It is shown that the theory of random functions permits the expansion of the effective tensor $X_{ijkl}$ for the elastic moduli with respect to correlation functions and that it leads in the second approximation in the Voigt-Reuss scheme to values that lie to one side of the $X_{ijkl}$, while in the third approximation it brackets the latter. The analysis is used to refine the Hashin limits to the elastic moduli for a mechanical mixture of isotropic components and polycrystalline aggregates of cubic structure.

There are two methods for calculating the effective elastic moduli of heterogeneous solids: virial expansion [2] (as a power series in the concentration of one of the components) and the method of correlation functions [2] (expansion with respect to relative fluctuation of the elastic moduli). Identical results should be obtained in the two cases if all terms are incorporated, but great mathematical difficulties restrict one to the lowest approximations. The first approximation in the virial method gives better results when the concentration of one component is low, while the method of correlation functions gives better results when the fluctuations in the elastic moduli are small and the concentrations are similar.

Methods have been developed for determining the upper and lower bounds in both approaches, and various schemes of averaging are used for this purpose in the correlation-function method. The upper bound is established by renormalizing the equation of equilibrium, while the lower one is found by renormalizing the equation of incompatibility.

The range of the bracketsing can be reduced by means of higher approximations. The range can be reduced in the limit to zero, which implies passing from an approximate effective tensor to the true one, which relates the means in stress and strain over the material. Here we show that the two methods of renormalization give identical results when all terms of the series are summed.

If the tensor has a Gaussian distribution, the moment functions of odd order are zero, while the even ones are expressed via combinations of the binary functions [3]. However, a mechanical mixture of several components is not Gaussian, and the odd moments are not zero. Splitting of the higher-order correlation functions is possible also for mechanical mixtures having determinate phase interfaces, but this involves various simplifying assumptions. A derivation is given for a moment of arbitrary order, which allows one to formulate the conditions under which such splitting is possible. The results are used in calculating the exact value of the effective bulk modulus for a medium with a homogeneous shear modulus.

1. To calculate higher-order correlation functions we seek the probability that at point $r_1$ there is component $i$, at point $r_2$ component $j$, etc. The joint probability of this is denoted by $P_{ijk}(r_1, r_2, r_3, \ldots)$. We derive the differential equations satisfied by $P_{ij}(r_1, r_2)$, etc., via the conditional probability $p_{ij}(r_1 | r_2)$, which defines the probability of finding component $i$ at $r_1$ if component $j$ is at $r_2$. We consider only a two-component system, so $i, j = 1, 2$. Then the probability $P_i(r + dr)$ of finding component $i$ at point $r + dr$ is expressed in terms of the probability $P_j(r)$ of finding component $j$ at $r$ by Smoluchowski's equation:

$$P_i(r + dr) = \sum p_{ij}(r + dr | r) P_j(r).$$ (1.1)

We assume that the mixture obeys the condition

$$p_{ij}(r + dr | r) = \delta_{ij} + A_{ij}(r) dr,$$ (1.2)

in which the matrix $A_{ij}(r)$ is defined by

$$A_{ij}(r) = \left[ \frac{\partial}{\partial r} p_{ij}(r) \right]_{r=0} \left( \sum_i A_{ij} = 0 \right)$$ (1.3)

and satisfies the normalization condition stated in parentheses.

For an isotropic and quasi-homogeneous medium

$$A_{ij}(r) = A_{ij}^0 \frac{r}{r} = A_{ij} n.$$ (1.4)

We substitute (1.2) into (1.1) to get

$$\nabla P_i(r) = \sum_j A_{ij} P_j(r).$$ (1.5)

We solve (1.5) with (1.4) and the normalization conditions to get

$$P_1(r) = a A_{11} + C_1 \exp\left(-\frac{r}{a}\right),$$

$$P_2(r) = a A_{22} + C_2 \exp\left(-\frac{r}{a}\right),$$

$$a \equiv (A_{12} - A_{21})^{-1}.$$ (1.6)

Then, in view of the condition of quasi homogeneity,

$$P_1(r) = c_1,$$ (1.7)

we find the relation of $A_{ij}$ to the component concentrations

$$a A_{ij} = \begin{bmatrix} c_1 & c_1 \\ c_2 & c_2 \end{bmatrix}.$$ (1.8)

We use the following equation to calculate the joint probability $P_{ij}(r_1, r_2)$:

$$P_{ij}(r_1, r_2) = \sum_k p_{ik}(r_1 | r_2) P_{kj}(r_1, r_2).$$ (1.9)

Then, as $P_{ij}(r_1, r_2) = P_{ij}(|r_1 - r_2|)$ for a quasi-homogeneous medium, we have

$$\frac{d}{dr} P_{ij}(r) = \sum_k A_{ik} P_{kj}(r).$$ (1.10)

Substitution for $A_{ik}$ from (1.8) gives

$$a \frac{d}{dr} P_{ij}(r) = -P_{ij}(r) + P_i P_j.$$ (1.11)

The following is the solution to (1.11) that satisfies $P_{ij}(0) = c_0 \delta_{ij}$:
\begin{equation}
\rho(r) = \exp\left(-\frac{r}{a}\right), \quad \alpha_{ij} = c_{ij} \quad \beta_{ij} = \delta_{ij} - c_{ij}.
\end{equation}

Then (1.12) gives the joint probability found under the condition that the passage from the initial state to the final state is performed by motion of a single point of radius \( r \). Now let the transition be performed by change in the coordinates of the start and end of the line, i.e., there is a transition from the state \( r_1, k \) to \( r_2, l \). This approach allows us to introduce the four-subscript matrix \( A_{ijkl} \), which will be used in what follows to calculate the joint probability \( P_{ijkl}(r_1, r_2, r_3) \). Then (1.9) is replaced by

\begin{equation}
P_{ij}(r_1, r_2) = \sum_{k} P_{ijkl}(r_1, r_2, r_3) P_{kl}(r_3, r_4).
\end{equation}

To calculate the matrix for transition from state \( kl \) to new state \( ij \) we resolve \( dr \) into the normal component \( dr_n = dr \cdot n \) and the tangential component \( dr_T = dr \cdot \tau \). If \( dr_T = 0 \), the transition occurs between points lying on the ray \( r/r = \text{const} \), while \( dr_n = 0 \) means transition between points on the sphere \( r = \text{const} \). The first transition alters the ratio of the radius to the scale of the correlations, while the second leaves it unchanged.

We therefore consider transitions as identical if the initial and final states have the same ratio of the distances to the scale of the correlations. A sphere represents the surface formed by the correlation-scale vector for an untextured medium, so we can put \( dr_T = 0 \) and consider only transitions that alter the absolute magnitude of the distance between points.

The following matrix then describes the transition from \( kl \), \( r \) to \( ij \), \( r + dr \):

\begin{equation}
P_{ij}(r+dr, r) = \delta_{ik} \delta_{jl} + A_{ijkl} dr,
\end{equation}

and substitution into (2.14) gives

\begin{equation}
\frac{d}{dr} P_{ij} = \sum_{k,l} A_{ijkl} P_{kl}.
\end{equation}

The equivalence of (1.16) and (1.10) means that \( A_{ijkl} \) takes the form

\begin{equation}
aA_{ijkl} = - (\delta_{ik} - P_i) (\delta_{jl} - P_j).
\end{equation}

Relation (1.19) imposes severe restrictions on the structure of the mixture. If we take account only of the mean grain sizes of the components in calculating the higher moments (these sizes are related to the correlation scale by the rule for mechanical mixing), this is equivalent to ignoring the grain shape, which can be taken into account, for example, via the set of moments \( \langle R^6 \rangle \), \( \langle R^2 R^4 \rangle \), etc., where \( R \) is the coordinate of the surface of a grain in a frame of reference whose origin coincides with the center of gravity, \( \omega = R/R \), and \( \langle \cdot \rangle \) denotes averaging with respect to angle. Moreover, no distinction is drawn between the matrix and an inclusion, so the coupling is considered identical for the regions filled by the two components.

Equation (1.16) is applicable if the component concentrations do not differ too greatly and the components have similar degrees of coupling. The method should give the best results for quasi-spherical grains, when \( \langle R - R' \rangle^2 \ll \langle R \rangle^2 \).

We now consider the calculations of \( P_{ijkl}(r_1, r_2, r_3) \). As the medium is isotropic and quasi-homogeneous, \( P_{ijkl}(r_1, r_2, r_3, r_4) \) can be put as \( P_{ijkl}(r', r'', r''' \rangle \), where

\begin{equation}
r' = |r_2 - r_3|, \quad r'' = |r_4 - r_3|, \quad r''' = |r_1 - r_4|.
\end{equation}

We envisage transitions such that \( r'' = \text{const} \) and \( r''' = \text{const} \) but \( r' \neq \text{const} \). Arguments similar to those for (1.15) and (1.17) then give

\begin{equation}
\frac{\partial}{\partial r'} P_{ijkl}(r', r'', r''') = \sum_{m,n} A_{ijklm} P_{lmn}(r', r'', r''') \quad (2.1)
\end{equation}

We substitute from (1.17) for the \( A_{ijklm} \) to get

\begin{equation}
\frac{\partial}{\partial r'} P_{ijkl}(r', r'', r''') = \frac{1}{a} [P_{ijkl}(r', r'', r''') - P_{ijk}(r') - P_{ijl}(r'') + P_{i} P_{j} P_{k}].
\end{equation}

This allows us to calculate the third-order correlation function for the tensor. We introduce moment \( N \) of the tensor in order to find the differential equation for the explicit form of this function. For simplicity we omit the tensor subscripts \( ^t_{pqrs} \equiv \lambda_i \) and get for this moment that

\begin{equation}
M_{a,b,...,n}(r_1, ..., r_n) = \langle \lambda^a_i (r_1) \lambda^b_j (r_2) \ldots \lambda^n_k (r_n) \rangle = \lambda^a_i \lambda^b_j \ldots \lambda^n_k P_{ijkl} (r_1, ..., r_n)
\end{equation}

with \( \sum_{i=1}^n x_i = N \). The centered moment of order \( N \) is then given by

\begin{equation}
u_{a,b,...,n}(r_1, r_2, ..., r_n) =
= \langle [\lambda (r_1) - \langle \lambda (r_1) \rangle]^b [\lambda (r_2) - \langle \lambda (r_2) \rangle] \ldots [\lambda (r_n) - \langle \lambda (r_n) \rangle] \rangle
= \sum_{i,j,l} \langle [\lambda_i - \langle \lambda_i \rangle]^b \ldots [\lambda_l - \langle \lambda_l \rangle] \rangle
= \langle [\lambda (r_1)]^b \ldots [\lambda (r_n)] \rangle P_{ijkl} (r_1, ..., r_n).
\end{equation}

We get from (2.3) and (2.4) for an isotropic quasihomogeneous medium that

\begin{equation}
M_N (r) = \langle \lambda^N (r) \rangle = \sum_{i=1}^N \gamma_i P_i (r) = \sum_{i=1}^N c_i,
\end{equation}

\begin{align*}
\mu_2 &= 0, \quad \mu_3 = 2M_2 - 2M_1, \quad \mu_3 = 3M_3 - 3M_2 - 2M_1, \\
M_{11} (r_1, r_2) &= M_1^2 + \mu_2 ^2 R^2 / a, \quad M_{11} (r_1, r_2, r_3) = \sum_{k,l} \gamma_k \gamma_l P_{ijkl} (r', r'', r'''), \\
M_{11} (r_1, r_2, r_3) &= M_{11} (r_1, r_2, r_3) + M_{11} (r') + M_{11} (r'') + M_{11} (r''') + 2M_{11}.
\end{align*}

This expression gives

\begin{equation}
\frac{\partial}{\partial r'} M_{11} (r', r'', r''') = \frac{\partial}{\partial r'} M_{11} (r', r'', r''') - M_{1} \frac{\partial}{\partial r'} M_{11} (r')
\end{equation}

The derivatives on the right-hand side of (2.6) are defined from (1.11), (2.2), and (2.3), so