}(\bar{q})\right)^{-2}\delta_{ij} \tag{25}$$

where $n^{(i)}(\bar{q})$ is the index of the mode and $\hat{u}^{(i)}(\bar{q})$ is a unit vector orthogonal to $\bar{q}$ which defines the polarization of the dielectric displacement of the mode. If $\underline{\bar{\bar{B}}}$ is a function of $|\bar{R}|$ alone, Equation 23 may be cast in a computationally useful form - viz.

$$\bar{\bar{\theta}}(\bar{q}) = \bar{\bar{1}}\cdot\underline{\bar{\bar{\sigma}}}(q) + (\bar{\bar{1}} - 3\,\hat{q}\hat{q})\cdot\underline{\bar{\bar{\tau}}}(q) \tag{26}$$

with

$$\underline{\bar{\bar{\sigma}}}(q) = i(8\pi/3)q_h^3\int_0^\infty j_0(qR)\, h_0^{(1)}(q_hR)\, \underline{\bar{\bar{B}}}(R)\, R^2\, dR$$
$$\approx (8\pi/3)q_h^2\int_0^\infty \underline{\bar{\bar{B}}}(R)\, R\, dR \qquad \text{for q small} \tag{27a}$$

$$\underline{\bar{\bar{\tau}}}(q) = i(4\pi/3)q_h^3\int_0^\infty j_2(qR)\, h_2^{(1)}(q_hR)\, \underline{\bar{\bar{B}}}(R)\, R^2\, dR \tag{27b}$$
$$\sim (4\pi/15)q^2\int_0^\infty \underline{\bar{\bar{B}}}(R)\, R\, dR \qquad \text{for q small}$$

The functions $j_n(x)$ and $h_n^{(1)}(x)$ are respectively the spherical Bessel and Hankel functions of the first kind[17]. The requisite number of independent components of $\underline{\bar{\bar{B}}}$ are set by the global symmetry of the composite. In particular, the $\underline{\bar{\bar{B}}}$ of an isotropic medium, like the elastic tetradic, has two independent components.

We have shown in the last paragraph that behavior of the mean coherent field can be understood with considerable accuracy by evaluating and interpreting $\bar{\bar{\theta}}(\bar{q})$. We examine now, briefly, an interesting means of studying the behavior of the coherence matrix in random media. The coherence matrix is, of course, a much more difficult problem, but it is an extremely useful in characterizing imaging and polarization[15] properties of propagation. Recently, it has been demonstrated that the Wigner distribution function (WDF) is a valuable tool in studying optical systems[16]. Indeed, we find the WDF representation of Equation 11 for the coherence matrix yields a valuable formulation of imaging problems. If we define the WDF's

$$\bar{\bar{W}}(\bar{r},\bar{q}) = (2\pi)^{-3}\int\langle\bar{f}(\bar{q}+\bar{Q}/2)\bar{f}^*(\bar{q}-\bar{Q}/2)\rangle \exp(i\bar{Q}\cdot\bar{r})\, dQ^3 \tag{28a}$$

$$\bar{\bar{W}}^o(\bar{r},\bar{q}) = (2\pi)^{-3}\int\langle\bar{f}(\bar{q}+\bar{Q}/2)\rangle\langle\bar{f}^*(\bar{q}-\bar{Q}/2)\rangle \exp(i\bar{Q}\cdot\bar{r})\, dQ^3 \tag{28b}$$

$$\underline{\bar{\bar{K}}}(\bar{r},\bar{q}) = (2\pi)^{-3}\int\langle\bar{\bar{t}}(\bar{q}+\bar{Q}/2)\rangle\langle\bar{\bar{t}}^*(\bar{q}-\bar{Q}/2)\rangle \exp(i\bar{Q}\cdot\bar{r})\, dQ^3 \tag{28c}$$

Equation 11 is transformed to

$$\bar{\bar{W}}(\bar{r},\bar{q}) = \bar{\bar{W}}^{o}(\bar{r},\bar{q}) + \iint \underline{\underline{\bar{\bar{K}}}}(\bar{r}-\bar{R},\bar{q}) : \underline{\underline{\bar{\bar{b}}}}'(\bar{q}-\bar{Q}) : \bar{\bar{W}}(\bar{R},\bar{Q})\ dR^3 dQ^3 \tag{29}$$

where

$$\underline{\underline{\bar{\bar{b}}}}'(\bar{Q}) = \int \underline{\underline{\bar{\bar{B}}}}'(\bar{\rho})\ \exp(i\bar{Q}\cdot\bar{\rho})\ d\rho^3 \tag{30}$$

$\bar{\bar{W}}(\bar{r},\bar{q})$ may be thought of as a "local" or spatially varying plane wave spectrum or, in terms of geometric optics, as a distribution of ray vectors. Equation 29 is a kind of "transport equation" for the WDF. We see clearly the two influences of randomness - i.e. a decay in the propagation of coherence reflected in $\underline{\underline{\bar{\bar{K}}}}(\bar{r}-\bar{R},\bar{q})$ and the scattering of rays reflected in $\underline{\underline{\bar{\bar{b}}}}(\bar{q}-\bar{Q})$. For lack of space, we cannot pursue the subject further, but the WDF view point is quite instructive and may provide a link to the elegant path integral formulations of propagation in random media [18].

"Double random" model of two-point correlations

A very simple statistical model [1] provides a useful means of separately parameterizing density and particle size in a ceramic. Suppose that we examine the microstructure along a lineal test segment. If a given point on the line falls on a void we characterize the point as "state 0." If it falls on a particle of type 1, we characterize the point as "state 1." We may make as many particle assignments as seem warranted by our knowledge of the microstructure. Distinct particle states may either represent physically distinct particles or different orientations of identical anisotropic particles. We define $P_{nm}(r)$ as the conditional probability of finding a state "n" at the point r on the line if a state "m" is measured at the origin. It is physically plausible that the probabilities obey rate equations of the form

$$\frac{d}{dr}(P_{om}) = \sum_{n=1}^{N}\left[-w_{on}P_{om} + w_{no}P_{nm}\right]$$

$$\frac{d}{dr}(P_{nm}) = -w_{no}P_{nm} + w_{on}P_{om} \qquad n \neq o \tag{31}$$

where w_{nm} is the transition probability from state n to state m. Rigorously (and nearly obviously) the rate w_{no} is the inverse of the mean particle size of the nth particle state. We can, of course, use these equations to model any level of multiple randomness, but the study of two particular types of "double randomness" is instructive. In the simplest case we suppose that there many particle states, but they are all characterized by a single transition probability w_{pv} (i.e. single average inverse particle size). In this case, the lineal porosity $P_v = w_{pv}(w_{vp} + w_{pv})$. By stereologic arguments [19] it can be shown that the lineal porosity is equivalent to the conventional volume porosity and, thus, the transition probabilities are simply related to measurable quantities. Suppose we associate with each particle state values of the functions μ and ν which may be components of the $\Delta\bar{\bar{\chi}}(r)$ dyadic. If we then solve Equations 31 for this type of double randomness we find a correlation function

$$\langle\mu(o)\nu(r)\rangle - \langle\mu(o)\rangle\langle\nu(o)\rangle = P_v(1 - P_v)\left[\langle\mu\nu\rangle_p - \mu_v\nu_v\right]\ \exp\{-r(w_{pv})/P_v\}$$
$$+ (1 - P_v)\left[\langle\mu\nu\rangle_p - \langle\mu\rangle_p\langle\nu\rangle_p\right]\ \exp\{-r(w_{pv})\} \tag{32}$$

where the subscipt p denotes averages over the particle states and v denotes values in the void state.

A slightly more complicated type of double randomness involves only two particle states each of which is characterized by a different transition probability. In a real ceramic, one is likely to find considerable particle size disparity which is best represented by a bimodal distribution. Such a microstructure is nicely approximated by this second form of double randomness. Because of space limitations, we cannot treat this case in any detail. It suffices to note that again the correlation functions involve two exponential decays, but now the decay rates η of the correlation are given by the roots of

$$\eta_{1,2}^2 - \eta_{1,2}(w_{01} + w_{02} + w_{10} + w_{20}) + (w_{10}w_{02} + w_{10}w_{20} + w_{20}w_{01}) = 0 \tag{33}$$

The lineal (and, hence, volumetric) porosity is given by $P_0 = w_{10}w_{20}/\eta_1\eta_2$ and the relative concentration of (state 1)/(state 2) particles by $w_{20}w_{01}/w_{10}w_{02}$. Thus, the correlation functions are again completely expressible in term of measurable quantities.

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