MACROSCOPIC ENGINEERING PROPERTIES OF POLYCRYSTALLINE MATERIALS
I: GENERAL METHODS

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August 1974

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Macroscopic Engineering Properties of Polycrystalline Materials

I: General Methods

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Abstract

We outline the formal application of integral equation methods (Green's function techniques) to the systematic computation of the effective elastic properties of polycrystalline materials from the specification of the statistical distribution of elastic properties of the constituent grains (or pores). These methods are not entirely new, but by the use of developments in the study of the electronic properties and lattice dynamics of disordered solids, we compare various methods on a common footing and indicate their limitations. Some details for the actual application of these methods are also presented.

I. Introduction

The calculation of the macroscopic elastic properties of a grainy polycrystalline material, or a heterogeneous mixture, is an old problem on which so much has been written that we make no attempt at an exhaustive review here.¹ Many of these earlier approaches were ingenious, though basically intuitive, and met with varying degrees of success, except for materials with strong anisotropy, porosity, or strong variation in composition. Among these methods are those of Eshelby,² Hill,³ Budiansky,⁴ Hershey,⁵ and Kröner (1958).⁶ A more rigorous technique has been based on variational arguments,⁷ but again a degree of good judgement in the choice of trial functions is essential to obtain practical results. Variational methods, however, are useful for obtaining bounds on the effective properties.

An essential difficulty to rigorous theoretical development is that the laws relating the local stress, strain, and material properties are generally stated in differential form and very little formal methodology has been developed to find solutions of differential equations with randomly varying coefficients, particularly in three dimensions. However, it is possible to cast the equations governing stress and strain fields in integral form for which formally straightforward iterative or perturbation methods of solution exist. In addition, approximations incurred in these methods can be precisely characterized. Traditionally, such methods were not often employed because of the labor of computing the resulting integrals. Modern computers have changed this; therefore, we argue that formal definiteness coupled with computational feasibility strongly favor the use of integral

equation methods. In applications to elastic properties Kröner,⁸ Zeller and Dederichs,⁹ and Korringa¹⁰ have made use of perturbation series and approximations to the integral equations.

A particularly useful approximation is often referred to as the "self-consistent effective medium theory." For some time how this approximation was related to perturbation theory was not known, but now¹ from the quantum mechanics of random solids, which is algebraically similar to the present problem, it is known that the effective medium is that medium which makes the average scattering by the constituent grains zero, if they scatter independently. In the solid state physics literature the effective medium method is called the "coherent potential approximation." Whether obtained by intuition or a formal perturbation approach, this recipe for determining macroscopic (average) properties is now believed to be the best possible when specific details of inter-particle scattering are not important.

Calculations based on this theory have been carried out by Kneer¹¹ and by Morris¹² for textured non-porous media composed of weakly anisotropic grains. In another paper¹³ we report on a more severe test of the theory, an application to porous polycrystalline graphite which is composed of highly anisotropic grains. In essence the final operational formulas we use are the same as Kneer's, but we shall arrive at these after a discussion which has several objectives: (1) to define "effective macroscopic properties" with some care, (2) to relate their calculation to quantum mechanical scattering theory, including its diagrammatic techniques, and (3) to provide a formulation which can be extended to dynamic properties.

We summarize in Section II the general philosophy of "effective" moduli, while in Section III we present the formal theory of fields in an inhomogeneous medium in the language of scattering theory and integral equations. In Section IV we discuss the special case of piecewise, continuous, random material parameters (i.e. statistically independent grains or pores) and the resulting self-consistent mean field theory; we also indicate the nature of other approximations which can be used. In Section V we collect some of the technical details necessary to apply the methods to actual materials.

II. Effective Macroscopic Properties

The definition of an effective macroscopic property is independent of any approximation method. Before we develop such methods for calculating an effective elastic modulus, we first define this term in detail. The manner of definition can be easily generalized to other macroscopic properties.

We consider a statistically homogeneous, i.e. macroscopically uniform, sample consisting of enough grains that probability distributions for composition and orientation can be well defined. We then relate sample averages of the stress and strain fields to their measured values. In this manner there is a direct, operational definition of effective elastic moduli. The first stage of this definition consists of the following: (1) Fourier analyze the components of the stress and strain fields by transforming their space and time variables to their dual variables $\underline{q}$ and ω . (2) Assume that a linear relation between the stress and strain transforms exists. (3) Designate the constant of proportionality as a generalized susceptibility, $\chi(\underline{q}, \omega)$. Since the material is macroscopically uniform, the generalized susceptibility is diagonal in $\underline{q}$ and ω . (4) Write

$$\sigma(\underline{q}, \omega) = \chi(\underline{q}, \omega) \epsilon(\underline{q}, \omega) \quad (1)$$

where σ is the stress field and ϵ is the strain field.

In an ultrasonic experiment $\underline{q}$ and ω are non-zero; for static elasticity $\underline{q} = 0$, $\omega = 0$. In this paper we are concerned almost exclusively with the latter case. However, before specializing to this case, we make some general remarks.

First, from the form of Eq. 1 it follows from the convolution theorem that in $\underline{r}$ -space χ is generally non-local:

$$\sigma(\underline{r}, \omega) = \int d\underline{r}' \chi(\underline{r} - \underline{r}', \omega) \epsilon(\underline{r}', \omega) \quad (2)$$

An exception is $\chi(\underline{q}, \omega)$ independent of $\underline{q}$ whence $\chi = \delta(\underline{r} - \underline{r}') \epsilon(\omega)$, but because of heterogeneity this can never occur in polycrystalline materials.

Second, a definition of Fourier transforms serves to define sample averages. For example,

$$\epsilon(\underline{q}, \omega) = \frac{1}{2\pi V} \iint d\underline{r} dt e^{-i(\underline{q} \cdot \underline{r} - \omega t)} \epsilon(\underline{r}, t) \quad (3)$$

automatically integrates over the sample. In particular, the effective static properties involve simple volume averages:

$$\begin{aligned} \epsilon(0;0) &= \frac{1}{V} \int d\underline{r} \epsilon(\underline{r}) \\ \sigma(0;0) &= \frac{1}{V} \int d\underline{r} \sigma(\underline{r}). \end{aligned} \quad (4)$$

We now come to our objective which is an operational definition of an effective modulus. This definition to have definite meaning must be independent of any particular sample. This independence in turn requires the volume V of the sample to be very large compared to the size of the grains and to include many grains. In the limit of $V \rightarrow \infty$, the sample contains many similar regions, each representative of the entire sample; a volume average averages over these representative regions, and thus becomes identical to an ensemble average which averages over many representative samples. Carrying out of the volume average of a sample requires, however, the complete specification of the structural details of that

sample. In practice, these details are never specified completely. What are specified are position-dependent distribution functions of various material parameters such as grain shape, composition, and orientation. Thus, an ensemble average replaces the volume average. Denoting the ensemble-averaged results of $\sigma(\underline{r})$ and $\epsilon(\underline{r})$ by $\langle \sigma \rangle$ and $\langle \epsilon \rangle$, we write

$$\chi^* = \langle \sigma \rangle \langle \epsilon \rangle^{-1}. \quad (5)$$

This defines the effective modulus. In (5) we dropped the explicit $(\underline{q}, \omega)$ dependence and will generally do so since we discuss mainly static properties; however, the same considerations apply for $\underline{q} \neq 0$, as long as grain size and structural correlation lengths are negligably small compared to a wave length.

The burden of calculation of the effective elastic moduli has been transferred to the determination of the stress and strain fields in a heterogeneous elastic medium. As stated in the Introduction, our view is that there are two approaches to determining these fields: intuition and general formalism. In the former category one may place the early approaches that used Eshelby's solutions for an inclusion and the various self-consistent methods based on imbedding that inclusion in an effective medium. Thus, the stress and strain fields of one grain are found by ingenious choice of a model problem, a "trick"; the whole sample is put together in some mathematically uncontrolled average way.

However, it is possible to write the general governing equation for the strain fields; for the static case in tensor notation, this equation is

$$\frac{\partial}{\partial x_j} \sigma_{ij}(\underline{r}) = \frac{\partial}{\partial x_j} [C_{ijkl}(\underline{r}) \epsilon_{kl}(\underline{r})] = F_i^A \quad (6)$$

Integral equations with Green's function kernels provide an explicit method for solving this equation exactly. (In the special case of a single inclusion, Eshelby's results can be reproduced.) The technical details of the solution are reviewed in Sections III and IV, but the point is that the general solution of the integral equation can be systematically approximated, which allows determination of $\langle \sigma \rangle$ and $\langle \epsilon \rangle$ from any specified statistical distribution of $C_{ijkl}(\underline{r})$. This determination furnishes the information needed to compute the effective macroscopic moduli. Moreover, beyond formal generality, the integral equation method is feasible for computation.

Finally, attention should be drawn to the fact that complete stress-strain field information is only obtainable from a knowledge of all Fourier components, and the local variations of these fields and the strain energy cannot be obtained from the effective modulus $\chi^*(0;0)$. This fact must be kept in mind, if one discusses total internal strain energy.

III. Solutions of the Equations for Static Elastic Fields

The (symmetric) strain is determined from the displacement field u_i by

$$\epsilon_{ij}(\underline{r}) = \frac{1}{2} \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right) = \epsilon_{ji}(\underline{r}) \quad (7)$$

We will frequently use the condensed notation $\epsilon_{ij} = \frac{1}{2}(u_{i,j} + u_{j,i})$, the comma denoting differentiation. The material distribution is locally characterized by an elastic stiffness $C_{ijkl}(\underline{r})$, and the local stress is

$$\sigma_{ij}(\underline{r}) = C_{ijkl}(\underline{r}) \epsilon_{kl}(\underline{r}). \quad (8)$$

Since the sample is uniform on the average, it is useful to separate the stiffness into spatially constant and varying parts,

$$C = C^0 + \delta C \quad (9)$$

where the tensor indices are to be understood whenever obvious; this separation is not unique. For static elasticity we want the quantities

$$\begin{aligned} \langle \epsilon \rangle &= \frac{1}{V} \int d\underline{r} \epsilon(\underline{r}) \\ \langle \sigma \rangle &= \frac{1}{V} \int d\underline{r} \sigma(\underline{r}). \end{aligned} \quad (4)$$

The defining equations for these fields, Eq. (6), can be written in several ways:

$$\sigma_{ij,j} = F_i^A \quad (10a)$$

$$(C_{ijkl}\epsilon_{kl})_{,j} = F_i^A \quad (10b)$$

$$C_{ijkl}^0\epsilon_{kl,j} + (\delta C_{ijkl}\epsilon_{kl})_{,j} = F_i^A \quad (10c)$$

where repeated indices imply the summation convention, and again the comma denotes differentiation. By writing Eq. (10c) as

$$C_{ijkl}^0 \epsilon_{kl,j} = -(\delta C_{ijkl} \epsilon_{kl})_{,j} + F_i^A, \quad (11)$$

one may formally regard the divergence of the stress variation produced by δC as a fictitious applied force. In general, traditional elasticity is concerned with the solutions of these differential equations when $\delta C = 0$. When there are applied forces, the standard method is to construct an integrating factor, which is quite the same as a Green's operator. Using this Green's (integral) operator, one can convert the above differential equation for $\epsilon(\underline{r})$ into an equivalent integral equation. We will first define the Green's operator and then make the conversion. Our discussion parallels Zeller and Dederichs⁹ and uses their notation.

For a uniform elastic medium C_{ijkl}^0 the displacement in the k th direction at $\underline{r}$ due to a point force applied in the m th direction at $\underline{r}'$ is given by the displacement Green's function

$$g_{km}(\underline{r}, \underline{r}') \quad (12)$$

which satisfies the second order differential equation

$$C_{ijkl}^0 g_{km,lj}(\underline{r}, \underline{r}') + \delta_{im} \delta(\underline{r} - \underline{r}') = 0. \quad (13)$$

We choose the boundary condition $g \rightarrow 0$ as $\underline{r} \rightarrow \infty$; for a large sample this choice is not critical. The linear operator allows one, by the superposition theorem, to write the solution of Eq. 11 as

$$u_i(\underline{r}) = u_i^0(\underline{r}) + \int d\underline{r}' g_{ik}(\underline{r}, \underline{r}') [-F_k^A(\underline{r}') + (\delta C_{klmn}(\underline{r}') \epsilon_{mn}(\underline{r}'))_{,l}] \quad (14)$$

where $u_i^0(\underline{r})$ is any solution of the homogeneous equation. For a uniform isotropic medium the Green's operator may be easily found. The solution is given by Love¹⁴ and is used by Eshelby² in a specific context. Equation (14) is in an inconvenient form since it mixes the displacement and strain fields. By computing $\frac{1}{2}(u_{i,j} + u_{j,i})$ from (14), one can work entirely in terms of the strain field, with a tensor Green's function defined by

$$G_{ijkl}(\underline{r}, \underline{r}') = \frac{1}{2}(g_{ik,jl'} + g_{jk,il'}). \quad (15)$$

This is an operator which gives the ij strain at $\underline{r}$ due to an applied stress kl at $\underline{r}'$ in a medium with C_{ijkl}^0 . Then from (14) with $F^A = 0$ we find (Zeller and Dederichs)

$$\epsilon_{ij}(\underline{r}) = \epsilon_{ij}^0(\underline{r}) + \int d\underline{r}' G_{ijkl}(\underline{r}, \underline{r}') \delta C_{klmn}(\underline{r}') \epsilon_{mn}(\underline{r}'). \quad (16)$$

This integral equation is the basic equation for most of what follows.

Equation (16) may be used to generate various approximations, just as in quantum mechanical scattering theory. For example, the first Born approximation is found by setting in the integral $\epsilon_{mn}(\underline{r}')$ equal to ϵ_{mn}^0 :

$$\epsilon = \epsilon^0 + G \delta C \epsilon^0 \quad (17)$$

where cartesian tensor indices have been suppressed, and the product of non-local operators such as $G(\underline{r}, \underline{r}')$ with fields such as δC implies the spatial integration written in detail in (16). Then, from the physical meaning of G , the interpretation of (17) is that $\epsilon = \epsilon^0 +$ strain at $\underline{r}$ produced by excess stress at $\underline{r}'$ due to $\delta C(\underline{r}')$. In a sense $\delta C(\underline{r}')$ "scatters" strain field to $\underline{r}'$ via the "propagator" $G(\underline{r}, \underline{r}')$. The first Born approximation is the leading

term in the iterated solution of (16):

$$\epsilon = \epsilon^0 + G\delta C \epsilon^0 + G\delta CG\delta C \epsilon^0 + \dots \quad (18)$$

In recent physics (solid state, field theory) scattering descriptions have been of great utility, but because of the cumbersome algebra in higher terms a shorthand has been developed to represent the formulae by diagrams. The actual symbols are not unique, but are simply defined for the purpose at hand. For example, we may represent (18) as shown in Fig. 1. The rules are:

- (1) ϵ^0 is a wavy line
- (2) G is a solid line
- (3) $\delta C(\underline{r})$ is a dotted line from a cross.
- (4) A vertex (intersection of lines) implies operator multiplication.

Thus the strain field ϵ_0 may

- (1) Propagate directly from $\underline{r}' \rightarrow \underline{r}$.
- (2) Propagate from $\underline{r}' \rightarrow \underline{x}$, develop an excess stress $\delta C(\underline{x})\epsilon^0$, which propagates from $\underline{x}$ via $G(\underline{r}, \underline{x})$ to produce a strain at $\underline{r}$. This strain is to be summed over all intermediate points $\underline{x}$ and thus describes the result of all variations δC to first order in δC .

(3) In higher orders the δC at intermediate points $\underline{x}$, $\underline{y}$, $\underline{z}$, etc. scatter the uniform strain field ϵ^0 . Two cases must be distinguished: all intermediate sites equal and two or more intermediate sites unequal. The repeated scattering by the same site $\underline{x} = \underline{y} = \underline{z}$ etc. gives the complete solution for a single "defect." Scattering from two or more unequal sites, $\underline{x}$, $\underline{y}$, for example, involves the properties at both $\underline{x}$ and $\underline{y}$ which in general

can only be specified by correlation functions. Figure 2 shows repeated scattering at the same grain; Fig. 3 shows scattering by two different grains. For each such diagram the rules yield a specific algebraic (integral) expression. For example, Fig. 3 is represented by $G\delta C G\delta C \epsilon^0 = \int d\underline{x} \int d\underline{y} G(\underline{r}, \underline{x}) \delta(\underline{x}) G(\underline{x}, \underline{y}) \delta C(\underline{y}) \epsilon^0$.

This discussion has been brief, but hopefully may introduce some readers to the solid state literature¹ relevant to this problem. An important point is that diagrams can be used to characterize precisely what approximations have been made.

It is particularly useful to sum formally the series (18) to obtain

$$\epsilon = \epsilon^0 + G[\delta C \frac{1}{I - G\delta C}] \epsilon^0$$

or

$$\epsilon = \epsilon^0 + G T \epsilon^0 \quad (19)$$

where $T = \delta C(I - G\delta C)^{-1}$ and the previously defined rules of multiplication apply. The unit operator is $I = \frac{1}{2}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})\delta(\underline{r}-\underline{r}')$.

A knowledge of T determines the total strain field; it is sometimes called the "T-matrix" in scattering theory because it completely specifies "transitions" out of the "incident" field ϵ^0 . Implementing the operator equation computationally is discussed in Sections IV and V.

To complete this section, we compute the effective static modulus:

$$\chi^* = C_{ijkl}^* = \langle \sigma \rangle_{ijmn} \langle \epsilon \rangle_{mnkl}^{-1}.$$

From (19), we find

$$\langle \epsilon \rangle = \epsilon^0 + G\langle T \rangle \epsilon^0, \quad (20)$$

and from (8) and (9),

$$\langle \sigma \rangle = \langle (C^0 + \delta C) \epsilon \rangle \doteq C^0 \langle \epsilon \rangle + \langle \delta C \epsilon \rangle. \quad (21)$$

But from (19) and (20), $\langle \delta C \epsilon \rangle = \langle T \rangle \epsilon^0$ whence

$$C^* = C^0 + \langle T \rangle \frac{1}{1 + G\langle T \rangle}. \quad (22)$$

Within the given definition of $\chi^* \equiv C^*$, Eq. (22) is a complete and exact solution, if $\langle T \rangle$ can be computed. Unfortunately, $\langle T \rangle$ can only be approximated, using the nature of the material for guidance. We next consider several such approximations.

IV. Approximations and Particular Models

When dealing with polycrystalline materials, we can assume without loss of generality that the stiffness can be decomposed into the sum of various contributions:

$$\delta C = \sum_{\alpha} \delta C^{\alpha} \theta_{\alpha}(\underline{r}) \quad (23)$$

where θ is the unity if $\underline{r}$ is in the α -th grain. The term grain is used generically and may even include pores. In general, a material has a distribution of species, particle shapes, and orientations of local elastic tensor; averages are performed over all of these distributions. We denote such averages by $\langle \rangle$. In a uniform sample volume averaging should be the same as distribution averaging at one position.

We now briefly discuss several increasingly more accurate approximations to (22):

(a) The Voigt Average.

If the strain is simply assumed everywhere to be constant, ϵ^0 , then the effective modulus is simply

$$C^* = \langle (C^0 + \delta C) \epsilon^0 \rangle \langle \epsilon^0 \rangle^{-1} = \langle C \rangle \quad (24)$$

This is a simple volume average of the stiffness. No variation in ϵ because of $\delta C(\underline{r})$ is accounted for. It is not surprising that the Voigt average is of limited value.

(b) Voigt Average as an Upper Bound

A next approximation is to set $T = \delta C$, i.e. the lowest term in the series expansion for T , whence

$$\begin{aligned} C^* &= C^0 + \langle \delta C \rangle \frac{1}{1 + G \langle \delta C \rangle} \\ &= C^{\text{Voigt}} - \frac{\langle \delta C \rangle G \langle \delta C \rangle}{1 + G \langle \delta C \rangle} \end{aligned} \quad (25)$$

The second term always subtracts from C^{Voigt} (which can be shown generally but with difficulty), and this is consistent with variational statements which prove that the Voigt average is an upper bound.

(c) Piecewise Averaging

If the specimen is statistically uniform, and if the particles are statistically independent, then when we average terms in T , those terms involving scattering at different sites will factor, i.e., for $\alpha \neq \beta$

$$\langle \delta C^\alpha G \delta C^\beta \rangle = \langle \delta C^\alpha \rangle G \langle \delta C^\beta \rangle. \quad (26)$$

On the other hand all the repeated scatterings at a given site defined a geometric series, so we sum those terms exactly. The series (18) can now be rearranged in terms of the single grain T-matrix

$$t^\alpha = \frac{\delta C^\alpha}{1 - G_\alpha \delta C^\alpha} \quad (27)$$

where G_α starts and returns to the α -th grain. A diagrammatic representation of this series is shown in Fig. 4. Thus

$$\epsilon = \epsilon_0 + G(\sum_\alpha t^\alpha) \epsilon_0 + G(\sum_\alpha t^\alpha) G(\sum_{\beta \neq \alpha} t^\beta) \epsilon_0 + \dots \quad (28)$$

The more complicated features of the terms involving two or more t 's are the subject of considerable discussion¹ in solid state physics; they involve the treatment of pairs and larger clusters of grains. We neglect these intergranular scattering effects in this next approximation and write

$$\langle T \rangle = \sum_{\alpha} \langle t^{\alpha} \rangle \quad (29)$$

which is equivalent to a volume average for a statistically uniform sample.

So far we have not specified C^0 , upon which all the t^{α} depend. A choice of $C^0 = C^{\text{Voigt}}$ in Eq. (29) leads to

$$C^* = C^{\text{Voigt}} + \left(\sum_{\alpha} \langle t^{\alpha} \rangle \right) \left(1 + \sum_{\alpha} \langle t^{\alpha} \rangle \right)^{-1} \quad (30)$$

This approximation is equivalent in the electronic literature to the "Average t-matrix Approximation", ATA. The single grain T-matrix solves the elastic problem for a single grain imbedded in C^0 , i.e., Eshelby's problem:

$$\epsilon = \epsilon_0 + G t^{\alpha} \epsilon_0. \quad (31)$$

The ATA simply volume averages that strain with no allowance for intergranular scattering.

Kröner (1968) has derived an expression for the elastic moduli of a perfectly disordered aggregate which is the first term in an expansion in $\sum_{\alpha} \langle t^{\alpha} \rangle$ of (30). Contributions from terms like Fig. 5 are not included in the Kröner approximation.

(d) Self-Consistent Effective Medium.

An improvement to (30) can be obtained by taking advantage of the freedom to choose and choosing C^0 so that it averages over the effects of the surrounding medium. This is equivalent to including the higher order terms in (28) on the average, but neglecting details of intergranular scattering. As Hershey,

Kröner (1958), Hill, and Budiansky have shown for inclusions imbedded in an effective medium, self-consistent conditions can be found and constitute a set of implicit equations for C^0 .

From the point of view of scattering theory, we start from (18)

$$C^* = C^0 + \langle T \rangle (1 + G\langle T \rangle)^{-1}, \quad (18)$$

T is a function of C^0 , but C^0 can be varied so that $\langle T \rangle = 0$.

Denoting that particular C^0 by $C_{sc.}$, then we have

$$C^* = C_{sc.} + 0, \quad (32)$$

This defines an effective medium, in which on the average the total scattering by all the grains is zero.

It is not feasible to compute T generally, so the self-consistent field condition is usually approximated using single grain T -matrices, including interparticle scattering on the average only. If the details of Vilicky, Kirkpatrick, and Ehrenreich¹³ are followed, the equation analogous to their Eq. (2.22) is

$$C^* = C^0 + \sum_{\alpha} \langle t^{\alpha} \rangle [1 + G\langle t^{\alpha} \rangle]^{-1}. \quad (33)$$

Then the implicit equation determining $C^* = C_{sc.}$ is simply

$$\langle t^{\alpha} \rangle = 0. \quad (34)$$

This requires averaging over types of grains, grain shape, and orientation. It is not possible to prove that this implicit, tensor equation has solutions, but in a variety of applications to date it has been possible to obtain good results.^{13,15} The effect of condition (34) on the strain field can be easily seen

from the ensemble average of (31): The self-consistent homogeneous field ϵ_0 equals $\langle \epsilon \rangle$. The technical details of the specific algebra, integrals, and computations are discussed in the next section, V.

It is not obvious that the single grain zero scattering condition (34) and self-consistent inclusion averaging of Hershey, Kröner, Budiansky, and Hill yield the same effective medium but in fact they do. In the literature of electronic properties of random alloys, the equivalence has been demonstrated exactly¹ in the approximation of neglecting detailed interparticle scattering i.e. pairs and larger clusters."

If this statement based on scattering theory is equivalent to the conditions developed by phenomenological arguments (Hershey, Kröner, Hill, Budiansky), what then is its advantage? There are at least four:

- (1) The application to various conditions of composition, structure, and anisotropy follows a specific recipe.
- (2) It is always possible to find out what error is being committed by computing higher terms in the scattered fields. No such recipe is given at all in the phenomenological derivations.
- (3) In principle structural correlations among the grains may be included by appropriate treatment of higher terms in the scattering.
- (4) The method applies to the dynamic case, i.e. $\omega \neq 0$.

V. Some Details for Application

The self-consistent elastic stiffness C^* satisfies

$$\langle t^a \rangle = 0. \quad (34)$$

In general, this is an implicit, non-linear, tensor equation involving all the stiffness constants and having sufficient complexity to require the use of computers. We now present some details of a straightforward scheme for such a computer solution. The scheme is an iterative process and uses matrix algebra; both of these techniques are readily adaptable to computers.

In general, the ensemble average in (34) involves averages over distributions of grain composition, orientation, and shape. Normally, a polycrystalline material is modeled with all the grains shaped identically; even so the ensemble average includes averages over the distribution of the orientations of this shape. For simplicity, we assume a model in which the grains either are spherical or have shapes precisely correlated with their elastic anisotropy. This model allows us to consider only distributions of grain composition and stiffness tensor orientation.

We write the implicit equation (34) for C^* as

$$\sum_{j=1}^N v_j \langle t^j(\Omega_j) \rangle_{\Omega_j} = 0$$

where $t^j(\Omega_j)$ is a fourth rank tensor defined by

$$t^j(\Omega_j) = (C^j(\Omega_j) - C^*)[I - G(C^*)(C^j(\Omega_j) - C^*)]^{-1} \quad (35)$$

with $C^j(\Omega)$ the stiffness tensor of a grain of type j whose principal axes have an orientation Ω_j with respect to the principal

axes of C^* , $G(C^*)$ the Green's function tensor for the strain field in a medium with stiffness C^* , I the identity tensor, and v_j the volume fraction of grains of type j . The angular brackets $\langle \rangle_{\Omega_j}$ indicate an average of the orientation Ω_j of each grain type, and the summation is over N distinct grain types. If "grain" denotes "pore", $C^j = 0$.

Self-consistency can be reached by the iterative algorithm relating the $(i+1)$ -th to i -th approximation

$$C_{i+1} = C_i + \sum_{j=1}^N v_j \langle \tau_i^j(\Omega_j) \rangle_{\Omega_j} \quad (36)$$

Equation (36) implies (34) when

$$|C_i - C_{i+1}| \leq \Delta |C_{i+1}| \quad (38)$$

where Δ is a suitably chosen, small positive number. When (38) is satisfied C_{i+1} is said to equal C^* . The iteration scheme may be initialized by $C_0 = C^{\text{Voigt}}$. Then the resulting first iteration C_1 is the Kröner approximation to the average T-matrix approximation.

The δC^α in (27) are non-zero only over the volume of the grain α . This has several implications for the product $G_\alpha \delta C^\alpha$:

- (1) The implicit integration is restricted to the grain volume.
- (2) The integration is only over G_α and yields a constant tensor $\mathcal{H}_\alpha$.
- (3) The product $G_\alpha \delta C^\alpha$ is thus equivalent for a given grain orientation to the product of two tensors $\mathcal{H}_\alpha$ and δC^α . We now write (37) as

$$\tau_i^j(\Omega_j) = (C^j(\Omega_j) - C_i) [I - \mathcal{H}_j(C_i)(C^j(\Omega_j) - C_i)]^{-1}. \quad (39)$$

The problem has been reduced to the addition, subtraction, multiplication, and inversion of fourth rank constant tensors. We now

describe an isomorphism between tensor elements and matrix elements which allows the tensor operations to be replaced by matrix operations.¹⁶

This isomorphism is established as follows: First, for a given tensor A_{ijkl} , pairs of indices ij and kl are converted to single indices α and β by the standard convention

$$\begin{aligned} 11 &\rightarrow 1 \\ 22 &\rightarrow 2 \\ 33 &\rightarrow 3 \\ 23, 32 &\rightarrow 4 \\ 13, 31 &\rightarrow 5 \\ 12, 21 &\rightarrow 6. \end{aligned}$$

Next, each tensor element A_{ijkl} is associated with a matrix element $A_{\alpha\beta}$ by the rules

$$\begin{aligned} A_{ijkl} &= A_{\alpha\beta}; \quad \alpha, \beta \leq 3 \\ A_{ijkl} &= \sqrt{2}A_{\alpha\beta}; \quad \alpha \text{ or } \beta > 3 \\ A_{ijkl} &= 2A_{\alpha\beta}; \quad \alpha, \beta > 3. \end{aligned}$$

The resulting matrices are of rank 6.

There are three remaining computational details: finding $C^j(\Omega_j)$, evaluating $\bar{\nu}_j$, and performing the averages over Ω_j . The quantities $C^j(\Omega_j)$ are found from the transformation law of a fourth rank tensor, using a rotation matrix from any convenient choice of Euler angles. If the orientations of C^j are not uniformly distributed, the average over Ω_j in general involves multiple integration over three Euler angles. Numerous, efficient, approximate

techniques exist for such integrations.¹⁷ If the orientations are uniformly distributed, the result of the averaging is an isotropic fourth rank tensor. Such tensors have two independent elements, and these elements can be found quite simply from the scalar invariants of t^j : $t_{i i \ell \ell}^j$ and $t_{i \ell i \ell}^j$.

The principal computational difficulty is evaluating H^j . The strain field Green's function tensor must be found and then integrated over the appropriate volume for the given grain shape. Kneer has reduced this problem to quadrature. If we write H^j as

$$H_{pqrs}^j = -\frac{1}{2}(E_{qpsr} + E_{pqsr}),$$

then from Kneer we have

$$E_{pqrs}^j = A_j \int_0^\pi \sin\theta \, d\theta \int_0^{2\pi} d\phi \, D_{pr}^{-1}(\underline{k}) k_q k_s \quad (40)$$

where D_{pr}^{-1} is the inverse matrix of the Fourier transform of the Green's operator g_{pr} , (13), and

$$A_j = \frac{1}{8\pi^3 V_j} \int_0^\infty dk \int_{V_j} d\underline{r}' e^{-i\underline{k} \cdot \underline{r}'} \int_{V_j} d\underline{r} e^{i\underline{k} \cdot \underline{r}}$$

with k , θ , ϕ the spherical coordinates in $\underline{k}$ -space, k_i the Cartesian components of $\underline{k}$, and V_j the volume of the shape of grains of type j . For simple geometric shapes the evaluation of A_j is straightforward, and for some shapes it may be done exactly. The integrand in (40) is a homogeneous function of k of zero order. The integration then is that of a function of θ and ϕ over their solid angle. Again we point out that efficient numerical techniques exist for performing such multiple integrations.¹⁷ The symmetry of H^j is determined by the symmetry of C^* . For C^* isotropic, explicit expressions for H^j may be written if the grains are spherical or

ellipsoidal⁹. When C^* is cubic or hexagonal, Kneer gives more complete integrals for β_j^j for spherical grains. The orthotropic case for spherical grains is discussed by Morris.

We have carried out several series of calculations that assume grains spherically shaped. Effective stiffness constants were found for polycrystalline materials of one type of crystal, an aggregate of two types of crystals, and an aggregate of one type of crystal and one type of pore. The results are reported elsewhere.¹⁵ In Table I we do, however, give for polycrystalline Cu a comparison of our results with similar results of Kneer and Morris.

The second series of calculations is for C^* transverse isotropic. The effective stiffness constants of polycrystalline carbon were computed. This material is highly porous and has grains that have highly anisotropic elastic properties. The results will be reported in detail elsewhere.¹³ We found Kneer's expression for E_{pqrs} when C^* is transverse isotropic to be partially incorrect; corrections are provided in the appendix.

The computation scheme outlined is certainly not unique. Improvements to the scheme that are tailored to particular problems are possible. Our purpose has been merely to indicate a definite method that has been found to be practical. Ultimately, our numerical technique is similar to those already used by Kneer and by Morris. Thus the application of integral equation methods is no more difficult than that of the more intuitive approaches. The advantages are ones of formal definiteness and flexibility as already discussed in Section IV.

Acknowledgements

We acknowledge a grant to Cornell University from the Union Carbide Corporation and the Atomic Energy Commission for partial support of this work.

Appendix

Concerning only the case of hexagonal bulk material, we found several errors in the listings of Kneer's equations. These expressions as they stand do not reduce to the correct isotropic result.

The expression for β immediately after his equation 21 should read

$$\beta = ab - 2(a+b)c+3.$$

In his appendix the following should be substituted:

$$A_{1313}^{(0)} = C_{11}C_{66}/2$$

$$A_{1313}^{(1)} = -3C_{11}C_{66} + C_{44}(C_{11} + C_{66}).$$

$$A_{1313}^{(2)} = 3C_{11}C_{66} - 2C_{44}(C_{11} + C_{66}) + C_{44}^2/2$$

$$A_{1313}^{(3)} = -C_{11}C_{66} + C_{44}(C_{11} + C_{66}) - C_{44}^2/2.$$

Table I

Comparison of self-consistent effective stiffness constants^a of polycrystalline Cu.

	Experiment ^b	Kneer	Morris	Present work
Bulk modulus	---	13.764	13.75	13.764
Shear modulus	4.83	4.826	4.83	4.826

^aAll constants in 10^{11} dyne/cm².

^bG. Kneer, Phys. Status Solidi 3, K331 (1963).

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* This work has been partially supported by the U. S. Atomic Energy Commission under Contract No. AT(11-1)-3161, Technical Report No. COO-3161-24. Additional support was received from the Union Carbide Corporation.

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Figure Captions

1. Integral equation (16) for the strain field. (a) Iterative solution. (b) Diagrammatic representation.
2. Diagram representing repeated scattering from the same grain.
3. Diagram representing scattering by two different grains.
4. The single grain T-matrix. (a) Diagrammatic representation of the single grain T-matrix. (b) Definition of the ensemble average of (a).
5. Averaged additional corrections to elastic constant C^{Voigt} . (a) Kröner(1967). (b) Average T-matrix approximation, Eq. (30).
6. Self-consistent approximation. (a) An averaged, modified single grain T-matrix, denoted by a square, is defined using a double-lined propagator which sums all single grain T-matrix scattering at grains other than this one. (b) The self-consistent elastic constant, according to (18). This Procedure is correct no matter what C^0 is. If $C^0 = C_{\text{s.c.}}$ then $\langle t_{\text{s.c.}} \rangle = 0$. [For details see Reference 1, section III.]

$$\epsilon = \epsilon^0 + G\delta C\epsilon^0 + G\delta CG\delta C\epsilon^0 + \dots$$

$$\epsilon = \text{wavy line} + \overset{\text{X}}{\text{vertical dashed line}} \text{---} \text{wavy line} + \overset{\text{X}}{\text{vertical dashed line}} \text{---} \overset{\text{X}}{\text{vertical dashed line}} \text{---} \text{wavy line} + \dots$$

Figure 1

Figure 2

Figure 3

$$t^a = \begin{array}{c} x \\ | \\ \vdots \end{array} + \begin{array}{c} x \\ \triangle \end{array} + \begin{array}{c} x \\ | \\ \triangle \end{array} + \dots$$

a

$$\langle t^a \rangle = \begin{array}{c} o \\ | \\ \vdots \end{array}$$

b

Figure 4

Figure 5

$$\langle \begin{array}{c} \times \\ | \\ \hline \end{array} + \begin{array}{c} \times \\ \triangle \\ \hline \end{array} + \begin{array}{c} \times \\ \triangle \\ \hline \end{array} + \dots \rangle = \begin{array}{c} | \\ \hline \end{array}$$

a

$$\begin{array}{c} \square \\ | \\ \hline \end{array} - \begin{array}{c} \square \\ \triangle \\ \hline \end{array} + \begin{array}{c} \square \\ \triangle \\ \hline \end{array} - \dots$$

b

Figure 6