Invariance conditions in the transmission from discrete lattice models to the continuum limit

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CERTAIN statements on the macroscopics properties of crystals can be derived from invariances against certain symmetry transformations. This has been shown for elastic properties and also for the elastic limit of lattice theory in simple cases. In this paper we derive such statements in more general cases and show that a unique and consistent transition from lattice theory including internal degrees of freedom is possible only if the conditions of translational and rotational invariance are satisfied.

Z niezmienniczości względem pewnych przekształceń symetrycznych można wysnuć niektóre wnioski o makroskopowych własnościach kryształów. Wykazano to w prostych przypadkach dla własności sprężystych, jak również dla granicy sprężystości teorii strukturalnej. W niniejszej pracy wyprowadzono takie wnioski w przypadkach bardziej ogólnych i pokazano, że jednoznaczne i konsekwentne przejście od teorii strukturalnej, włączając wewnętrzne stopnie swobody, jest możliwe tylko wtedy, gdy spełnione są warunki niezmienniczości względem translacji i obrotu.

Из инвариантности по отношению к некоторым преобразованиям симметрии можно вывести некоторые следствия о макроскопических свойствах кристаллов. Это показано в простых случаях для упругих свойств, как тоже для предела упругости структурной теории. В настоящей работе выведены такие следствия в более общих случаях и показано, что однозначный и последовательный переход к структурной теории, включая внутренние степени свободы, возможен только тогда, когда удовлетворены условия инвариантности по отношению к трансляциям и вращениям.

Introduction

INVARIANCE conditions play an important role in physics; this has been known for a long time. Nevertheless, they are sometimes disregarded, what often leads to incorrect conclusions. In general, such conditions are related to symmetries of space and time and give statements about physical quantities.

In the elastic theory the invariance conditions are generally satisfied mainly by choosing appropriate variables which are invariant by themselves, such as the finite strain or corresponding variables for other degrees of freedom. But even in the elastic theory rotational invariance is not always taken into account, and it is only in recent years that some of its consequences have been discussed.

The main concern of my discussion is lattice theoretical aspect of such invariances where the elastic theory is the limiting case. Therefore, some of the elastic statements are required. On the other hand, invariance conditions in the lattice theory have very often been disregarded; the aim of this contribution is to stress its importance in general cases.

As a rule there are a number of symmetries and invariance conditions which set restrictions to physical constants. Some of them are related to lattice symmetries, others to space inversion (parity) and time reversal, which may equally be essential. But in this report we will consider the influence of translational and rotational invariance; the latter has sometimes been omitted in lattice theory.

The importance of such conditions in lattice theory was first pointed out by K. HUANG and M. BORN [1–5]. In their approach to the continuum limit they did not even use the invariance conditions because they postulated certain symmetries of lattice sums. But these symmetries can be related to invariances. It is only with these conditions that there can be unique and consistent way to the elastic limit. This has been shown for lattices of mass-points [4, 5]. More complicated systems with internal degrees of freedom of the constituents have not been discussed. Having thus introduced some elastic and lattice theoretical foundations we shall deal with some extensions of the usual approach. Objections have been raised against rotational invariance in the lattice theory and it was even suggested to cancel such well-known principles as of virtual work. But it should be pointed out strongly that the condition of rotational invariance, for example, has to be used explicitly only in the case of models (like Born-von Kármán) in the lattice theory. However, if one starts with a given potential energy of the whole system, then these conditions are satisfied identically and no explicit use has to be made. But in model calculations they are required. Sometimes, it is also convenient to use them in cases of a given potential in order to derive general statements without using the potential explicitly. We will discuss rotational invariance in connection with homogeneous (compact) systems mainly.

This is even more important in lattices with defects or at the surfaces of lattices because one can use it, for example, in deciding whether a certain model is stable or not. However, most effects can be stretched by means of the simpler discussion mentioned.

1. Continuum foundations of invariances

In the continuum limit one first has to specify the appropriate variables; these variables must not only describe the elastic deformation of the continuous medium, but also the magnetization, polarization and, if present, the external fields acting on the body. The displacements of every volume element of the medium are described by the field $\mathbf{u}(\mathbf{X})$ in the Lagrangian way, where $\mathbf{X}$ is the material coordinate (in the initial state). A deformation is then described by the displacement gradient (distorsion) which is different from zero:

$$(1.1) \qquad \frac{\partial u_i}{\partial X_k} = \frac{\partial x_i}{\partial X_k} - \delta_{ik} = u_{i|k} = v_{ik} \neq 0, \quad v_{ik} \neq v_{ki}.$$

Here, x_i is the space coordinate of the volume element after deformation and $v_{ik}(\mathbf{X})$ describes the state of the deformed medium if we restrict ourselves only to compact media with sufficient slowly varying fields. The energy density of the medium can then be expanded with respect to the v_{ik} and the other possible variables

$$(1.2) \qquad w = w(v_{ik}, m_i, h_k),$$

where we have introduced a dependence on magnetization and magnetic field. It may also depend on $m_i|_k$ and further variables which we will not discuss since they can be handled in a similar way.

Rotational and translational invariance means that (1.2) may not change under a rotation or translation of the space coordinate system (provided, of course, there is no preferred direction in space). Let D be the rotation matrix, then by rotation[1]

$$\mathbf{x} \Rightarrow \mathbf{x}' = D\mathbf{x}, \quad v_{ik} \Rightarrow v'_{ik} = D_{im}\frac{\partial x_m}{\partial X_k} - \delta_{ik} = D_{im}v_{mk} + D_{ik} - \delta_{ik}.$$

For most purposes it is sufficient and easier to consider infinitesimal rotations defined by

(1.4) $$D_{ik} = \delta_{ik} + \Omega_{ik}, \quad \Omega_{ik} = -\Omega_{ki}$$

and

(1.5) $$x'_i = x_i + \Omega_{ik}x_k, \quad v'_{ik} = v_{ik} + \Omega_{im}v_{mk} + \Omega_{ik},$$
$$m'_i = m_i + \Omega_{ik}m_k, \quad h'_i = h_i + \Omega_{ik}h_k.$$

Then (1.2) must be independent of D or Ω for all possible deformations v_{ik} and m_i, h_k. The variables given do not change with a translation. But if there are such variables, say $u_i^{(\alpha)}$ with

(1.6) $$u_i^{(\alpha)} \Rightarrow u_i^{(\alpha)\prime} = u_i^{(\alpha)} + t_i \quad \text{for all } \alpha$$

under a translation of the coordinate system, then (1.2) has to be invariant of t_i too.

We use an arbitrary expansion of (1.2) just to compare it with lattice theory. This expansion might be[2]

(1.7) $$w = S_{ik}v_{ik} + \frac{1}{2}S_{ik,jl}v_{ik}v_{jl} + P_i m_i + N'_{i,jl}m_i v_{jl} + \frac{1}{2}D_{ik}m_i m_k$$
$$+ \frac{1}{2}V'_{ik,jl}m_i m_k v_{jl} + M_i h_i + L'_{i,jl}h_i v_{jl} + G_{ik}h_i m_k + \frac{1}{2}T'_{ik,jl}h_i m_k v_{jl} + \ldots$$

where, of course in many cases, some terms might be equal to zero. The first two terms represent elastic deformations, the others give the dependence on further variables and interactions.

Now, if we insert (1.5) into (1.7) and remember that w must be independent of Ω_{ik} for all the possible v_{ik}, m_i, h_i, we obtain

$$(1.8)_{1-6} \quad \left.\begin{array}{l} S_{jl} \\ S_{ik,jl} + S_{jk}\delta_{il} \\ N'_{i,jl} + P_j\delta_{il} \\ L'_{i,jl} + M_j\delta_{il} \\ V'_{ik,jl} + D_{jk}\delta_{il} + D_{ij}\delta_{kl} \\ T'_{ik,jl} + G_{jk}\delta_{il} + G_{ij}\delta_{kl} \end{array}\right. \begin{array}{l} = \\ = \\ = \\ = \\ = \\ = \end{array} \left\}\begin{array}{l} \\ \text{symmetric} \\ \text{against} \\ \text{interchange} \\ \text{of } j, l \\ \end{array}\right. \left.\begin{array}{c} 9 \to 6 \\ 45 \to 21 \\ 27 \to 18 \\ 27 \to 18 \\ 54 \to 36 \\ 81 \to 54 \end{array}\right\}\begin{array}{l} \\ \text{number} \\ \text{of coefficients} \\ \end{array}$$

+ relations for higher order coefficients.

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[1] We always use summation convention for the lower indices.

[2] One should not drop the commas in such coefficients because the properties of symmetry depend essentially on the "position of the comma".

These relations reduce the number of independent constants essentially[3]. This can be accounted for by dividing the coefficients into their symmetric and antisymmetric parts, i.e., we put

(1.9)

$$S_{ik,jl} - S_{kl}\delta_{ij} \quad = C_{ik,jl} = C_{ik,lj} = C_{ki,jl},$$

$$\frac{1}{2}(N'_{i,jl} + N'_{i,lj}) \quad = N_{i,jl} = N_{i,lj},$$

$$\frac{1}{2}(N'_{i,jl} - N'_{i,lj}) \quad = \frac{1}{2}(P_l\delta_{ij} - P_j\delta_{il}),$$

$$\frac{1}{2}(V'_{ik,jl} + V'_{ik,lj}) = V_{ik,jl} = V_{ik,lj},$$

$$\frac{1}{2}(V'_{ik,jl} - V'_{ik,lj}) = \frac{1}{2}(D_{lk}\delta_{ij} + D_{il}\delta_{jk} - D_{jk}\delta_{il} - D_{ij}\delta_{kl}),$$

where use has been made of (1.8). If we insert this into the expansion (1.7), we obtain a new expansion with symmetric variables and symmetric coefficients:

$$(1.10) \quad w = S_{ik}\eta_{ik} + \frac{1}{2}C_{ik,jl}\eta_{ik}\eta_{jl} + P_i m_i^* + N_{i,jl}m_i^*\eta_{jl} + \frac{1}{2}D_{ik}m_i^* m_k^*$$

$$+ \frac{1}{2}V_{ik,jl}m_i^* m_k^* \eta_{jl} + M_i h_i^* + L_{i,jl}h_i^*\eta_{jl} + G_{ik}h_i^* m_k^* + \frac{1}{2}T_{ik,jl}h_i^* m_k^* \eta_{jl}.$$

In this expansion, *every term is rotationally invariant,* because the new variables are invariant. They are defined in the following way. η_{ik} is the finite strain tensor

$$(1.11) \qquad\qquad \eta = \frac{1}{2}(v + \tilde{v} + \tilde{v}v).$$

whereas the starred variables are

$$(1.12) \qquad \begin{aligned} \mathbf{m}^* &= \tilde{R}\mathbf{m} \quad \text{or} \quad m_i^* = m_k R_{ki}, \\ \mathbf{h}^* &= \tilde{R}\mathbf{1} \qquad\quad h_i^* = h_k R_{ki} \end{aligned}$$

with

$$(1.13) \qquad \begin{aligned} R &= (1+v)\cdot(1+2\eta)^{-1/2} = R(v), \\ R\tilde{R} &= \tilde{R}R = 1, \quad \det R = +1. \end{aligned}$$

It can be seen immediately that, with (1.5), these variables are invariant with respect to rotations D of the coordinate system. For small deformations it is

$$(1.14) \qquad\qquad R = 1 + \omega + \frac{1}{2}(\omega\omega - \varepsilon\omega - \omega\varepsilon)\ldots$$

where

$$v = \varepsilon + \tilde{\omega}, \quad \varepsilon = \tilde{\varepsilon}, \quad \omega = -\tilde{\omega}.$$

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[3] If there are variables of the kind indicated in (1.6), translational invariance gives conditions for certain sums, e.g., $\sum\limits_{\alpha} C_i^{(\alpha)} = 0$, if $C_i^{(\alpha)}$ is the coefficient of $u_i^{(\alpha)}$.

Of course, both expansions (1.7) and (1.10) can be used, but (1.7) only with (1.8). We have indicated both expansions because they are necessary in the continuum limit of lattice theory. Although the finite strain theory is old, it seems to me that (1.12) was first given by TOUPIN [11, 12]. The physical meaning is that there are rotational motions if there is a magnetization in a finite strained material. Written explicitly, one of these terms is in lowest order

$$(1.15) \qquad \frac{1}{2} D_{ik}(m_i m_k + m_i m_j \omega_{jk} + m_j m_k \omega_{ji}).$$

The effects related to these rotation terms are not large in general but they can be observed in some cases. It can be seen from (1.15) that this term vanishes in isotropic (cubic) materials, but higher order terms are present even then. Their influence has been discussed by MELCHER [15–18] and others only in recent years.

2. Lattices of mass-points

In a microscopic discrete lattice theory the appropriate variables are the atomic displacements from equilibrium positions:

$$(2.1) \qquad \mathbf{x}^m = \mathbf{X}^m + \mathbf{u}^m$$

(m: number of atom, $\mathbf{X}^m$ corresponds to the material coordinate). Rotational invariance of the potential energy means

$$(2.2) \qquad \Phi(\mathbf{x}) = \Phi(D\mathbf{x}).$$

Expansion about the equilibrium positions gives

$$(2.3) \qquad \Phi(\mathbf{x}) = \Phi_0(\mathbf{X}) + \sum_m \Phi_i^m u_i^m + \frac{1}{2} \sum_{mn} \Phi_{ij}^{mn} u_i^m u_j^n + \dots$$

and after rotation of the coordinate system

$$(2.4) \qquad \begin{aligned} \mathbf{x}' &= D\mathbf{x} = D\mathbf{X} + D\mathbf{u} \quad \text{or with} \quad D = 1 + \Omega, \\ x_i' &= X_i + \Omega_{ik} X_k + u_i + \Omega_{ik} u_k. \end{aligned}$$

If we insert this into (2.3) and regard that Φ has to be as independent of Ω, we obtain the conditions of rotational invariance

$$(2.5) \qquad \begin{aligned} \sum_m \Phi_i^m X_k^m &= \sum_m \Phi_k^m X_i^m, \\ \sum_n \Phi_{ij}^{mn} X_k^n + \Phi_j^m \delta_{ik} &= \sum_n \Phi_{ik}^{mn} X_j^n + \Phi_k^m \delta_{ij} \end{aligned}$$

and so on. These relations are very similar to Eq. $(1.8)_{1,2}$, (1.9) but their correspondence is not complete. As the potential energy is also invariant against (infinitesimal) translations, we obtain further

$$(2.6) \qquad \sum_m \Phi_i^m = 0, \qquad \sum_n \Phi_{ij}^{mn} = 0, \text{ etc.}$$

With the help of (2.6), (2.5) can be transformed in many ways. With the definitions

(2.7)
$$\hat{S}_{ik} = \frac{1}{V} \sum_m \Phi_i^m X_k^m$$

and

$$\hat{S}_{il,jk} = \frac{1}{V} \sum_{mn} \Phi_{ij}^{mn} X_l^m X_k^n$$

(V: volume) it follows

(2.8)
$$\hat{S}_{ik} = \hat{S}_{ki},$$
$$\hat{S}_{il,jk} + \hat{S}_{jl}\delta_{ik} = \hat{S}_{il,kj} + \hat{S}_{kl}\delta_{ij},$$

and a number of similar relations. One of these further relations is

(2.9)
$$\underbrace{\frac{1}{2}(\hat{S}_{il,jk} + \hat{S}_{ik,jl}) - \hat{S}_{kl}\delta_{ij}}_{\hat{C}_{ij,kl}} = \underbrace{\frac{1}{2}(\hat{S}_{kj,li} + \hat{S}_{lj,ki}) - \hat{S}_{ij}\delta_{kl}}_{\hat{C}_{kl,ij}},$$

which is very similar to (1.9); definition (2.7) together with (2.9) gives also

(2.10) $$\hat{C}_{ij,kl} = \frac{1}{2V} \sum_{mn} \Phi_{ij}^{mn}(X_l^m X_k^n + X_k^m X_l^n) = -\frac{1}{2V} \sum_{mn} \Phi_{ij}^{mn}(X_k^m - X_k^n)(X_l^m - X_l^n),$$

where use has been made of (2.6). From (2.10) it can immediately be seen that

(2.11) $$\hat{C}_{ij,kl} = \hat{C}_{ij,lk} = \hat{C}_{ji,kl}.$$

At this point it should be emphasized that all the relations follow from (2.5) and (2.6) and are valid for all sets of mass-points interacting by a potential energy; the set needs not necessarily be a lattice but it must have equilibrium positions.

Now, we can establish the connection between the microscopic description and the elastic theory (1) by using homogeneous deformations or (2) by using slowly varying displacements (as in a wave-field). In the first case we set

(2.12) $$u_i^m = v_{ik} X_k^m,$$

in the second case

(2.13) $$u_i^n - u_i^m = u_{i|k}(X_k^n - X_k^m) + \frac{1}{2} u_{i|kl}(X_k^n - X_k^m)(X_l^n - X^m).$$

In both cases this implies a number of assumptions: the deformations of *all* the atoms are described by (2.12, 13), which means that there is no individual sublattice displacement or every atom is a center of inversion (e.g., Bravais-lattices); there are no defects or surface-effects in the lattice or the lattice is infinite; the range of forces is small compared to the varying displacements or the elastic limit is just that described by (2.12, 13). Since all the essentials can be seen with these assumptions, we restrict our considerations

to such cases. Otherwise we would have additional terms which are not affected by rotational invariance[4]. From (2.3) and (2.12) we have

$$(2.14) \qquad w = \frac{1}{V}(\Phi - \Phi_0) = \hat{S}_{ik}v_{ik} + \frac{1}{2}\hat{S}_{ik,jl}v_{ik}v_{il} + \dots$$

with $\hat{S}$ defined in (2.7). (2.14) is the energy-density of the crystal and can be identified with the pure elastic expression (1.7), that means $S = \hat{S}$ (in Bravais-crystals, otherwise there are further terms). (1.8), (2.8) and similar relations guarantee the maintenance of rotational invariance.

For practical purposes, definitions (2.7) are unfavourable because they contain absolute distances from a given point. For example, surface effects can be eliminated only through complicated manipulations. It is then better to start with the equation of motion derived from (2.3)

$$(2.15) \qquad M\ddot{u}_i^m = -\Phi_i^m - \sum_n \Phi_{ij}^{mn}u_j^n + \dots$$

From this relation it can be seen that the Φ_i^m have to vanish at least in the interior of the crystal to allow for an elastic limit (existence of equibrium). If there are individual forces on every atom, an elastic limit is not possible. We assume, therefore,

$$(2.16) \qquad \Phi_i^m = 0$$

in the interior of the crystal. Then, using (2.13) and the symmetry relations for lattices in which every atom is a center of inversion, we obtain ($u_i^m \Rightarrow u_i$)

$$(2.17) \qquad \varrho\ddot{u}_i = \hat{C}_{ij,kl}u_{j|kl}, \quad \varrho = M/V_z$$

and

$$\hat{C}_{ij,kl} = -\frac{1}{2V_z}\sum_n \Phi_{ij}^{mn}(X_k^m - X_k^n)(X_l^m - X_l^n).$$

This definition of $\hat{C}$ is identical with that in (2.10); if the range of forces is small compared with the dimensions of the crystal and if the crystal is large enough to neglect surface contributions to the sums in (2.10, 17), with order words, the sum over n in (2.17) has to be independent of m (there are N bulk terms, but $N^{2/3}$ surface terms).

(2.17) has to be compared with the corresponding elastic equation of motion derived from (1.7) or (1.10):

$$(2.18) \qquad \varrho\ddot{u}_i = \frac{1}{2}(S_{ik,jl} + S_{il,jk})u_{j|kl}.$$

and by comparison with (2.17)

$$(2.19) \qquad \hat{C}_{ij,kl} = \frac{1}{2}(S_{ik,jl} + S_{ii,jk})$$

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[4] If there are no external fields, etc.

(cf. (2.9); (2.19) holds for Bravais-lattices, otherwise there can be additional terms)[5] (2.19) is consistent, and that is the essential point, only if the condition of rotational invariance is satisfied because this condition guarantees the correct symmetries in the indices. Further (2.19) can be uniquely solved for the $S_{ik,jl}$ or the elastic constants *if, and only if*, the conditions of rotational and translational invariance are satisfied. From (2.19) and (1.9) we then have

$$\hat{C}_{ij,kl} = \frac{1}{2}\,(C_{ik,jl}+C_{il,jk})+S_{kl}\,\delta_{ij},$$

$$\hat{C}_{kj,il} = \frac{1}{2}\,(C_{ki,jl}+C_{kl,ji})+S_{il}\,\delta_{kj},$$

(2.20)

$$-\hat{C}_{ki,jl} = -\frac{1}{2}\,(C_{kj,il}+C_{kl,jj})-S_{jl}\,\delta_{ki},$$

$$\hat{C}_{ik,jl} = \hat{C}_{ij,kl}+\hat{C}_{kj,il}-\hat{C}_{ki,jl}-S_{kl}\,\delta_{ij}-S_{il}\,\delta_{kj}+S_{jl}\,\delta_{ki}.$$

This allows to determine elastic constants from lattice theoretical quantities. It must be emphasized that (2.20) are the elastic constants of a homogeneous medium without any surface effects. If there are stresses in the initial state, these are contained in (2.20), i.e., in the relation between microscopic and macroscopic quantities. By using the symmetry relations these stresses can also be obtained from (2.20) apart from a homogeneous pressure

(2.21) $$p = -\frac{1}{3}\,(S_{11}+S_{22}+S_{33}),$$

e.g.,

$$S_{12} = \hat{C}_{ii,12}-\hat{C}_{21,ii}, \quad i \text{ arbitrary, no summation!}$$

In general, however, the elastic constants are defined without stresses in the initial equilibrium state, so that $S_{kl} = 0$.

Equations (2.17,18) clearly show that by sound wave measurements one determines the $\hat{C}$-coefficients, not the true elastic constants C. But the condition of rotational invariance guarantees that both can be determined from each other.

Some further consequences of the conditions (2.5) should be mentioned. The condition of rotational invariance connects force constants of the n-th and $(n+1)$-th order. In an anharmonic theory, elastic as well as lattice theory, one obtains similar but more complicated relations for the higher order constants. One can even show [6, 9] that a pure harmonic theory is not possible, this means that such a lattice or medium would be instable. In elastic limit as well as in lattice theory it is necessary that every crystal be anharmonic in order to be stable.

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[5] In non-centrosymmetric crystals there is an additional term in (2.18,19,20) which has the form

$$\sum_{\mu\nu} \hat{C}^{\mu}_{m,ik}\, R^{\mu\nu}_{mn}\, \hat{C}^{\nu}_{n,jl}.$$

It can be shown that this term is rotationally invariant and has the correct symmetries [8, 9]. Therefore we will not go into details here.

This does not contradict a lattice model with spring constants between the atoms because every spring acts anharmonically if stretched perpendicular to its direction. But even though this is only a small anharmonic effect, it is sufficient to satisfy the conditions of rotational invariance. Of course, the "real anharmonic" forces in general are larger by an order of magnitude.

The condition of rotational invariance is even more restrictive in the vicinity of defects or any perturbations. We consider a free surface where, again, the significance of this condition can be seen. In finite elastic media there exists the possibility of surface waves (Rayleigh waves, etc.) These waves propagate in a direction lying on the surface and have exponentially decreasing amplitudes perpendicular to the surface. To treat these waves we have to solve (2.18) with the boundary condition of a free surface, i.e., all forces (or stresses) on this surface have to vanish, for a $x_3 = 0$ surface e.g.,

$$(2.22) \qquad S_{13} = S_{23} = S_{33} = 0 \quad \text{at } x_3 = 0.$$

But instead of solving (2.18) with (2.22) we can also discuss the solution of

$$(2.23) \qquad \varrho \ddot{u}_i = S_{ik,jl|k} u_{j|l} + S_{ik,jl} u_{j|kl}$$

and this is even the more general equation of motion, valid for non-homogeneous elastic media (varying elastic constants). It can be derived just as the usual equation of motion (2.18) with the assumption that the $S_{ik,jl}$ depend on position. If the elastic constants can be described by a step-function at $x_3 = 0$, jumping from zero to the value of the interior of the crystal, it can be shown that (2.23) is equivalent to (2.18) and (2.23). In (2.23) it holds with $S_{ik} = 0$

$$(2.24) \qquad S_{ik,jl|k} = S_{ik,lj|k}.$$

Now let us look for the corresponding lattice expression. The equation of motion is (2.15) with $\Phi_i^m = 0$. By introducing the slowly varying displacement field (2.13) we now obtain

$$(2.25) \qquad \varrho \ddot{u}_i = -\frac{1}{V_z} \sum_n \Phi_{ij}^{mn}(X_l^n - X_l^m) u_{j|l} + \hat{C}_{ij,kl} u_{j|kl}$$

which is different from (2.17) in the first term.

In infinite homogeneous lattices or in the interior of finite lattices the first term on the right side vanishes because every atom is then a center of inversion (Bravais-lattices). The second term is the general term of homogeneous media.

In inhomogeneous media, e.g. near a surface, the first term in (2.25) does not vanish since there is no inversion symmetry for surface atoms! It corresponds to the first term in (2.23) with symmetry given by (2.24). Therefore, the first term in (2.25) must have the same symmetry which is possible only if (2.5) is satisfied!

In inhomogeneous media the second term depends on the position (x, m); but this again gives a correspondence between the lattice and elastic theory and can be discussed in the usual way. Moreover, the first term depends on m, but this dependence is limited to the surface if the inhomogeneities are limited to the surface. A step-function in the elastic constants corresponds to the case where the first term in (2.25) is different from zero only for m-values on the surface.

What should be emphasized again is that surface waves calculated by means of the lattice theory coincide with elastic waves or even have an elastic limit only if the condition of rotational invariance is satisfied. This has been overlooked in a great number of papers.

3. Lattices of rigid molecules

The objections first raised against the symmetries of elastic constants and to the agreement of the elastic and lattice theory of all crystals were then transferred to molecular crystals, where the molecules can be looked upon as being rigid. In these molecular crystals we have translations (of the center of mass) and rotations (librations) as the possible motions, which means that we have additional degrees of freedom compared with the usual lattices of "mass-points".

Of course, molecular crystals can also be looked upon as being composed of (very many) mass-points. Then, such lattices can be treated as crystals with many optical branches (of high frequencies) and we can make use of the procedures in the preceding sections to obtain the elastic constants. These constants have then all the properties mentioned there.

But we will show here that we can obtain the same result if we treat the molecules as rigid bodies with additional degrees of freedom connected with librations. Complications arise if in the equilibrium state different molecules of the lattice have different orientations. But as this gives only additional terms in the elastic constants we restrict ourselves to "simple" molecular crystals where all the molecules have the same orientation (in equilibrium) and there is only one molecule per unit cell.

Then the equation of rigid molecule motion is

(3.1)
$$M\ddot{u}_i^m = -\sum_n \Phi_{\substack{u\,u\\i\,j}}^{mn} u_j^n - \sum_n \Phi_{\substack{u\,\omega\\i\,j}}^{mn} \omega_j^n,$$

$$I_{lk}\ddot{\omega}_k^m = -\sum_n \Phi_{\substack{\omega\,u\\i\,j}}^{mn} u_j^n - \sum_n \Phi_{\substack{\omega\,\omega\\i\,j}}^{mn} \omega_j^n.$$

Here u_j^n are the translations (of the mass center), ω_j^n are small librations, I_{ik} is the tensor of the moment of inertia and the $\Phi_{\substack{\alpha\,\beta\\i\,j}}^{mn}$ couple the different motions.

Again, we can look for the conditions of translational and rotational invariance and find out

(3.2)
$$\sum_n \Phi_{\substack{\alpha\,u\\i\,j}}^{mn} = \sum_m \Phi_{\substack{u\,\beta\\i\,j}}^{mn} = 0,$$

(3.3)$_1$
$$\sum_n \left\{ \Phi_{\substack{\alpha\,u\\i\,j}}^{mn} X_k^n - \Phi_{\substack{\alpha\,u\\i\,k}}^{mn} X_j^n \right\} = \sum_n \Phi_{\substack{\alpha\,\omega\\i\,l}}^{m\mu} \varepsilon_{lkj},$$

or

(3.3)$_2$
$$\sum_n \Phi_{\substack{\alpha\,u\\i\,j}}^{mn} X_k^n \varepsilon_{lkj} = \sum_n \Phi_{\substack{\alpha\,\omega\\i\,l}}^{mn}$$

with $\alpha, \beta = u$ or ω and ε_{lkj}: total antisymmetry tensor of 3rd rank. It should be mentioned that these relations imply equilibrium in the initial state; otherwise we have additional terms. The condition of rotational invariance (3.3) now connects translational and librational motions and this even twice (with $\alpha = u$ and $\alpha = \omega$). In other words, the coupling constants of translational and librational motions (different kinds of motions, variables) are connected by these relations. This is the essential point. In order to obtain the elastic limit, we introduce slowly varying fields again:

(3.4)
$$u_j^n - u_j^m = u_{j|k}(X_k^n - X_k^m) + \frac{1}{2} u_{j|kl}(X_k^n - X_k^m)(X_l^n - X_l^m),$$

$$\omega_j^n - \omega_j^m = \omega_{j|k}(X_k^n - X_k^m).$$

Then

(3.5)
$$\varrho \ddot{u}_i = \hat{C}_{ij,kl} u_{j|kl} - \hat{C}_{m,ik} \omega_{m|k},$$

$$\frac{1}{V_z} I_{ik} \ddot{\omega}_k = \hat{C}_{i,jl} u_{j|l} + \hat{C}_{im} \omega_m$$

with

(3.6)
$$\hat{C}_{ij,kl} = -\frac{1}{2V_z} \sum_n \Phi_{u\; u}^{m\; n}_{\;i\; j} (X_k^m - X_k^n)(X_l^m - X_l^n),$$

$$\hat{C}_{i,jk} = -\frac{1}{V_z} \sum_n \Phi_{\omega\; u}^{m\; n}_{\;i\; j} (X_k^n - X_k^m),$$

$$\hat{C}_{ij} = -\frac{1}{V_z} \sum_n \Phi_{\omega\; \omega}^{m\; n}_{\;i\; j};$$

in deriving (3.5) use has been made of (3.2) and of the fact that there is only one molecule per unit cell. Further, using (3.2) and

$$\hat{S}_{ik,jl} = -\sum_{mn} \Phi_{u\; u}^{m\; n}_{\;i\; j} X_k^m X_l^n$$

we obtain

(3.7)
$$\hat{C}_{ij,kl} = \frac{1}{2}(\hat{S}_{ik,jl} + \hat{S}_{il,jk}).$$

The condition of rotational invariance gives the symmetry relations

(3.8)
$$\hat{C}_{ij} = \hat{C}_{ji}; \quad \hat{C}_{i,jk} - \hat{C}_{i,kj} = \hat{C}_{il}\varepsilon_{lkj},$$

$$\hat{S}_{il,jk} - \hat{S}_{il,kj} = -\hat{C}_{s,il}\varepsilon_{skj},$$

$$\hat{S}_{il,jk} - \hat{S}_{li,jk} = -\hat{C}_{s,jk}\varepsilon_{sli}$$

and by direct inspection

(3.9)
$$\hat{C}_{ij,kl} = \hat{C}_{ji,kl} = \hat{C}_{ij,lk}, \quad \hat{S}_{il,jk} = \hat{S}_{jk,il}.$$

The relations (3.8,9) now guarantee that the solution of (3.5) has an elastic limit and gives the elastic constants with the correct symmetries.

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ω_m has to be eliminated from (3.5) where the term $\ddot{\omega}_k$ can be neglected in accordance to the following argument: in the long wave limit $u_{j|l}$ and $\omega_{m|k}$ vanish, therefore, $\ddot{u}_i$ as well which describes the acoustical waves in this limit. $\ddot{\omega}_k \sim \hat{C}_{im}\omega_m$ must describe the librational modes with non-vanishing frequencies in this limit. These frequencies are "high" compared with the elastical acoustical ones which are of sole interest here. We can neglect librational waves and use stationary solutions of $(3.5)_2$:

$$\omega_m = -(\hat{C}^{-1})_{mn}\hat{C}_{n,jl}u_{j|l},$$

(3.10)

$$\varrho\ddot{u}_i = \{\hat{C}_{ij,kl}+\hat{C}_{m,ik}(\hat{C}^{-1})_{mn}\hat{C}_{n,jl}\}u_{j|kl} = \frac{1}{2}\{S_{ik,jl}+S_{il,jk}\}u_{j|kl}$$

according to (2.18). This relation can be solved for the elastic constants $S_{ik,jl}$ or $C_{ik,jl}$ provided that (3.8) and (3.9) and (3.2,3) are satisfied. Then, and only then, all the coefficients again have the correct symmetries. For the sake of simplicity, we introduce

(3.11) $$A_{ij,kl} = \hat{C}_{ij,kl}+\frac{1}{2}\hat{C}_{m,ik}(\hat{C}^{-1})_{mn}\hat{C}_{n,jl}+\frac{1}{2}\hat{C}_{m,il}(\hat{C}^{-1})_{mn}\hat{C}_{n,jk}$$

and obtain, similarly as in before

(3.12) $$C_{ik,jl} = A_{ij,kl}+A_{kj,il}-A_{ki,jl}.$$

In a sound wave measurement one measures only the sum given in (3.10). The elastic constants, defined by static deformations, can be obtained from (3.10) only through (3.11,12). However, they have the correct symmetries as well as the coefficients in (3.10).

It must be emphasized further that, if one starts with a given potential energy, then the conditions (2.5,6) and (3.2,3) are always satisfied; but if one does model calculations with free parameters, these conditions are true conditions, which have to be satisfied and which set restrictions to the possible models.

4. Lattices of atoms with internal degrees of freedom

The situation becomes even more complicated if one considers interactions in external fields, e.g., in magnetic and electrical fields, which act on internal degrees of freedom like spins, etc. One is then forced to consider additional terms which describe the corresponding interactions with spins or electrical polarisation and so on. The elastic limit of these effects can be found in the description given at the beginning.

Lattice theory, of course in the long wave limit, has to coincide with this elastic theory, and again this is consistent only if there is a rotational invariant expansion in the lattice theory as well. In order to compare the lattice theory with the elastic limit, we can start as previously. The energy of the crystal might be given by a spin-dependent and a usual potential energy:

(4.1) $$E = \frac{1}{2}\sum_{mn}J_{ij}^{mn}\sigma_i^m\sigma_j^n+h_i\sum_n K_{ij}^n\sigma_j^n+\Phi(x_i^m).$$

Ising- and Heisenberg-models are special forms of this theorem (4.1) can be expanded with respect to displacements from equilibrium positions to give

$$(4.2) \quad E = h_i \left\{ \sum_n K^n_{ij} \sigma^n_j + \sum_{np} K^{np}_{ijk} \sigma^n_j u^p_k \dots \right\}$$

$$+ \frac{1}{2} \sum_{nm} J^{mn}_{ij} \sigma^m_i \sigma^n_j + \frac{1}{2} \sum_{mnp} J^{mnp}_{ijk} \sigma^m_i \sigma^n_j u^p_k + \Phi_0(X^m_i) + \frac{1}{2} \sum_{mn} \Phi^{mn}_{ij} u^m_i u^n_j.$$

This energy has to be invariant against translations and infinitesimal rotations of the coordinate system

$$u^m_i \Rightarrow u^m_i + a_i, \qquad\qquad \sigma^m_i \Rightarrow \sigma^m_i,$$

$$u^m_i \Rightarrow u^m_i + \Omega_{ik} X^m_k + \Omega_{ik} u^m_k, \qquad \sigma^m_i \Rightarrow \sigma^m_i + \Omega_{ik} \sigma^m_k.$$

Then the conditions of translational invariance are

$$(4.3) \qquad \sum_p \Phi^{mn}_{ij} = \sum_m \Phi^{mn}_{ij} = 0,$$

$$\sum_p K^{mp}_{ijk} = 0, \qquad \sum_p J^{mnp}_{ijk} = 0,$$

and those of rotational invariance

$$(4.4) \qquad \left. \begin{aligned} \sum_n \Phi^{mn}_{jk} X^n_l &= \\ \sum_p J^{mnp}_{ijk} X^p_l + J^{mn}_{kj}\delta_{il} + J^{mn}_{ik}\delta_{jl} &= \\ \sum_p K^{np}_{ijk} X^p_l + K^n_{kj}\delta_{il} + K^n_{ik}\delta_{jl} &= \end{aligned} \right\} \quad \begin{aligned} &\text{symmetric against,} \\[1em] &\text{interchange of } k, l. \end{aligned}$$

These relations have a certain similarity to the relations (1.8); they are necessary and sufficient for a consistent elastic limit of (4.2).

In order to obtain the correspondence to the elastic limit explicitly one has to use again the method of homogeneous deformation (2.12) or of slowly varying fields (long waves, 2.13) together with the energy density from (4.2) or the corresponding equations of motion:

$$M^m \ddot{u}^m_i = \dot{p}^m_i = -h_l \sum_n K^{nm}_{lj} \sigma^n_j - \frac{1}{2} \sum_{np} J^{pnm}_{lji} \sigma^p_l \sigma^n_j - \sum_n \Phi^{mn}_{ij} u^n_j,$$

$$(4.5)$$

$$\dot{\sigma}^m_i = +\varepsilon_{ijk} h_l \left\{ K^m_{lj} \sigma^m_k + \sum_p K^{mp}_{ljs} \sigma^m_k u^p_s \right\} + \varepsilon_{ijk} \left\{ \sum_n J^{mn}_{jl} \sigma^m_k \sigma^n_l + \sum_{np} J^{mnp}_{jls} \sigma^m_k \sigma^n_l u^p_s \right\}.$$

One first obtains certain lattice theoretical coupling constants which have symmetry properties expressed by (4.3) and (4.4) basically. With the help of these properties, and

only if (4.3,4) are satisfied, these lattice theoretical constants can be related to the expansion coefficients of the elastic theory in (1.7) or (1.10). We will not give the details here because some of the expressions are very "long". But in any case the elastic limit exists consistently only if (4.3,4) are satisfied; in other words, the rotational terms in (1.7) and (1.10) are necessary for a consistent theory.

The effects of these terms and corresponding higher ones are small in general and occur in lowest approximation only in anisotropic materials. However, some effects caused by these terms have been observed since 1970. The first investigations seem to have been made by MELCHER [18] who studied the antiferromagnetic system MnF_2. A large effect of the rotation terms can be seen near the spin-flop-transition which gives effective elastic constants.

It is clear that the rotational invariance is of similar importance in every system which is elastically coupled to further degrees of freedom and to external fields. In the case of electrical fields and electrical (sublattice) polarisation in crystals this has already been pointed out by TOUPIN [1956], and in a more general form, by LAX and NELSON [14]. The main point is always the same: one has to use the appropriate rotational invariant variables in the continuum theory and, to obey the conditions of translational and rotational conditions in the microscopic lattice theory. The theory can also be extended to energy densities depending on higher derivatives of displacements, of magnetic moments, of fields. In any case one has to look for the corresponding invariant variable and the related microscopic invariance conditions.

Of course, there might be and there are in general further conditions which set restrictions to the expansion parameters. These further conditions are related to the lattice symmetries and set the usual restrictions to the number of independent constants. Other invariance conditions are those related to space inversion (parity) and time reversal. Sometimes these are also essential.

But it seems to me that the most important conditions are those of translational and even more of rotational invariance. They must be satisfied in every system with no exception and only they can guarantee a continuum theory as a consistent limit to the microscopic theory.

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