

Ordered versus disordered materials: consequences for the continuum model(*)

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In this paper the class of solid materials is divided into perfectly ordered, partially disordered and perfectly disordered materials. After discussing the propagation of elastic waves it is found that the frequency spectra of these groups of materials exhibit remarkable differences which can be observed in macroscopic experiments.

W niniejszej pracy klasę ciał stałych podzielono na idealnie uporządkowane, częściowo nieuporządkowane i idealnie nieuporządkowane. Przedyskutowano zagadnienie rozprzestrzeniania się fal sprężystych i wykazano, że widma częstości tych grup materiałów wykazują istotne różnice, które mogą być obserwowane w doświadczeniach makroskopowych.

В настоящей работе класс твердых тел разделен на идеально упорядоченные, частично неупорядоченные и идеально неупорядоченные тела. Обсуждена задача распространения упругих волн и показано, что спектры частот этих групп материалов обладают существенными разностями, которые могут наблюдаться в макроскопических экспериментах.

1. Introduction

ACTUAL materials are built up from huge numbers of tiny physical particles such as atoms and molecules. These particles like to arrange themselves into configurations in such a manner that the free energy of this physical system becomes a minimum, i.e., it reaches an equilibrium state. This statement applies whether or not the system is subordinated to constraints of any kind. It is clear that the presence of such constraints increases the free energy as compared to the unconstrained situation. The constraints may also be of such a nature that they prevent the system from reaching the equilibrium within the time of observation. In such cases one often has to deal with metastable states.

Under Helmholtz conditions free energy has the form $F = U - TS$. A decisive feature of this formula is the minus sign. As a rule, well-ordered configurations of particles have low values of the internal energy U and entropy S , whereas heavily disordered configurations possess high values of these quantities. In addition, the higher is the temperature, the more effective will the given value of the entropy be in determining the configuration of the body. For this reason, the tendency is to have ordered configurations at lower temperatures and disordered ones at higher temperatures.

The order-disorder phenomena can be observed on various scales in solids. The greatest possible disorder in the sense of entropy occurs on the atomic scale. Complete disorder on this scale exists in the sense of an idealization only because even the most

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amorphous bodies contain a certain short-range order. This range can be from one to a few atomic distances.

Distinct from these amorphous bodies are media that possess a far-range order when measured on the atomic scale. The range of this order is usually limited by certain elements of disorder such as dislocations in crystals and grain and phase boundaries in polycrystalline and composite materials. One expects the behaviour of such materials to depend on length parameters which are of the order of 10^{-5} – 10^{-4} cm in the case of a dislocated metal crystal and of an order larger than this, but small when compared with the external dimensions of the workpiece, in the case of polycrystals and composites. Finally, the perfectly ordered body, i.e., the perfect crystal, is again fiction, as we have said it with respect to for the perfectly disordered body⁽¹⁾.

The large scale disorder discussed above does not contribute appreciably to the entropy as it is clear intuitively. It is for this reason that we shall focus our attention on polycrystals and composites. If we restrict ourselves further to reversible deformations, i.e., to elasticity, then we also need not consider the vibrational part of the entropy and the problem becomes a purely mechanical.

Within this restriction to polycrystals and composites we shall distinguish materials with ordered and disordered constitutions. We shall reserve the term "ordered" for structures that can completely be constructed by periodic repetitions of some elementary domain, just as a crystal can be built up by periodic repetitions of an elementary atomic cell. Thereby, we allow the periodicity to be interrupted at the surface of the body. Many composites of technical importance are, in fact, constructed in such a manner. Disordered materials are either partially disordered or perfectly disordered. The latter notation is reserved for materials the constitution of which does not contain any principle of order. To give two examples: a 2-phase composite which consists of a matrix with inclusions is never perfectly disordered because the assertion that one of the two constituents forms the matrix and the other the inclusions implies a principle of order. A Poisson cell material often used to characterize a polycrystal is never perfectly disordered because the cell boundaries of such a material are plain, what again implies a principle of order.

Since partially and perfectly disordered materials are best treated within the same theoretical framework, it seems adequate to comprise these materials under the heading of disordered materials. By doing this we have implicitly decided to speak of a grade of disorder rather than of a grade of order. Incidentally, this corresponds to the common understanding of entropy as a measure of disorder and not of order.

The aim of this paper is to discuss to what extent the grade of disorder on the microscopical scale, as we shall call it, influences the macroscopical elastic properties of a material. It is quite obvious that these properties will change very little from a specimen which is perfectly ordered to a specimen which is *almost* perfectly ordered. Distinct differences, if existent, will appear above all if we compare perfectly ordered with considerably disordered materials. Thus, we shall focus our attention on this difference.

The question to be discussed in this paper can also be put as follows: is it possible

(¹) We shall not discuss many other more or less ordered material structures that exist in nature and are used in practice. As an example let us just mention the great variety of high polymers.

to distinguish between ordered and disordered materials by macroscopic measurements of the elastic properties? We shall deal with this question by comparing elastic wave propagation in the two kinds of materials, because wave propagation is a relatively transparent phenomenon that reflects the totality of the dynamical properties of a material in a very natural manner. Since so much is well known about waves in ordered materials our main effort is directed towards an understanding of the main features of elastic waves in disordered media. There is a vast literature in this field to which I certainly cannot do justice in this brief paper.

For a thorough review of the work done up to 1968 the reader is referred to an excellent article by U. FRISCH [1] who, among others, states (on p. 77) that the mathematical difficulties are very large "so that, at present, a rigorous study of wave propagation in random media is out of the question". To my knowledge, this situation has not changed since 1968. In fact, there are many more open than solved problems in this field.

2. Ordered materials

The grade of disorder we have talked about is a quantity that typically refers to the bulk of the material. It is therefore adequate to assume, as has been indicated before, that all characteristic lengths of the material are microscopic, namely, very small when compared to the external, i.e. macroscopic dimensions of the body. For general investigations it is then sensible to apply periodic boundary conditions with the ordered body, whereas the disordered body can, with some precaution, be taken to be infinite.

In this section we shall consider the ordered body. As it has been mentioned it is most convenient, for our purpose, to deal with elastic wave propagation. If we recall the pattern of the theory of electrons in crystals we can predict that many results of this theory will be valid for ordered composites, too. Of course, there are also differences due to the fact that electrons obey Schrödinger's wave equation, whereas here we are dealing with elastic wave propagation.

The most important result with respect to our problem is that there exist FLOQUET or BLOCH waves in our ordered medium and that these possess a frequency spectrum which is divided into bands such that frequencies falling within the bands specify possible elastic waves, whereas those falling between the bands, in the so-called frequency gaps, are forbidden in the sense that no waves with such frequencies can exist. There is also some similarity to the phonon spectra of the lattice theory in which so-called acoustical and optical bands with gaps in between are distinguished in the frequency spectrum. The apparent difference in the number of bands is due to the fact that, n being the number of particles in the elementary cell, there are $3n$ degrees of freedom for these particles. This leads to $3n$ bands in the case of phonons but to an infinite number of bands in the case of electrons and of ordered continua in which the number of degrees of freedom per elementary domain is unlimited.

One of the properties of the band structure of the frequency spectrum is that waves with a given wavelength can propagate with different frequencies. This statement is valid for arbitrary wavelengths, in particular for very long ones. Hence, the $3(n-1)$ optical branches of a phonon frequency spectrum certainly have spatially macroscopic aspects.

A further feature of such frequency spectra consists in the fact that there exists a smallest possible wavelength, namely $2a$, if a is the distance of periodicity in the concerned direction of the cell, or domain structure. Such features of the frequency spectra of electrons and phonons have a great influence on the related physical phenomena. It would then be very surprising if a corresponding statement were not to apply to elastic continua that are ordered on a larger scale.

3. Disordered media

3.1. Preliminaries about the statistical theory

In our introduction we contrasted ordered with disordered bodies. Should we emphasize more the mathematical nature of the problem, we could speak of deterministic and stochastic media instead. The two pairs of notions are not exactly equivalent. In fact, any elastic problem of a disordered body can be solved, at least in principle, if sufficient information about the disorder is given in a deterministic way. Such possibilities, however, are excluded from our considerations because our aim is a macroscopic description of materials that are ordered or disordered on a microscopic scale. In the case of ordered materials, as defined in Sect. 1, we can give a macroscopic deterministic characterization of the periodicity which defines the order. Hence, the mathematical problem is deterministic by its very nature.

The mentioned aim of our investigation makes it senseless to describe any deviation from the defined ordered body in deterministic terms. This implies that in the specification of disordered materials we renounce certain microscopic information and content ourselves with a characterization of the disorder in macroscopic, i.e., statistical terms. As an immediate consequence we must restrict ourselves to situations in which an ergodic hypothesis can be applied; this essentially means that the medium is random enough so that differentiations and integrations on the macroscale have a meaning. Single experimental results can then only be predicted in the form of expectation values or, to put it in a different way, our results apply to statistical ensembles rather than to selected specimens. The ergodic hypothesis also implies that ensemble averages are equal to averages over so-called representative or macroscopic volume elements which are large enough to contain the whole statistics of the constitution in themselves, but are, at the same time, very small when compared with the external dimensions of the body. If the material in question does not allow for the definition of such volume elements, then the situation is not sufficiently stochastic and must be excluded from our further investigations.

3.2. Basic equations; condition of consistency for effective operators

Let us give briefly some of the basic equations of the elasticity theory of disordered media. First we write the equations governing the behaviour of a linearly elastic, inhomogeneous medium in the short (operator) form

$$(3.1) \quad Lu = f.$$

The Eq. (3.1) and the linear operator L are, as discussed in Ref. [2], supposed to comprise the (volume) equations of motion, the surface conditions, e.g. those of the infinite body or of the 1st or 2nd boundary value problem (BVP), and the initial conditions, for instance those giving the displacement field u and the velocity $v = \partial u / \partial t$ at some time, too. The physical meaning of the corresponding parts of the "source field" f is easily recognized: the volume part represents the density of the volume forces, the surface part is zero (infinite body) or represents either the surface displacements (1st BVP) or the density of the surface forces (2nd BVP); the initial part comprises the displacements and particle velocity at time t_0 . Bearing this in mind we are in a position to demand that the Eq. (3.1) has a unique solution which we write by means of the well-known Green operator G of the (inhomogeneous) elastic medium as

$$(3.2) \quad u = Gf, \quad LG = I,$$

where I is the unit element of our operator space.

The material information is contained in L or in $G = L^{-1}$. In statistical problems we shall now consider, we renounce the microscopic details so that we do not know L and G . If f is non-random, then, from the Eq. (3.2)₁ we obtain

$$(3.3) \quad \langle u \rangle = \langle G \rangle f.$$

The operator $\langle G \rangle^{-1}$ is also called the effective operator and denoted by L^{eff} . In fact, if the Eq. (3.1) is averaged and compared with Eq. (3.3) one finds with $\langle f \rangle = f$ that

$$(3.4) \quad \langle Lu \rangle = L^{\text{eff}} \langle u \rangle.$$

This equation is often used as a definition of L^{eff} . The other common definition of L^{eff} is

$$(3.5) \quad \langle uLu \rangle = \langle u \rangle L^{\text{eff}} \langle u \rangle.$$

It is consistent with the definition (3.4) if the left-hand side of the Eq. (3.4), when multiplied by $\langle u \rangle$, is equal to that of the Eq. (3.5). Using $Lu = f$ once more, one obtains in this way the condition of consistency in the form

$$(3.6) \quad \langle u \rangle \langle f \rangle = \langle uf \rangle.$$

Note that in our present notation both products in (3.6) involve an integration over the volume of the body. If the Eq. (3.6) is valid, then, the fluctuations of u do not contribute to the average $\langle uf \rangle$ which is essentially the total energy content of the body. Then, the mean field $\langle u \rangle$ contains the whole energy. This situation is encountered in the ordinary elastostatics of disordered media, but not in the theory of elastic wave propagation.

The statement of the Eq. (3.6) is that u be uncorrelated with f . With $u = Gf$, the Eq. (3.6) becomes $\langle Gf \rangle \langle f \rangle = \langle Gff \rangle$. This is satisfied if, and only if, f is non-random. In the case of free wave propagation the volume and surface parts of f vanish identically, thus obeying the condition of consistency. As to the initial condition we can, of course, assume u and v to be non-random at time t_0 . This is a somewhat unpleasant restriction which one might like to remove in more quantitative investigations. As a passing remark let us add that the condition of consistency, in particular its form (3.6), is related to a condition used by HILL [3] in his theory of bounds for elastic moduli of heterogeneous media.

3.3. Energy flux in disordered media

Situations in which the condition of consistency is satisfied are much simpler than other situations because, according to the Eq. (3.3), the solutions do employ the average $\langle G \rangle$ in the place of the full statistical information about the Green operator G . Nevertheless, even these simpler situations may be extremely involved as this can be seen in the case of the propagation of free waves in our disordered medium. The reason for the complication here is that the volume part of

$$(3.7) \quad L^{\text{eff}} \rightarrow \left(\varrho \frac{\partial^2}{\partial t^2} - \partial_j c_{ijkl}(\mathbf{r}) \partial_i \right)^{\text{eff}}$$

in volume, where ϱ is the mass density and $c_{ijkl}(\mathbf{r})$ are the elastic moduli at point \mathbf{r} , cannot be decomposed into a kinetic part $L_{\text{kin}}^{\text{eff}}$ and a potential part $L_{\text{pot}}^{\text{eff}}$. Such partial operators, if they existed, should be defined by

$$(3.8) \quad \langle L_{\text{kin}} u \rangle = L_{\text{kin}}^{\text{eff}} \langle u \rangle, \quad \langle L_{\text{pot}} u \rangle = L_{\text{pot}}^{\text{eff}} \langle u \rangle.$$

However, they do not satisfy the condition of consistency in the volume. In fact, we have for instance

$$(3.9) \quad L_{\text{pot}} u = -L_{\text{kin}} u,$$

where $-L_{\text{kin}} u$ are the inertia forces that produce the field u at time t . These forces are always random functions of position in our problem. Now, if $L_{\text{kin}}^{\text{eff}}$ and $L_{\text{pot}}^{\text{eff}}$ do not exist, then there are no ϱ^{eff} and c_{ijkl}^{eff} either⁽²⁾. As a consequence there is no wave which propagates with the wave speed $\sqrt{c^{\text{eff}}/\varrho^{\text{eff}}}$, where c^{eff} is an elastic modulus for a longitudinal or transversal wave.

In order to obtain some insight into the real situation let us consider the energies involved. According to a well-known formula, the energy flux $\Phi(t)$ at time t through a closed surface S inside the body is

$$(3.10) \quad \Phi(t) \equiv - \int dV \partial_j (\sigma_{ij} v_i) = \partial_t \int dV w(\mathbf{r}, t), \quad w(\mathbf{r}, t) \equiv \frac{1}{2} (p_i v_i + \sigma_{ij} \varepsilon_{ij}),$$

where $\partial_t \equiv \partial/\partial t$ and σ_{ij} , ε_{ij} are the components of stress and strain, respectively; $p_i \equiv \varrho v_i$ is the density of momentum and the integration of the energy density w extends over the volume V bounded by S . From our outline of the ergodic hypothesis it follows that for any random function $f(\mathbf{r}, t)$ the equality $\int dV f(\mathbf{r}, t) = \int dV \langle f(\mathbf{r}, t) \rangle$ holds⁽³⁾. Applying this to the Eq. (3.10) we obtain

$$(3.11) \quad - \int dV \partial_j \langle \sigma_{ij} v_j \rangle = \partial_t \int dV \frac{1}{2} \langle p_i v_i + \sigma_{ij} \varepsilon_{ij} \rangle,$$

where we have interchanged differentiation and averaging in accordance with the ergodic hypothesis.

⁽²⁾ In monophasic polycrystals there is random c but constant ϱ which can then be called ϱ^{eff} . The case with random ϱ and constant c is seldom encountered. ϱ^{eff} and c^{eff} exist simultaneously if, and only if, the medium is nonrandom.

⁽³⁾ If f is identified with the energy density w , then, the equation means that there is no energy deficit when passing from a deterministic to a stochastic description.

It can easily be shown that the condition of consistency, if it were valid for L_{kin}^{eff} and L_{pot}^{eff} , would imply that

$$\langle p_i v_i \rangle = \langle p_i \rangle \langle v_i \rangle \quad \text{and} \quad \langle \sigma_{ij} \varepsilon_{ij} \rangle = \langle \sigma_{ij} \rangle \langle \varepsilon_{ij} \rangle,$$

the latter being the original Hill condition applied to the case of random media. Since, as we have argued, these conditions are not valid in our application we conclude that there does not exist by itself an elastic wave with particle velocity $\langle v_i \rangle$, stresses $\langle \sigma_{ij} \rangle$ and strains $\langle \varepsilon_{ij} \rangle$.

Volume integrals over averages of products as they occur in the Eq. (3.11) can be decomposed according to a formalism described earlier in ref. [4]. Let a, b, g and h be four random scalar or tensorial functions of position; furthermore, let a, b be stationary and ab constant. We are now interested in the integral

$$(3.12) \quad \int dV_1 \langle g(1)h(1) \rangle = \int dV_1 \langle g(1) \rangle \langle h(1) \rangle + \int dV_1 \langle g'(1)h'(1) \rangle,$$

where the primes, as throughout this paper, denote deviations from the mean so that, for example, $g' = g - \langle g \rangle$, $\langle g' \rangle = 0$. In this consideration we shall replace, for reasons of simplicity, the position vectors r_1, r_2 , etc. by the corresponding numbers 1, 2, etc. Since, under the ergodic hypothesis

$$(3.13) \quad ab = \frac{1}{V} \int dV_2 \langle a(2)b(2) \rangle = \frac{1}{V} \int dV_2 \langle a(2) \rangle \langle b(2) \rangle + \frac{1}{V} \int dV_2 a'(2)b'(2)$$

the last integral in the Eq. (3.12) is

$$(3.14) \quad \int dV_1 \langle g'(1)h'(1) \rangle = \frac{\langle a \rangle \langle b \rangle}{ab} \int dV_1 \langle g'(1)h'(1) \rangle + \frac{1}{ab} \int dV_1 \langle g'(1) \rangle \frac{1}{V} \int dV_2 a'(2)b'(2)h'(1).$$

Now we have $\langle a \rangle \langle b \rangle \neq ab$ and, under the ergodic hypothesis, averaging and differentiating may be interchanged so that we can rewrite the Eq. (3.14) as

$$(3.15) \quad \int dV_1 \langle g'(1)h'(1) \rangle = \frac{1}{AV} \int \int dV_1 dV_2 \langle g'(1)a'(2)b'(2)h'(1) \rangle = \frac{1}{AV} \int \int dV_1 dV_2 \langle g'(1)a'(2) \rangle \langle b'(2)h'(1) \rangle + \langle (g'(1)a'(2))' (b'(2)h'(1))' \rangle,$$

where $A \equiv ab - \langle a \rangle \langle b \rangle$. The same procedure can be repeated as long as one wishes and with the final result

$$(3.16) \quad \int dV_1 \langle g(1)h(1) \rangle = \int dV_1 \bar{g}(1)\bar{h}(1) + \frac{1}{AV} \int \int dV_1 dV_2 \bar{g}(1, 2)\bar{h}(1, 2) + \frac{1}{A^2V^2} \int \int \int dV_1 dV_2 dV_3 \bar{g}(1, 2, 3)\bar{h}(1, 2, 3) + \dots,$$

where for the sake of simpler representation we have introduced the abbreviations

$$(3.17) \quad \begin{aligned} \bar{g}(1) &\equiv \langle g(1) \rangle, & \bar{g}(1, 2) &\equiv \langle g'(1)a'(2) \rangle, & \bar{g}(1, 2, 3) &\equiv \langle (g'(1)a'(2))' a'(3) \rangle \dots \\ \bar{h}(1) &\equiv \langle h(1) \rangle, & \bar{h}(1, 2) &\equiv \langle b'(2)h'(1) \rangle, & \bar{h}(1, 2, 3) &\equiv \langle b'(3)(b'(2)h'(1))' \rangle \dots \end{aligned}$$

etc. The \bar{g} - and \bar{h} -quantities are cross correlation functions between g and a on the one hand and h and b on the other hand. They are linear in g and h .

The formalism will now be applied to the Eq. (3.11) which is possible without any restriction in spite of the occurrence of the differentiation in the first integral. For simplicity, we shall soon omit all tensor indices. If we identify the random functions a, b, g, h according to

$$(3.18) \quad g \rightarrow \sigma_{ij}, \quad a \rightarrow c_{pqrs} \varrho, \quad h \rightarrow v_j, \quad b \rightarrow \varrho c_{pqrs}^{-1}$$

and if we make appropriate use of the bar notation explained by the Eqs. (3.17), we obtain for the left-hand side of the Eq. (3.11)

$$(3.19) \quad - \int dV \partial \langle \sigma v \rangle = - \int dV_1 \partial^{(1)} [\bar{\sigma}(1) \bar{v}(1)] - \frac{1}{AV} \int \int dV_1 dV_2 \partial^{(1)} [\bar{\sigma}(1, 2) \bar{v}(1, 2)] \\ - \frac{1}{A^2 V^2} \int \int \int dV_1 dV_2 dV_3 \partial^{(1)} [\bar{\sigma}(1, 2, 3) \bar{v}(1, 2, 3)] - \dots,$$

where $\partial^{(1)}$ means differentiation with respect to point 1. The choice of a and b in (3.18) is somewhat arbitrary. We have chosen what, to our mind, is the most natural combination of ϱ and c_{ijkl} .

Performing the $\partial^{(1)}$ -differentiations in the Eq. (3.19) and using the equation of motion

$$(3.20) \quad \partial_t p_i - \partial_j \sigma_{ij} = 0$$

we find for the energy flux $\Phi(t)$ through the surface S of the arbitrary volume V that

$$(3.21) \quad \Phi(t) = \partial_t \left\{ \int dV_1 \frac{1}{2} \langle \bar{p}(1) \bar{v}(1) + \bar{\sigma}(1) \bar{\varepsilon}(1) \rangle \right. \\ \left. + \frac{1}{AV} \int \int dV_1 dV_2 \frac{1}{2} \langle \bar{p}(1, 2) \bar{v}(1, 2) + \bar{\sigma}(1, 2) \bar{\varepsilon}(1, 2) \rangle \right. \\ \left. + \frac{1}{A^2 V^2} \int \int \int dV_1 dV_2 dV_3 \frac{1}{2} \langle \bar{p}(1, 2, 3) \bar{v}(1, 2, 3) + \bar{\sigma}(1, 2, 3) \bar{\varepsilon}(1, 2, 3) \rangle + \dots \right\}.$$

This equation is also obtained if the decomposition formalism is directly applied to the right-hand side of the Eq. (3.11).

3.4. General features of the elastic wave spectrum in disordered materials

The important point of the rigorous equations (3.19) and (3.21) is that the total energy flux appears in a decomposed form, namely as contributions from various motions. The wave corresponding to the leading term will be called the main wave whereas the other terms arise from waves that we call correlation waves (of increasing order) because the oscillating quantities $\bar{u}(1, 2)$, $\bar{u}(1, 2, 3)$ etc. are cross correlation functions. The fundamental question now is whether these waves can travel separately by themselves, a situation on which we cannot decide from the appearance of the Eqs. (3.19) and (3.21) alone. From our observation, that in the case of wave propagation in random media $L_{\text{kin}}^{\text{eff}}$ and $L_{\text{pot}}^{\text{eff}}$ do not exist in the volume, we have concluded that always $\langle p_i v_i \rangle \neq \langle p_i \rangle \langle v_i \rangle$ and $\langle \sigma_{ij} \varepsilon_{ij} \rangle \neq \langle \sigma_{ij} \rangle \langle \varepsilon_{ij} \rangle$. This means that at least the main wave does not propagate alone, by itself. If, say, at some time t_0 , such a wave were to be enforced by a suitable distribu-

tion of external forces which are then suddenly removed, one would observe that the correlation waves would build up very quickly on costs of the energy of the main wave. This fact can be attributed to coupling terms in the equations of motion which follow, for instance, from the equation for the energy flux in the usual manner. We then expect that due to this coupling single correlation waves cannot travel alone by themselves, either. If such a wave existed at time t_0 , then it would give away parts of its energy in order to build up other wave modes, among them the main wave. These observations suggest that there exists an interaction between the various wave modes such that energy is exchanged between them and a dynamical equilibrium is established.

This picture does not exclude the possibility that any one of the waves has a particularly large amplitude compared to the others. I hesitate to predict anything about the existence, the wavelengths and frequencies of such waves. In analogy to wave propagation in ordered media, it is tempting to compare the three main waves with those of the three acoustical branches and the correlation waves with those of the optical branches of various order. The analogy would be clearest if we allowed the correlation waves also to travel with long wavelengths and to possess minimum frequencies which increase along with increasing order of the correlation. It may be plausible that this picture is not too far away from the truth if the medium is near the ordered state.

Now let us discuss the question of frequency gaps. In ordered materials these gaps are understood to result from the so-called Bragg reflections which plane waves suffer if they propagate through equally spaced reflecting planes. If the wavelength and the angle of coincidence satisfy the Bragg condition, then there are so many coherent reflections at the planes that the wave dies out exponentially over a few plane spacings.

One may now conceive of introducing some disorder in such a way that either the plane spacings become somewhat random or the distinguished planes cease to be good. The latter situation can be imagined easily if these planes are formed by a sort of lattice planes of regularly arranged inclusions. If either the regularity in position is given up or the shapes vary from one inclusion to the other, then we no longer have good lattice planes. We expect that in such a situation the stopping of waves in the Bragg region is less pronounced; this means that the waves, which satisfy the Bragg condition only approximately but still cannot pass through the ordered material, may be able to come through the corresponding disturbed medium. We may then say that the transition from perfect order to partial disorder has weakened the frequency gap without removing it completely.

Should the disorder be rather large then the picture might change completely. There is nothing left which resembles the Bragg reflections discussed before. Nevertheless, the fact remains that waves of certain wavelengths cannot pass through the medium because there are too many reflections along the inclusion or cell boundaries. These reflections become effective when the wavelength approaches the characteristic disorder parameters of the material and remain effective at a smaller wavelength. This prediction is supported by strong evidence in the almost historic Christiansen filter experiment many students of physics at my university had to perform during their studies. This experiment concerns light waves rather than sound waves. For our conclusion, however, this difference is not relevant. The Christiansen filter consists of a pulverized glass filled with some transparent fluid which has an index of refraction similar to that of the glass particles. Glass and

fluid together form a disordered two-phase system, the fluid representing the matrix and the glass particles the inclusions.

The experiment shows that light waves with wavelengths that are smaller than the characteristic disorder length do not pass through except if the index of refraction of the fluid is changed, for instance by raising the temperature, in such a way that it approaches that of the glass. In such a case the fluctuations of the index of refraction become so small that the system behaves like a homogeneous medium.

There is no reason why the medium should become transparent again if one moves towards smaller and smaller wavelengths except if the index of refraction in the two phases changes in such a manner that it becomes practically the same. This does in fact occur if one passes to x-rays. A corresponding phenomenon does not exist in the case of sound waves. Hence we conclude that waves below a critical wavelength cannot propagate through a highly disordered material.

On the other hand, there is no reason why waves of larger wavelengths should not propagate through such a medium. Since increasing frequency means decreasing wavelength we expect that, at sufficiently low frequencies, an entity of coupled waves can propagate through the medium. The distribution over the various wave modes of the energy will depend on the frequency. At high frequencies no wave propagation is possible. Between low and high frequencies there may exist a more or less extended transition region.

4. Conclusion

Having as our aim the exposition of differences in the macroscopic behaviour of materials which are ordered or disordered on a microscopic scale we have discussed the problem of wave propagation in those two kinds of materials. The striking feature is that the sound wave frequency spectrum of ordered materials possesses a band structure which exhibits distinct frequency gaps, i.e., bounded frequency ranges in which wave propagation is not possible. Such gaps do not seem to exist in perfectly disordered materials which, therefore, do not possess a band structure. Insofar as partially disordered materials are concerned the tendency is that the frequency gap weakens more and more along with increasing disorder. These results are also significant for long wavelengths and therefore imply that differences in the macroscopic dynamical behaviour of ordered and disordered materials can be detected by purely macroscopic experiments.

A further important difference of ordered and disordered elastic media consists in the fact that single wave modes can exist in ordered media, whereas in the disordered media many wave modes are always coupled together in a sort of dynamical equilibrium. As a result, the main wave can be described in some approximation by a complex propagation vector the imaginary part of which describes the flux of energy into the correlation modes. This has been shown by F. C. KARAL and J. B. KELLER [5].

The discussion has given rise to a number of questions. It would be particularly desirable to clarify to what extent the waves of the optical branches of ordered materials are analogous to the correlation waves in partially disordered materials.

We have not discussed the important issue of convergence of developments such as (3.19) and (3.21). The result of a recent investigation (see Ref. [2]) is that the convergence is always ensured in polycrystals whereas there may be divergence in the case of composites. The criterium is whether the relative fluctuations $\langle c \rangle^{-1} c'$ of the elastic moduli are smaller or larger than ≈ 1 . In polycrystals they are smaller than ≈ 1 because all cells consist of the same material and, therefore, have limited fluctuations.

I should like to conclude this paper with the following final remark: Most of the predictions made are rather qualitative and many of them were based on physical intuition. It was my attempt to make clear that there are many interesting unsolved problems in the field of disordered materials, particularly that of wave propagation. Further investigations have to be more quantitative. For this purpose one can derive coupled wave equations for the main wave and the correlation waves, for instance starting from the equation for the energy flux. The equations become much simpler if one considers one-dimensional problems, for example wave propagation in rod-shaped, one-dimensionally disordered materials. Unfortunately, the solutions of such one-dimensional problems may be, even qualitatively, very different from those of three-dimensional ones. For reference see for instance P. DEAN [6].

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