Effective properties of random elastic media(*)

E. KRÖNER (STUTTGART)

THE CONCEPT of "effective medium" is explained using elasticity for illustration. Examples of physical situations are described in which the effective medium theory is not applicable. The present stage of the theory is reviewed. Because of practical importance a unifying formula is discussed which allows us to calculate the bounds of Voigt, Reuss, Hashin-Shtrikman, third order as well as the self-consistent values of the effective elastic moduli with one single computer programme. Comparisons with experiments are reported and their merits and demerits are discussed.

Wyjaśniono koncepcję „ośrodka efektywnego" na przykładzie teorii sprężystości. Opisano przykłady fizyczne, do których teoria ośrodka efektywnego nie może być zastosowana. Zanalizowano obecny stan wiedzy w tej teorii. Biorąc pod uwagę ważność praktyczną teorii przedyskutowano szczególny związek, który pozwala określić granice poprawności dla efektywnych modułów sprężystości wyznaczonych dla modeli Voigta, Reussa, Hashina-Shtrikmana i teorii trzeciego rzędu na podstawie jednego programu numerycznego. Rezultaty obliczeń porównano z wynikami doświadczeń, dokonując oceny istoty tych ostatnich.

Выяснена концепция „эффективной среды" на примере теории упругости. Описаны физические примеры, для которых теория эффективной среды не может быть применена. Анализируется настоящее состояние знаний в этой теории. Имея в виду практическую важность теории, обсуждено частное соотношение, которое позволяет определить пределы правильности для эффективных модулей упругости определенных для моделей Фойхта, Рейсса, Хашин-Штрикман и теории третьего порядка на основе одной численной программы. Результаты расчетов сравнены с результатами экспериментов, проводя оценку сущности этих последних.

1. Introduction

IN THIS lecture we shall deal with the macroproperties of solid materials which, on a micro-scale, possess an at least partially random, or disordered, constitution. Polycrystalline aggregates and composites are prominent representatives of these materials to which also solid solutions, solid polymers, porous solids and other substances belong. Some of these materials have a regular, or ordered, constitution as well. It is important to realize that such materials have macroproperties which can deviate considerably from those of substances with a disordered constitution. Everybody knows about the distinct differences between the properties of (single-element) solids with crystalline arrangement of atoms and those with irregular arrangement of atoms, i.e. amorphous bodies. Composites which consist of a matrix in which inclusions of equal species are distributed regularly resemble crystal lattices. In particular, the periodic array of the inclusions causes physical fields that are described by Floquet, or Bloch, type solutions, similar to those occurring in the quantum theory of crystals. As a consequence, certain waves cannot propagate through the composite but suffer Bragg type reflections, in contrast to the situation with inclusions

(*) Lecture presented at the 19th Polish Solid Mechanics Conference.

that are distributed in an irregular fashion. Clearly, predictions on microscopically-ordered materials must be based on theories which are rather different from those to be used for disordered materials. Only the latter theories will be discussed in this lecture.

We furthermore restrict ourselves to the so-called linear properties. These are characterized by linear or linearized field equations and constitutive equations. Elasticity as well as viscoelasticity, electrical and thermal conductivity, thermal expansion, magnetic susceptibility and optical polarizability, as far as they can be handled in linear approximation, are all examples of such properties. Since this is a conference on solid mechanics we shall take our illustrations from the field of elasticity.

In certain situations linearization might not be admissible in dealing with the above mentioned properties. The theory to be discussed below will cover also such cases, provided the nonlinear problem can be solved by the iterative solution of a linear problem. This holds when the nonlinearity is sufficiently small.

Basically, nonlinear phenomena like fracture, fatigue, creep and plasticity require completely different methods which are by far not so well developed as those of the linear properties. Thus we shall omit these properties completely.

The number of papers on our subject which have been published since the pioneering work of W. Voight in 1887 is immense. In many of them the success of the theory is assessed by comparing it with experimental results. Such comparisons can, in fact, provide reliable support to a theory. On the other hand, they can be very misleading when they are applied uncritically which has been done but too often. The point is this: In order to derive theoretical predictions, which for comparison with experiment should be given in the form of numerical values of some physical quantities, for instance of elastic moduli, one has to specify quantitatively the conditions under which these predictions hold. Only if the experiments also match with these conditions can comparisons lead to reliable statements. Some of the conditions will now be discussed.

2. Critical remarks on experimental data for elastic moduli

Modern apparatus measure elastic moduli with an accuracy of one part in a thousand and better. Hence we need not be concerned about deviations between theoretical idealizations and experimental realities as long as such deviations remain below 0.1%. In the following we shall, however, also consider deviations of order per cent. Clearly, one has been to be very careful about these cases.

The material with random or partially random constitution will be called *macromaterial*. In the present discussion we restrict ourselves to macromaterials which consist of great numbers of continuously space-filling domains in all spatial directions. The properties of interest, for instance the elastic moduli, in a domain are called *local*. It is usually assumed that the local properties are distributed uniformly in each domain and that they change abruptly from one domain to its neighbour. The domains can be the grains, or crystallites, in a polycrystal, the inclusions and also the matrix in a composite of the matrix-inclusion type, the pores and the material part in a sponge-type structure etc. It is furthermore tacitly assumed in the problem discussed here that the interfaces between material domains do not open themselves nor allow gliding during the elastic deformation.

Real macromaterials do not satisfy all of the above conditions rigorously. Crystallites in a polycrystal or composite can be considered as elastic continua with uniform elastic moduli only if they are large enough so that the discreteness of the atomic scale does not come into play. Fortunately, this requirement is satisfied with an accuracy of better than 0.1% for almost all materials of practical interest.

The grain or phase boundaries between adjacent crystallites or phases, can be considered as shell-shaped inclusions with a thickness of the order of a few angstroms. They usually possess reduced elastic moduli. As long as the volume occupied by these boundaries is sufficiently small compared to the volume of the macromaterial, their contribution to the macrobehaviour remains below 0.1% unless opening or gliding occurs in the interface. Opening of phase boundaries is observed in certain composites. Crack opening is a related phenomenon. Grain boundary glide takes place at elevated temperature in many polycrystalline materials. The gliding is a viscous rather than an elastic process. In extreme situations these effects can lower the apparent elastic moduli as compared to the ideal ones by several per cent. Fortunately, it is usually possible to predict whether the one or the other phenomenon will occur in a certain physical situation.

More serious because rather universal is the effect of lattice defects, in particular dislocations, on the apparent elastic moduli. The constituents of most materials of practical importance are crystalline and hence are filled with numerous dislocations. Under applied stresses these dislocations bow out thereby producing a reversible glide. By this the material appears to be weaker than it would be in the absence of the dislocations. The effect on the shear moduli can easily be of the order of a few per cent as has been explained by N. F. MOTT [1] and J. FRIEDEL [2]. G. BRADFIELD and H. PURSEY [3] have developed a method by which the unwanted effect can be avoided. They prepare a specimen which contains in small concentration solute atoms of a different species precipitated along the dislocations. In this way, these are pinned down and become practically immobile under the applied (small) stress. Of course, the addition of foreign atoms itself changes the moduli. This effect, however, is often below 0.1% or, if not, can be corrected away by extrapolation to zero concentration.

Another effect which can be called the texture effect has also been treated by G. BRADFIELD and H. PURSEY [3]. Most calculated moduli of macromaterials hold for macroscopically isotropic materials. It is difficult, however, to prepare specimens which possess this property with an accuracy of more than a few per cent. This means that the test specimens usually have a slight texture which can be found out from X-ray measurements or from the direction-dependence of the gross elastic moduli. For this reason the experimental values need a texture correction which again can amount to a few per cent.

Since the corrections discussed are of the order a few per cent, it is these which determine the final reliability of the result. To give an example: K. A. GSCHNEIDER Jr. [4] gives the most likely value of the shear modulus of a macroscopically isotropic copper crystal as $4.51 \cdot 10^{11}$ dyn/cm^2. This value was deduced from the results of older publications which were not at all concerned about dislocation bowing out and texture. On the other hand, Bradfield and Pursey who took into account both effects obtained 4.83 instead of 4.51. An essential part of the difference can be attributed to the texture effect.

The discussion of this section was meant to show once more that reported good agree-

ment between theoretical and experimental results is not necessarily a proof that the theory
and experiment were good.

3. Micro- and macroinformation

As mentioned in the beginning, we restrict ourselves to materials of a statistical nature.
This means that the discussed theory of the macroproperties of such materials is basically
statistical. To say it in a more formal language: we are concerned with macromaterials
the constitution of which has an effectively ergodic character. As long as we restrict our-
selves to static situations, the ergodic hypothesis states that ensemble averages are equal
to averages over so-called macrovolume elements which are large as compared to the
dimensions of the fluctuations (grain size etc.) but small in comparison with the external
dimensions of the experiment. In such a situation differentiation and integration on the
macroscale can be defined and can be interchanged with ensemble averaging.

In order to make theoretical predictions we need certain information about the ma-
terial. According to a general theorem of the probability theory, it is possible to divide
the complete information which contains every detail of the constitution into micro-
and macroinformation. The latter is also called statistical information. Statistical theory
means that the microinformation is discarded completely. This is done because no interest
lies in details which can be observed on the microscale only. For instance, for the macro-
behaviour it is irrelevant whether four grains of particular lattice orientation join at point **r**.
Only macroscopic statements like "the majority of the grains is lengthy" or "almost all
grain boundaries are plane" count.

By omitting the microinformation we pass over to statistics. As a consequence, our
predictions lose their deterministic character and assume the status of expectations. This
means that deviations from the predictions will occur which, however, become all the more
unimportant the larger the number of statistical elements, here the number of grains etc., is.

We shall see that the statistical information itself can be immensely complex so that
it is not possible to incorporate it completely when going to quantitative predictions.
Omitting a part of the statistical information means that expectations can no longer be
calculated sharply. One rather has to rely on approximations or try to find bounds for the
expectation. The latter possibility has the advantage that rigorous results can be obtained.
The well-known Voigt and Reuss bounds for the elastic moduli of macroscopically iso-
tropic materials are examples of rigorous results. The information needed to calculate
them consists in the 1-point probability density for the local elastic moduli. The Voigt
and Reuss bounds are the best bounds derivable with this information.

If the 2-point probability density, which also includes the information of the 1-point
density, is given, then the bounds can be drawn closer. The occurrence of the probability
densities can be used for a classification of bounds. Those derived with the 1-point density
are the first order bounds, those with the 2-point density are the second order bounds and
those with the 3-point density are the third order bounds. More generally, if the n-point
probability density is given where n assumes the values 1, 2, 3, ..., then one can derive
bounds of order n.

The problem can now be stated in a quantitative way as follows: Given the n-point probability density for the distribution of the local material property, find the closest bounds for the desired expectations. Here "closest" means "closest under the given information".

In the above considerations the set of n-point probability densities can be replaced by the set of n-point correlation functions which are defined by means of the probability densities and contain the same information. For instance, the 2-point correlation of the local tensor c of elastic moduli is defined as

$$\langle c_1 c_2 \rangle = \int c_1 c_2 P(c_1, c_2) dc_1 dc_2,$$

where $c_1 = c(r_1)$, $P(c_1, c_2)$ is the 2-point probability density for c and the integrations go over all values of c.

The probability densities and the correlation functions are very natural means for describing the internal structure of a macromaterial. Consider, for instance, a polycrystal in which many lengthy grains are aligned in some preferred direction. Then the probability of finding two points r_1 and r_2 in the same grain is comparatively large when the connection line of the two points is parallel rather than perpendicular to the preferred direction. In other words, the probability density, thus also the correlation function, reflects the anisotropy of the grain arrangement in a quantitative manner.

Similarly, the 3-point probability density gives information about lengthy grains which are not aligned but rather possess an axes distribution over all spatial directions. Plane grain boundaries show themselves in the 4-point probability density etc.

Of course, the macroinformation need not be given in terms of probability densities or correlation functions. But these certainly form the most systematic tool available at present. There exists, however, a whole branch of science called stereology which is concerned with the macrodescription of microstructures. These approaches which emphasize the geometrical or topological feature of those structures certainly are interesting and deserve attention. The inclusion of this subject into the present lecture would, however, go far beyond the allotted space and time.

4. The concept of effective medium

So far we have formulated our problem of deriving from some information either approximate values or bounds for certain expectations, i.e. average outcomes of experiments. We now have to specify quantitatively the experiments for which we are to calculate expectations. We are mainly interested in materials which are non-uniform on a microscale but homogeneous on the macroscale. It is then suggestive to assume that with respect to macroresponse the material behaves as if it were completely homogeneous. This, however, is not generally true. An important example where it is not true is the propagation of elastic waves in macromaterials. Such waves suffer multiple diffractions and reflections at the grain or phase boundaries. In the wave equation of the mean field these effects are represented by extra terms, namely by mixed correlation functions between local moduli and local field. Thus the effective wave equation of the macromaterial is different from that of a homogeneous material. Hence also the solutions are different in the two cases.

On the other hand there are situations where the macromaterial does, in fact, behave like a homogeneous material as we shall explain in the following. Assume that the local material property is represented by a linear operator $\mathscr{L}$. The operator is supposed to be the same for all times t but to depend on the space points $\mathbf{r}$ in a random manner. Next we introduce a field u which is also random in space. We consider u to be caused by volume and surface sources as well as by initial values. To the entity of the sources and initial values we refer as the source field f. By definition, the operator $\mathscr{L}$ when applied to u yields f [1]

$$(4.1) \qquad \qquad \mathscr{L}u = f.$$

We treat situations in which the field u follows uniquely from the sources f and where $u = 0$ for $f = 0$. Under these conditions a unique Green operator, also called the Green function $\mathscr{G}$, can be defined as $\mathscr{L}^{-1}$ so that

$$(4.2) \qquad \qquad u = \mathscr{G}f.$$

For illustration let us consider an elastic medium which is deformed by a volume force density $\mathbf{F}(\mathbf{r}, t)$, a surface force density $\mathbf{A}(\mathbf{r}, t)$, an initial displacement field $\mathbf{u}(\mathbf{r}, 0) \equiv \mathbf{u}_0(\mathbf{r})$ and an initial velocity field $\dot{\mathbf{u}}(\mathbf{r}, 0) \equiv \mathbf{v}_0(\mathbf{r})$. This problem is known as the combined initial and Neumann problem. The field u is now written as $\mathbf{u}(\mathbf{r}, t)$. Equation (4.2) then takes the detailed form [2]

$$(4.3) \quad \mathbf{u}(\mathbf{r}, t) = - \int_0^t dt' \int_V dV' \mathscr{G}(\mathbf{r}, t; \mathbf{r}', t') \cdot \mathbf{F}(\mathbf{r}', t') + \int_0^t dt' \int_S dS' \mathscr{G}(\mathbf{r}, t; \mathbf{r}', t') \cdot \mathbf{A}(\mathbf{r}', t)$$

$$+ \int_V dV' \left\{ \mathscr{G}(\mathbf{r}, t; \mathbf{r}', 0) \cdot \mathbf{v}_0(r') - \mathbf{u}_0(r') \cdot \left[\frac{\partial}{\partial t'} \mathscr{G}(\mathbf{r}, t; \mathbf{r}', t') \right]_{t'=0} \right\},$$

where $\mathscr{G}$ is the Green's function belonging to the present initial and boundary conditions. Observe that in this case the right hand side of Eq. (4.2) is not a simple convolution.

We now consider ensemble averaged fields $\langle u \rangle$ which are caused by spatially random sources f. In accordance with our statistical concept we shall not know all the details about the source distribution but rather the ensemble average $\langle f \rangle$ only. We say that the material behaves as an effective medium with respect to the physical experiments under investigation if and only if $\langle f \rangle$ determines $\langle u \rangle$ completely. In that case $\langle u \rangle$ does not depend on the fluctuations $f' \equiv f - \langle f \rangle$ of the sources. In the following, primes shall denote deviations from the mean except when they are applied to $\mathbf{r}$ and t.

In order to find a criterion for a material to behave effectively in the above sense we take the ensemble average of Eq. (4.2) writing

$$(4.4) \qquad \qquad \langle u \rangle = \langle \mathscr{G} \rangle \langle f \rangle + \langle \mathscr{G}f' \rangle.$$

The last term does not contribute if and only if

$$(4.5) \qquad \qquad \langle \mathscr{G}f \rangle = \langle \mathscr{G} \rangle \langle f \rangle.$$

---

[1] Operators are written in script letters.

[2] For the derivation of this formula cf. P. M. Morse and H. Feshbach [5]. The classical uniqueness proof originates from F. Neumann. Instead of the Neumann problem one can also consider the Dirichlet or the mixed problem.

Then f is not correlated with $\mathscr{G}$, hence not correlated with the material parameters. Equation (4.5) is always satisfied if f is non-random.

A different approach towards the condition (4.5) for the effective medium to exist starts from the definition

$$(4.6) \qquad \langle \mathscr{L}u \rangle = \mathscr{L}^{\text{eff}} \langle u \rangle$$

of the effective operator $\mathscr{L}^{\text{eff}}$. With $u = \mathscr{G}f$ and $\mathscr{G} = \mathscr{L}^{-1}$ one finds

$$(4.7) \qquad \langle f \rangle = \mathscr{L}^{\text{eff}} \langle \mathscr{G} \rangle \langle f \rangle + \mathscr{L}^{\text{eff}} \langle \mathscr{G}f' \rangle.$$

Since $\langle f \rangle$ is independent of f', the last term, thus also $\langle \mathscr{G}f' \rangle$, must vanish as before. Equation (4.7) shows that $\langle \mathscr{G} \rangle$ is the Green function belonging to $\mathscr{L}^{\text{eff}}$. Observe that $\mathscr{L}^{\text{eff}} \neq \langle \mathscr{L} \rangle$.

If $\langle \mathscr{G}f' \rangle$ does not vanish, i.e. if Eq. (4.5) is not satisfied, then Eq. (4.7) contradicts Eq. (4.6). Hence Eq. (4.5) is the consistency condition for the existence of an effective operator. For our Eq. (4.3) the consistency condition assumes the form

$$(4.8)_1 \qquad \langle \mathscr{G} \cdot \mathbf{F} \rangle = \langle \mathscr{G} \rangle \cdot \langle \mathbf{F} \rangle, \qquad \langle \mathscr{G} \cdot \mathbf{A} \rangle = \langle \mathscr{G} \rangle \cdot \langle \mathbf{A} \rangle,$$

$$(4.8)_2 \qquad \langle \mathscr{G} \cdot \mathbf{u}_0 \rangle = \langle \mathscr{G} \rangle \cdot \langle \mathbf{u}_0 \rangle, \qquad \langle \mathscr{G} \cdot \mathbf{v}_0 \rangle = \langle \mathscr{G} \rangle \cdot \langle \mathbf{v}_0 \rangle.$$

Let us now turn to elastostatics. Here the Green function becomes time-independent. Hence the time integrations and the last integral can be omitted in Eq. (4.3). If there are no volume forces, then the consistency condition is $\langle \mathscr{G} \cdot \mathbf{A} \rangle = \langle \mathscr{G} \rangle \cdot \langle \mathbf{A} \rangle$. Assume that the surface forces have a fluctuating part $\mathbf{A}'$ which is correlated with the elastic moduli. Evidently, $\mathbf{A}'$ changes the sign over distances of the correlation length, for instance some grain diameter. Then, in accordance with Saint-Venant's principle, the stress and strain fields decay towards the interior over a distance of this order of magnitude. If the grains etc. are very small compared to the size of the body, the contribution to $\langle \mathbf{u} \rangle$ becomes negligible. In this manner the condition of consistency looses its relevance: random elastic media behave as an effective medium under surface loads as long as the correlations of the elastic moduli are of sufficiently short range.

Volume forces are mostly gravitational. They have a fluctuating part $\mathbf{F}'$ if the mass density fluctuates. This occurs in composites but not in polycrystals. It is rather clear from Eq. (4.3) that $\mathbf{F}'$, when it is correlated with $\mathscr{G}$, contributes to $\langle \mathbf{u} \rangle$. Hence a medium with fluctuating mass density does not behave as an effective medium under gravitational forces.

We now pass to elastodynamics. The above statements concerning $\mathbf{A}'$ and $\mathbf{F}'$ remain valid. In the discussion of the consistency conditions we then concentrate on Eq. $(4.8)_2$. The important question is whether initial fluctuations $\mathbf{u}_0'$ and $\mathbf{v}_0'$, unlike $\mathbf{A}'$, contribute to $\langle \mathbf{u} \rangle$ at later times. Assume that $\mathbf{F}$, $\mathbf{A}$ as well as $\mathbf{v}_0$ and $\mathbf{u}_0$ vanish at $t = 0$, whereas $\mathbf{u}_0'$ does not vanish. Such a situation can be thought to be produced by applying fluctuating volume forces $\mathbf{F}$ which are suddenly removed at $t = 0$. The exact solution for $\langle \mathbf{u} \rangle$ can be taken from Eq. (4.3) as

$$(4.9) \qquad \langle \mathbf{u}(\mathbf{r}, t) \rangle = - \int_V dV' \left\langle \mathbf{u}_0'(\mathbf{r}') \cdot \left[\frac{\partial}{\partial t'} \mathscr{G}(\mathbf{r}, t; \mathbf{r}', t') \right]_{t'=0} \right\rangle.$$

The function $\mathbf{u}_0'(\mathbf{r}')$ can be chosen in such a way that at an arbitrary fixed space-time point $\mathbf{r}, t$ the integrand is positive or negative definite. Then $\langle \mathbf{u} \rangle$ does not vanish for that

point and, of course, not for neighbouring points either. We interpret this by saying that a ⟨**u**⟩-wave develops as a consequence of the initial distribution. Thus $\mathbf{u}_0'$, and similarly $\mathbf{v}_0'$, if they are correlated with $\mathscr{G}$, contribute to ⟨**u**⟩, in contrast to **A**'. In other words: in dynamical situations a random elastic medium does not behave as an effective medium. One finds that wave propagation in such media is an extremely involved phenomenon.

5. Results of effective medium theory. Elastostatics

In the general theory of Green functions there is a rigorous operator equation, due to F. J. Dyson, which expresses one Green function, $\mathscr{G}$ say, in terms of another Green function, $\mathscr{G}^0$ say:

$$(5.1) \qquad \mathscr{G} = (\mathscr{I} + \mathscr{G}^0 \delta\mathscr{L})^{-1}\mathscr{G}^0, \qquad \delta\mathscr{L} \equiv \mathscr{L} - \mathscr{L}^0.$$

Here all quantities are operators of the same operator space. $\mathscr{I}$ is the unit operator. Equation (5.1) is easily derived from the equations $\mathscr{L}\mathscr{G} = \mathscr{I} = \mathscr{L}^0\mathscr{G}^0$.

We now identify $\mathscr{L}$ with the statical operator of a random elastic medium and $\mathscr{L}^0$ with a simpler, namely non-random, operator which will be fixed later. Averaging Eq. (5.1) and taking the inverse gives us $\mathscr{L}^{\text{eff}}$. If the correlations of the elastic moduli are short range, i.e. microscopic, then $\mathscr{L}^{\text{eff}}$ can be represented in terms of an effective elasticity tensor operator $\mathscr{C}^{\text{eff}}$ in exactly the same fashion as $\mathscr{L}$ is represented by the local tensor $\mathscr{C}$. One finds (C. Eimer [6], R. Zeller and P. H. Dederichs [7] and others)[3]

$$(5.2) \qquad \mathscr{C}^{\text{eff}} = \langle c\mathscr{A}\rangle\langle\mathscr{A}\rangle^{-1}, \qquad \mathscr{A} \equiv (\mathscr{I} + \Gamma^0 \delta c)^{-1}$$

where Γ^0 is a modified Green function defined by

$$(5.3) \qquad \Gamma^0_{ijkl}(\mathbf{r}, \mathbf{r}') = \partial_j \partial_l' \mathscr{G}_{ik}(\mathbf{r}, \mathbf{r}')|_{(ij),(kl)},$$

(ij) and (kl) denote symmetrization in ij and kl.

Provided the geometric series contained in Eq. (5.2) converges, Eq. (5.2) is equivalent to

$$(5.4) \qquad \mathscr{C}^{\text{eff}} = \mathscr{C}^0 + \langle \delta c\rangle - \langle c'\Gamma^0 c'\rangle + \langle c'\Gamma^0 \delta c\Gamma^0 c'\rangle$$
$$- \langle (c'\Gamma^0 \delta c)'\Gamma^0 (\delta c\Gamma^0 c')'\rangle + \langle (c'\Gamma^0 \delta c)'\Gamma^0 \delta c\Gamma^0 (\delta c\Gamma^0 c')\rangle - \ldots + \ldots$$

As before, the primes denote deviations from the mean, $\mathscr{G}^0 \equiv (\mathscr{L}^0)^{-1}$ is formed with $\mathscr{C}^0$ and $\delta c \equiv c - \mathscr{C}^0$. The $\mathscr{C}^{\text{eff}}$ given by Eq. (5.4) is an infinite series the general term of which can be written down but is of no interest here. Equation (5.4) represents a formal solution of the problem to find $\mathscr{C}^{\text{eff}}$ when the information on c is given by the full set of correlation functions including ⟨c⟩. Observe that the Γ^0 are non-random so that they can be taken outside of the (ensemble) average brackets in Eq. (5.4) which then produce the correlation functions of increasing order. Equations (5.2) and (5.4) are formal rather than explicit solutions because they contain multiple integrals which cannot easily be calculated for order higher than 3.

As to the convergence of the series in Eq. (5.4) we can say that it always converges for macroisotropic polycrystals consisting of crystallites with cubic lattice symmetry [8].

---

[3] In Eqs. (5.2) and (5.4) all c's and Γ^0's are to be understood as operators. For instance, $c = c(\mathbf{r}')\delta(\mathbf{r}, \mathbf{r}')$ where $c(\mathbf{r}')$ is the local tensor of elastic moduli at point $\mathbf{r}'$. Multiplication with Γ^0 implies an integral operation.

It is highly probable that it converges even for all polycrystals. The series does not converge for composites in which the constituents have very different stiffnesses. The most spectacular example is that of porous media.

As has been proved by P. H. DEDERICHS and R. ZELLER [9], an upper bound to $\mathscr{C}^{\text{eff}}$ is obtained if, regardless of the chosen $\mathscr{C}^0$, the series in Eq. (5.4) is truncated after any term of odd order in δc and c'. For instance, $\mathscr{C}^0 + \langle \delta c \rangle = \langle c \rangle$ is the upper bound of first order. In fact, it is the Voigt bound which we also denote by $\mathscr{C}^{(1)}$.

In the following we restrict ourselves to polycrystals but keep in mind that corresponding results apply to composites. In order to calculate the bound $\langle c \rangle$ from c we must know the texture weight function([4]) for the averaging as well as the elastic moduli of the crystallite. We shall assume further that the crystallite moduli are available if we know the material species. To give an example for the weight function: this function is completely specified by saying that it is isotropic. Such a statement does not imply, though, that the polycrystal is isotropic. In fact, a polycrystal with isotropic texture function can well be anisotropic. It is known that this happens when the grain shape distribution is anisotropic, an effect which shows itself in the correlation functions of second and higher order only.

If not even the weight function is known, i.e. if we only know that we have a polycrystal of copper, say, then we can still construct an upper bound which, of course, lies above $\mathscr{C}^{(1)}$ and will be called $\mathscr{C}^{(0)}$. In fact, we know that $\mathscr{C}^0 + \langle \delta c \rangle$ is an upper bound. Apparently, $\mathscr{C}^0$ itself is an upper bound provided that $\langle \delta c \rangle \leqslant 0$, i.e. $\mathscr{C}^0 \geqslant \langle c \rangle$. The size of two 4th rank tensors $\mathscr{C}$ and c, say, which can be represented by 6×6 Voigt type matrices $(\mathscr{C}_{ij})$ and (c_{ij}) can be compared by means of the corresponding quadratic forms. Then $\mathscr{C} > c$ implies that

$$(5.5) \qquad \mathscr{C}_{ii} > c_{ii}, \qquad \mathscr{C}_{ii}\mathscr{C}_{jj} - \mathscr{C}_{ij}^2 > c_{ii}c_{jj} - c_{ij}^2$$

(no summation, $i, j = 1, 2, 3, 4, 5, 6$) with the addition that some but not all of these 21 quantities may also be equal.

Since we do not possess the information necessary for calculating $\langle c \rangle$, we must do this with the most unfavourable weight functions, i.e. with those yielding the largest possible $\langle c \rangle$. These are obtained for polycrystals that are effectively monocrystals the preferred axes of which might have any orientation with respect to the axes of the specimen, for instance a rod axis. If for all these orientations $\mathscr{C}^0 \geqslant \langle c \rangle$ is to hold and if the optimum, i.e. lowest, bound is wanted, then $\mathscr{C}^0$ must be isotropic such that the $\mathscr{C}_{ii}^0$ and $\mathscr{C}_{ii}^0\mathscr{C}_{jj}^0 - \mathscr{C}_{ij}^0\mathscr{C}_{ij}^0$ are equal to the largest corresponding values the local elastic moduli c_{ij} can possibly assume. The $\mathscr{C}^0$ (uniquely) defined by this prescription, which likewise applies to composites, is equal to the above mentioned $\mathscr{C}^{(0)}$, the zeroth upper bound. It plays a significant role in the bound theory of Z. HASHIN and S. SHTRIKMAN [10]; see also L. J. WALPOLE [11] and refs. [8, 9].

Having defined $\mathscr{C}^{(0)}$ we can extend the mentioned bound theorem of Dederichs and Zeller as follows: If the series (5.4) is truncated after any term of even order in δc ad c', and if $\mathscr{C}^{(0)}$ is substituted for $\mathscr{C}^0$, then we obtain an upper bound.

([4]) This function is usually given in terms of generalized spherical harmonics. Since harmonics of order higher than 4 do not contribute in the averaging of a 4th rank tensor we need not know the full but only a partial texture function.

We mention in passing that, following M. J. BERAN and J. MOLYNEUX [12], the bounds of third and higher order constructed in this way can be somewhat improved by the introduction of free variational parameters.

From second order on the terms of the series (5.4) contain integrals which become increasingly involved and have been done only for relatively simple forms of the correlation functions. An important class of materials for which this is possible are those graded according to the three macroproperties: homogeneity, isotropy and disorder. The following discussion applies to polycrystals with a macroisotropic grain shape and size distribution. Extensions towards aggregates with grain shape anisotropy seem to be possible but have not yet been developed. The grading is done by means of integers l, m and n, respectively. This scheme implies that the considered property shows itself in the correlation function up to the order given by the particular number. The property disorder by definition excludes any principle of order in the statistical constitution of the material. For instance, a principle of order for all correlation functions is implied if the distribution of the local moduli is correlated with the grain shapes or sizes. Thus a polycrystal in which the large or elongated grains are in average harder than the other grains is not disordered in the above sense. The other characteristics of disorder concern the grain topology alone. Disorder of grade 3 requires that particular grain shape(s) or size(s) which are distinguishable in macroexperiments must not exist in the considered polycrystal.

Let us return to Eq. (5.2). It is easy to decompose the modified Green function Γ^0 into parts one of which is a delta function part $E\delta(\mathbf{r}, \mathbf{r}')$ where E is a constant tensor of 4th rank. In the isotropic case E is determined by

$$(5.6) \qquad E_{ijkl} = [-(3k+4\mu)\,\delta_{ij}\,\delta_{kl} + 9(k+2\mu)\,I_{ijkl}]/15\mu(3k+4\mu).$$

k and μ are compression and shear modulus of the tensor $\mathscr{C}^0$ ([5]). If the polycrystal possesses a well-defined texture, then the tensor E becomes anisotropic and can be calculated by using a computer programme of F. GHAHREMANI [13]. The tensor E is named P in Ghahremani's work. Since the grain shape statistics assumed here is isotropic, one has to consider spherical inclusions in this programme.

If $E\delta(\mathbf{r}, \mathbf{r}')$ is substituted for Γ^0 in Eq. (5.2), we obtain

$$(5.7) \qquad C = \langle ca \rangle \langle a \rangle^{-1}, \qquad a \equiv (I + E\delta c)^{-1}.$$

This formula is very helpful. It comprises the most important bounds of the effective medium theory. Which particular bound is obtained depends on the choice of C^0. Recall that C^0 determines E and dc. The results are given in Table 1.

Table 1

$C^0 =$	∞	$C^{(0)}$	$C^{(1)}$	$C(SC)$	$C_{(1)}$	$C_{(0)}$	O
$C =$	$C^{(1)}$	$C^{(2)}$	$C^{(3)}$	$C(SC)$	$C_{(3)}$	$C_{(2)}$	$C_{(1)}$
$n =$	1	2	3	(3)	3	2	1

([5]) Remember that $\mathscr{C}^0 = C^0\delta(\mathbf{r}, \mathbf{r}')$.

Observe that both C^0 and C decrease monotonically when going from left to right. $C^{(n)}$ and $C_{(n)}$ are respectively upper and lower optimum bounds for materials which are classified as overall grade n, meaning $l = m = n$. $C^{(1)}$, $C_{(1)}$, $C^{(2)}$, $C_{(2)}$ bear extra names; they are respectively called the bounds of Voigt, Reuss and Hashin-Shtrikman. $C(SC)$ is the author's self-consistent tensor of elastic moduli [14] for which we have inserted $n = (3)$ in order to indicate that $C(SC)$ is a good approximation for materials which are (at least) overall grade 3. In fact, $C(SC)$ always lies between the bounds of these materials. The equality $C^0 = C$ expresses the condition of self-consistency in the calculation of $C(SC)$.

The proofs for the results given in Table 1 can be found in the works of various authors. We mention J. L. WALPOLE [11], P. H. DEDERICHS and R. ZELLER [9] and E. KRÖNER [8, 15]. Note that the gradation scheme in [15] is changed slightly compared to that originally introduced in [8]. In order to find the lower bounds it was necessary to consider compliances rather than moduli. In fact, in an analogous manner as we worked with distributions of local moduli one uses distributions of local compliances and derives upper bounds for effective compliances. After inversion these bounds turn into lower bounds of the effective moduli.

The bounds given by Eq. (5.7) in connection with Table 1 are rigorous within the initially discussed model. A pleasant feature of these results is that a whole set of important bounds can be calculated with only one computer programme. If the material is not disordered in the above defined sense, then it is much more difficult to obtain rigorous results. Such cases occur frequently in composites. One then depends upon reasonable assumptions or guesses. It seems that also in this field the work has become more systematical. For a recent review see J. P. WATT, G. F. DAVIES and R. J. O'CONNELL [16].

6. Conclusion

Following the scheme of Table 1 we show in Fig. 1 the bounds of first, second and third order as well as the self-consistent value of the effective shear modulus G of polycrystalline copper. For completeness, also the bounds of order 0 are included. The single crystal values used in the calculation are G. Bradfield's $c_{11} = 16.9$, $c_{12} = 12.2$, $c_{44} = 7.55$, all numbers in 10^{11} dyn/cm^2. For comparison with experiment we have chosen Bradfield's result 4.83 for the effective shear modulus, not because is compares so well but rather because this author took extreme care in avoiding the disturbing effects discussed in Sect. 2. In particular, Fig. 1 conveys a good impression of how much narrowing of the bounds can be achieved by passing from bounds of lower order to those of higher order.

Note that all bounds are optimum in the sense that they are the narrowest possible bounds under the given information, i.e. the overall grade. It is sensible also to speak of an overall grade 0 in the case of $C^{(0)}$ and $C_{(0)}$. In fact, the assumptions leading to these bounds do not contain any requirement on the homogeneity, isotropy or disorder. The fact that the bounds are optimum implies that they are also exact. Since they all relate to different amounts of information, we do not call any of them better than any other. In fact, narrowness is no measure of the quality of bounds so long as these are optimum.

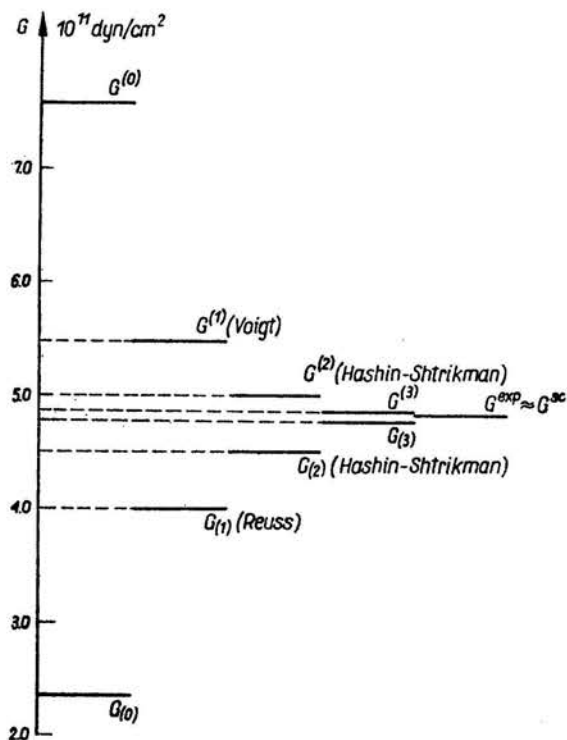Figure 2 shows how well the experimental shear modulus G^{exp} of the polycrystals falls

FIG. 1. Calculated bounds of the effective shear modulus G of macroisotropic polycrystalline copper. Experimental values for mono- and polycrystal from Bradfield as quoted by G. KNEER [17].
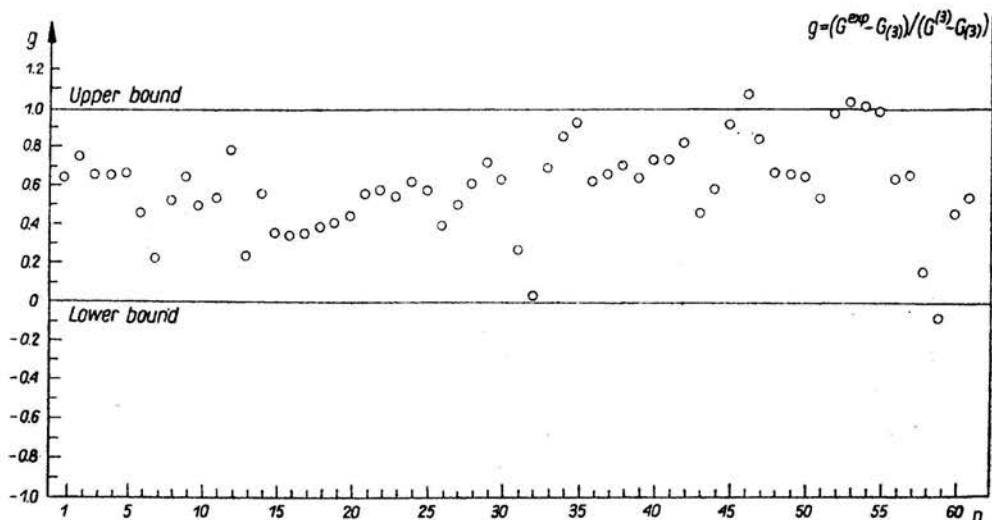

FIG. 2. Normalized third order bounds and experimental values of the shear modulus of various polycrystalline alloys of copper, silver and iron. n denotes the number of the specimen. From H. H. WAWRA, H. KOCH and E. KRÖNER [18].

[196]

within the third order bounds. We have used scaled units for the measured shear moduli so that the third order bounds are the same for all materials. In these units the ordinate measures $g = (G^{exp} - G_{(3)})/(G^{(3)} - G_{(3)})$ so that G^{exp} coincides with $G_{(3)}$ if $g = 0$ and with $G^{(3)}$ if $g = 1$. The abscissa gives the number of the polycrystalline specimen which can be identified by consulting the original paper of H. H. WAWRA, H. KOCH and E. KRÖNER [18]. In selecting experimental data we have once more used the criterium of dislocation and texture corrections.

The striking impression of Fig. 2 is that the experimental values scatter over the whole interval between the two bounds. From this we conclude that in spite of the endeavour of the experimentalist it was not possible to prepare specimens which were homogeneous isotropic and disordered of a grade higher than 3. In fact, Fig. 1 suggests that bounds of fourth order would be much closer than those of third order so that most of the experimental values shown in Fig. 2 would lie outside the fourth order bounds. In view of this prediction the extreme numerical effort the calculation of fourth order bounds would need does not seem worthwhile, at least not at the present stage of preparational techniques.

In conclusion, I should like to apologize to all workers in the field whose valuable work I could not mention due to the shortage of space and time.

References

1. N. F. MOTT, Phil. Mag., 43, 1151, 1952.
2. J. FRIEDEL, Phil. Mag., 44, 444, 1953.
3. G. BRADFIELD and H. PURSEY, Phil. Mag., 44, 437, 1953.
4. K. A. GSCHNEIDER Jr., in: Solid State Physics, eds. F. SEITZ and D. TURNBULL, Academic Press, Vol. 16, 275, New York 1964.
5. P. M. MORSE and H. FESHBACH, Methods of theoretical physics, vol. 1, McGraw Hill, New York 1953.
6. C. EIMER, Arch. Mech., 19, 521, 1967; 20, 87, 1968.
7. R. ZELLER and P. H. DEDERICHS, Phys. Stat. Sol., (b) 55, 831, 1973.
8. E. KRÖNER, J. Mech. Phys. Solids, 25, 137, 1977.
9. P. H. DEDERICHS and R. ZELLER, Z. Physik, 259, 103, 1973.
10. Z. HASHIN and S. SHTRIKMAN, J. Mech. Phys. Solids, 11, 127, 1963.
11. L. J. WALPOLE, J. Mech. Phys. Solids, 14, 151, 1966.
12. M. J. BERAN and J. MOLYNEUX, Q. Appl. Math., 24, 107, 1966.
13. F. GHAHREMANI, Mech. Res. Comm., 4, 89, 1977.
14. E. KRÖNER, Z. Physik, 151, 504, 1958.
15. E. KRÖNER, Mech. Res. Comm., 4, 389, 1977.
16. J. P. WATT, G. F. DAVIES and R. J. O'CONNELL, Rev. Geophys. Space Phys., 14, 541, 1976.
17. G. KNEER, Phys. Stat. Sol., 9, 825, 1965.
18. H. H. WAWRA, H. KOCH and E. KRÖNER, Jap. J. Mat. Sci. Soc., 14, 131, 1977.

INSTITUT FÜR THEORETISCHE UND ANGEWANDTE PHYSIK, DER UNIVERSITÄT
UND MAX-PLANCK-INSTITUT FÜR METALLFORSCHUNG, STUTTGART, BRD.

http://rcin.org.pl