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# PARTICLE ASPECTS OF PHONONS

Det Kongelige Danske Videnskabernes Selskab  
Matematisk-fysiske Meddelelser 37, 5



Kommissionær: Munksgaard

København 1969

### **Synopsis**

The possibility of introducing dynamical variables for a single phonon is investigated. A space time description of localized phonons leads to position and direction variables. Lattice translations, point group transformations and local rotations are used to define variables analogous to momentum, angular momentum, parity and spin of an ordinary particle.

## 1. Introduction

A system with  $3N$  degrees of freedom which can perform harmonic vibrations may be treated quantum mechanically in two formally different ways. In the first case it is described as  $3N$  distinguishable oscillators, preferably the normal vibrations of the system. In the second case the excitations of the system are treated as a gas of indistinguishable Bose-particles, phonons.

The purpose of the present note is to investigate how far it is possible to carry the latter description. In particular we shall try to introduce dynamical variables of a single phonon analogous to the variables of ordinary particles like spatial coordinate, momentum, orbital angular momentum and spin angular momentum.

An analysis of the so called pseudomomentum of a phonon has been carried through by SÜSSMANN<sup>(1) (2)</sup>. The concept of phonon spin is discussed by VONSOVSKII and SVIRSKII<sup>(3)</sup> and by LEVINE<sup>(4)</sup> in the continuum limit for an isotropic and a cubic material respectively. In the present note which extends previous work<sup>(5)</sup>, these and other concepts are treated, starting from an atomistic description of the vibrating system.

## 2. Space-time Description of Phonons

Consider a crystal with  $N$  atoms. Phonons are introduced on the basis of the harmonic approximation in which the crystal is described by the Hamiltonian

$$H = \frac{1}{2}(p_r T_{rs} p_s + u_r V_{rs} u_s) \quad (1)$$

(summation over indices occurring twice being understood).

Here the summation indices  $r$  and  $s$  run over all  $3N$  degrees of freedom, i. e. they label both the equilibrium positions of the atoms and the cartesian

components of the momenta  $p$  and displacements  $u$ . The matrix  $T$  is diagonal ( $T_{rs} = (1/m_r)\delta_{rs}$ , where  $m_r$  is the mass related to the  $r$ 'th degree of freedom) but the formalism which we are going to describe is valid also for the more general case of non-diagonal  $T$ . The potential energy is assumed to be positive definite\*;  $V$  is a symmetric matrix\*\*.

For convenience we introduce mass-adjusted canonically conjugate variables

$$\pi_r = (T^{1/2})_{rs} p_s \quad v_r = (T^{-1/2})_{rs} u_s \quad (2)$$

where  $T^{1/2}$  is the real, symmetric, positive definite square root of the matrix  $T$ . In our case  $(T^{1/2})_{rs} = (1/\sqrt{m_r})\delta_{rs}$ .

In terms of the new variables the Hamiltonian becomes

$$H = \frac{1}{2}(\pi_r \pi_r + v_r D_{rs} v_s) \quad (3)$$

where

$$D = T^{1/2} V T^{1/2} \quad (4)$$

is the so called dynamical matrix of the system. It is a symmetric, positive definite and real  $3N \times 3N$  matrix.

From the assumed properties of the matrix  $D$  it follows that there exists one and only one symmetrical, positive definite and real matrix  $M$  which fulfils the relation

$$M^4 = D. \quad (5)$$

This matrix has a reciprocal  $M^{-1}$  because it is positive definite.

The actual calculation of functions of hermitian matrices, e. g.  $D^{1/4}$ , is most conveniently done in the following way: The dynamical matrix  $D$  is diagonalized. The diagonal elements are replaced by their function values, e. g. by their positive fourth roots, and finally the matrix is transformed to the original representation. In ref.<sup>(5)</sup> this is described in more detail using the infinite linear chain with nearest neighbour interaction as an example\*\*\*.

A mathematical treatment of functions of matrices is given in several textbooks, e. g. A. I. MAL'CEV<sup>(6)</sup>.

By means of  $M$  we introduce  $3N$  creation and  $3N$  destruction operators  $b_r^\dagger$  and  $b_r$  through the definitions

\* If needed, this may be enforced by adding, e. g., a small fictitious term proportional to  $u_r u_r$  and letting the proportionality constant go to zero in the final results.

\*\* The most general case, with  $H$  containing terms of the type  $p_r G_{rs} u_s$  as in the presence of magnetic forces or Coriolis forces, is considered in Appendix I.

\*\*\* We take the opportunity to correct an error in ref.<sup>(5)</sup>; the right side of eq. 4.11 should be multiplied by 4.

$$v_s = \sqrt{\frac{\hbar}{2}} (M^{-1})_{sr} (b_r + b_r^\dagger) \quad (6)$$

$$\pi_s = \frac{1}{i} \sqrt{\frac{\hbar}{2}} M_{sr} (b_r - b_r^\dagger) \quad (7)$$

with the inverse relations

$$b_r = \frac{1}{\sqrt{2\hbar}} (M_{rs} v_s + i(M^{-1})_{rs} \pi_s) \quad (8)$$

$$b_r^\dagger = \frac{1}{\sqrt{2\hbar}} (M_{rs} v_s - i(M^{-1})_{rs} \pi_s). \quad (9)$$

From (8) and (9) together with the canonical commutation relations for  $\pi_r$  and  $v_r$  it follows that the  $b$  and  $b^\dagger$  operators obey the commutation relations characterizing destruction and creation operators for Bose-particles

$$\left. \begin{aligned} [b_r, b_s^\dagger] &= b_r b_s^\dagger - b_s^\dagger b_r = \delta_{rs} \\ [b_r, b_s] &= [b_r^\dagger, b_s^\dagger] = 0. \end{aligned} \right\} \quad (10)$$

When we introduce (6) and (7) in (3) we obtain the Hamiltonian in terms of the creation and destruction operators

$$H = \frac{\hbar}{2} (M^2)_{rr} + \hbar b_r^\dagger (M^2)_{rs} b_s. \quad (11)$$

Thus  $H$  is written as the sum of a zero point energy

$$E_0 = \frac{\hbar}{2} \text{trace} (M^2) \quad (12)$$

and an excitation Hamiltonian, the form of which is characteristic for a system of non-interacting bosons.

The boson operators  $b_r$  and  $b_r^\dagger$  defined by (6) and (7) are said to destroy and create a localized phonon at the atom and at the coordinate axis denoted by  $r$ . If the system given by the Hamiltonian (3) is large, then the excitation energy of the localized phonons is to a large extent localized in the neighbourhood of the atom corresponding to the index  $r$  (appendix II).

The form of eq. (11) makes it natural to consider the matrix  $\hbar M^2 = \hbar \sqrt{D}$  as the Hamiltonian matrix for a single phonon. Henceforth it will be denoted by  $h$

$$h_{rs} = \hbar(M^2)_{rs} = \hbar(\sqrt{D})_{rs}. \quad (13)$$

Having defined  $b$  and  $b^\dagger$  operators and the one-particle Hamiltonian we can proceed in principle as in the case of an ordinary boson gas. The ground state of the system (including the time dependent phase factor  $\exp(- (i/\hbar)E_0t)$ ) is denoted  $|0\rangle$  and obeys the equations  $b_r|0\rangle = 0$  for all  $r$ . From this state a complete set of states is obtained by successive application of the creation operators. In particular the state  $b_s^\dagger|0\rangle$  will be said to contain one phonon with the position and direction given by  $s$ . It is an eigenfunction with the eigenvalue  $s$  of the dynamical variable

$$\hat{r} = \sum_r r b_r^\dagger b_r \quad (14)$$

which may be called the position-direction operator of the phonon.

The most general one-phonon state is

$$|\psi\rangle = \sum_r \psi_r(t) b_r^\dagger |0\rangle \quad (15)$$

where  $\psi_r(t)$  will be called the Schrödinger function of the phonon in the  $r$ -representation.  $|\psi_r(t)|^2$  is the probability that when exactly one phonon is present it has the position and direction  $r$ . From (14), (11) and (13) together with the general Schrödinger equation

$$i\hbar \frac{\partial}{\partial t} |\psi\rangle = H |\psi\rangle \quad (16)$$

we find that  $\psi_r(t)$  obeys the one-phonon Schrödinger equation

$$i\hbar \frac{d\psi_r}{dt} = h_{rs} \psi_s. \quad (17)$$

The analogy with the non relativistic quantum theory for ordinary particles is nearly complete. The main difference is that the position variable of the phonon in this formalism can take on only discrete values, i. e. the equilibrium positions of the vibrating particles. Therefore (17) is a difference equation in  $r$  instead of a differential equation.

Similarly we can discuss states with more than one phonon.

The procedure outlined above is to some extent arbitrary. Starting from canonically conjugate coordinates and momenta many linear transformations lead to Boson operators. In fact it can be shown that the most general transformation of the form

$$\left. \begin{aligned} u_r &= \sqrt{\frac{\hbar}{2}}(A_{rs}b_s + A_{rs}^*b_s^\dagger) \\ p_r &= \frac{1}{i}\sqrt{\frac{\hbar}{2}}(B_{rs}b_s - B_{rs}^*b_s^\dagger) \end{aligned} \right\} \quad (18)$$

which leads to operators obeying the commutation relations (10) is such that (in matrix notation)

$$\left. \begin{aligned} A &= S^{-1}W \\ B &= S(1 + iC)W \end{aligned} \right\} \quad (19)$$

where  $S$  and  $C$  are real and symmetric matrices, while  $W$  is a unitary matrix. (The matrix  $A$  must have a reciprocal matrix in order to ensure that (18) can be reversed).

However, in order to have a reasonable particle concept, the total number of particles,  $\hat{n} = b_r^\dagger b_r$ , must be a constant of motion under movements described by the unperturbed Hamiltonian (1).

If we impose this condition on the transformation and use the Hamiltonian (1) with positive definite  $T$  and  $V$  we find that the matrix  $C$  in (19) must vanish, while the matrix  $S$  must obey the relation

$$S^2 T S^2 = V \quad (20)$$

by which the positive definite, real, symmetric matrix  $S^2$  is uniquely determined. It now readily follows from (18) that all acceptable phonon variables are connected by transformations of the type  $b'_r = U_{rs}b_s$ , where  $U_{rs}$  is a unitary matrix.

Among the acceptable sets of phonon variables we have chosen a particular one (defined by (8) and (9)) as describing spatially localized phonons. In certain simple cases, notably when all masses are equal and all diagonal elements of the dynamical matrix are equal, the choice is justified by the fact that the excitation energy becomes spatially localized to the highest degree possible. In the general case this is not necessarily so, but the transformation leading to maximum localization of energy cannot then be described in terms of simple functions of the matrices  $T$  and  $V$ . Such a functional relationship is important for the following discussions; in all cases we therefore define localized phonons by means of (8) and (9). With equal right we might have chosen other transformations which coincide with (8) and (9) in case the matrices  $T$  and  $V$  commute. We could, e. g., have reversed the rôles of  $T$  and  $V$  in the procedure leading from the Hamiltonian (1) to the localized phonons given by (8) and (9), or—most symmetrical with respect to kinetic and potential energy—we could have chosen a symmetrical  $A (= S^{-1})$  in (18). The actual choice agrees most closely with standard methods and concepts described in the literature.

In Appendix II the localization of excitation energy is treated in more detail.

### 3. Transformations and Dynamical Variables

The most important dynamical variables of a system are those (like momentum and angular momentum) which are connected with coordinate transformations or symmetry operations. We consider transformations of the particle variables  $u_r$  and  $p_r$  of the form

$$\left. \begin{aligned} u'_r &= L_{rs}u_s \\ p'_r &= L_{rs}p_s \end{aligned} \right\} \quad (21)$$

where the transformation matrix  $L$  is real and orthogonal

$$\tilde{L} = L^{-1}. \quad (22)$$

These transformations include translations and rotations of the displacement pattern of the system. Special cases are treated in the next paragraph.

Introducing (21) in the Hamiltonian (1) we obtain new matrices  $T'$  and  $V'$  defining the kinetic and potential energy:

$$T' = LTL^{-1} \quad V' = LVL^{-1}. \quad (23)$$

They are again real, symmetric and positive definite matrices; consequently transformed matrices  $D'$ ,  $M'$  and  $h'$  together with transformed creation and destruction operators  $b_r^{\dagger'}$  and  $b_r'$  can be defined as before. All matrices like  $D$ ,  $M$  and  $h$  are seen to transform according to the rule (23) whereas vectors like  $v$ ,  $\pi$ ,  $b^\dagger$  and  $b$  transform like  $u$  and  $p$ ; in particular

$$\left. \begin{aligned} b'_r &= L_{rs}b_s \\ b_r^{\dagger'} &= L_{rs}b_s^\dagger \end{aligned} \right\} \quad (24)$$

The ground state is unchanged under this transformation and the phonon number operator is unchanged.

The transformation (24) can be written as a contact transformation; in fact, we can always find a hermitian operator

$$\hat{O} = b_r^\dagger O_{rs} b_s \quad (25)$$

where

$$O_{rs}^* = O_{sr} \quad (26)$$

such that (24) becomes

$$\left. \begin{aligned} b'_r &= \exp(i\hat{O}) b_r \exp(-i\hat{O}) \\ b_r^{\dagger'} &= \exp(i\hat{O}) b_r^\dagger \exp(-i\hat{O}). \end{aligned} \right\} \quad (27)$$



Each transformation like (21) thus motivates the introduction of a dynamical variable  $\hat{O}$ . From (25) we see that  $\hat{O}$  commutes with the phonon number operator and thus  $\hat{O}$  can be said to describe a property of the single phonon. If the Hamiltonian (1) is form invariant under the transformation (21) the Hamiltonian matrix  $h$  commutes with  $L$  and thus  $h' = h$ . From this it follows that the operator  $\exp(i\hat{O})$  commutes with the Hamiltonian operator. In this case it is therefore a constant of motion.

The actual calculation of  $\hat{O}$  can be performed according to the following procedure (for the proof see appendix III).

- a) Find a complete orthonormal set of eigenvectors  $B_r^\alpha$  of the matrix  $L$ , i. e. solve

$$L_{rs}B_s^\alpha = \lambda_\alpha B_r^\alpha. \quad (28)$$

Since  $L$  is unitary, any eigenvalue  $\lambda_\alpha$  has modulus 1.

- b) For each eigenvector determine a real number  $l_\alpha$  so that

$$\lambda_\alpha = \exp(il_\alpha). \quad (29)$$

- c) The matrix  $O_{rs}$  may then be chosen as

$$O_{rs} = \sum_\alpha l_\alpha B_r^{\alpha*} B_s^\alpha (= i(\ln(L))_{rs}). \quad (30)$$

- d) Introducing (30) into (25) we find

$$\hat{O} = \sum_\alpha l_\alpha b_\alpha^\dagger b_\alpha \quad (31)$$

where

$$\left. \begin{aligned} b_\alpha &\stackrel{\text{def}}{=} \sum_r B_r^\alpha b_r \\ b_\alpha^\dagger &\stackrel{\text{def}}{=} \sum_r B_r^{\alpha*} b_r^\dagger \end{aligned} \right\} \quad (32)$$

constitute a new set of phonon destruction and creation operators.

The state  $b_\alpha^\dagger |0\rangle$  is a one phonon eigenstate of the operator  $\hat{O}$  with the eigenvalue  $l_\alpha$ .

It is seen that the scheme described is completely analogous to the usual transformation theory in the quantum mechanics of particles.

The operator  $\hat{O}$  is not uniquely defined by the rules given so far. In fact it is evident from the definition of  $l_\alpha$  that  $\exp(i\hat{O})$  is not changed when we add arbitrary integer multiples of  $2\pi$  to the numbers  $l_\alpha$  in (30). In each single case the definition can be made unique by a suitable convention (e. g. a continuity convention for continuous groups or limitation of wavevectors to the first Brillouin zone in case of the lattice translation group).

It might be considered more straightforward to introduce dynamical variables by writing the transformations of displacements and momenta (21) as contact transformations,

$$\left. \begin{aligned} \exp(i\hat{F})u_r \exp(-i\hat{F}) &= L_{rs}u_s \\ \exp(i\hat{F})p_r \exp(-i\hat{F}) &= L_{rs}p_s. \end{aligned} \right\} \quad (33)$$

In general, however, an operator  $\hat{F}$  satisfying (33) will not be a one-phonon operator, i. e. when expressed in terms of the phonon variables  $b$  and  $b^\dagger$ , it will contain terms of the form  $b_r b_s$  and  $b_r^\dagger b_s^\dagger$ . Thus it does not commute with the phonon number operator, in contrast to  $\hat{O}$ . In particular,  $\hat{F}$  and also  $\exp(i\hat{F})$  may change the ground state into a superposition of the ground state and states with phonons present. For these reasons, in the general case  $\hat{F}$  cannot be said to describe a property of a single phonon.

More specifically we can find under what conditions the operator  $\hat{F}$  can be a one-phonon operator. We try a solution to (33) of the form

$$\hat{F} = b_r^\dagger F_{rs} b_s. \quad (34)$$

Expressing  $u$ 's and  $p$ 's in terms of  $b$ 's and  $b^\dagger$ 's by means of (2), (6) and (7) and using eq. (III, 1) (see Appendix III) we find that a solution of the type (34) exists if and only if the matrix  $L$  commutes with the matrix  $T^{-1/2} M^2 T^{-1/2}$ . If, in particular,  $L$  commutes with both the potential and kinetic energy matrices,  $V$  and  $T$ , the equations determining  $\hat{O}$  and  $\hat{F}$  become identical, so that  $\hat{F}$  can be chosen equal to  $\hat{O}$ .

In the general case explicit solutions to (33) may be written down, but they are usually not of much interest. We only mention that if the matrix  $O_{rs}$  is anti-symmetric, the following expression satisfies (33):

$$\hat{F} = \frac{i}{\hbar} u_r O_{rs} p_s. \quad (35)$$

This may be proved by means of eq. (III,1) (Appendix III).

#### 4. Application to Phonons in Crystals

To each atom in a crystal belong a displacement vector  $\mathbf{u}$  and a momentum vector  $\mathbf{p}$ . Introducing phonon variables we get for each atom a three component creation and a three component destruction operator which transform as vectors under rotation. The equilibrium positions of the atoms will be described as a lattice with a basis. If there are  $N$  lattice points with  $\nu$  atoms in the basis we have altogether  $3N\nu$  creation operators forming  $N\nu$  vectors. An atom will be labelled by the lattice vector  $\mathbf{r}$  and the basis vector  $\mathbf{c}$  of its equilibrium position which is  $\mathbf{r} + \mathbf{c}$ . The phonon operators are denoted  $\mathbf{b}_{\mathbf{r}\mathbf{c}}$  and  $\mathbf{b}_{\mathbf{r}\mathbf{c}}^\dagger$  or in components  $b_{\mathbf{r}\mathbf{c}}^\alpha$  and  $b_{\mathbf{r}\mathbf{c}}^{\alpha\dagger}$ , where  $\alpha$  labels three cartesian coordinate

axes with unit vectors  $\mathbf{e}_\alpha$ . Following eq. (14) we can then define dynamical variables

$$\left. \begin{aligned} \hat{\mathbf{r}} &= \sum_{rc} \mathbf{r}(\mathbf{b}_{rc}^\dagger \cdot \mathbf{b}_{rc}) \\ \hat{\mathbf{c}} &= \sum_{rc} \mathbf{c}(\mathbf{b}_{rc}^\dagger \cdot \mathbf{b}_{rc}) \\ \hat{\mathbf{e}} &= \sum_{rc\alpha} \mathbf{e}_\alpha b_{rc}^{\alpha\dagger} b_{rc}^\alpha \end{aligned} \right\} (36)$$

In the one-phonon case they constitute a complete commuting set of dynamical variables with eigenstates  $b_{rc}^{\alpha\dagger} |0\rangle$  and corresponding eigenvalues  $\mathbf{r}'$ ,  $\mathbf{c}'$ , and  $\mathbf{e}_{\alpha'}$ . They may be called the lattice position operator, the basis position operator and the polarisation direction operator respectively.

The transformations of the type (21) which can be applied to phonons in crystals include permutations of the field vectors among the sites of the crystal and changes of the directions of the field vectors. Of greatest interest are those which can be described as simple spatial operations. This is the case with the following transformations:

- a) The cyclic translational group of the crystal lattice applied to the field vectors. This is applicable to all ideal crystals. It affects  $\hat{\mathbf{r}}$  but not  $\hat{\mathbf{c}}$  and  $\hat{\mathbf{e}}$ .
- b) Proper and improper rotations of the field vectors without permutation among sites. They are applicable to all crystals. They affect  $\hat{\mathbf{e}}$  but not  $\hat{\mathbf{r}}$  and  $\hat{\mathbf{c}}$ .
- c) If the lattice of a crystal is mapped into itself by a certain point group transformation like rotation, reflection or inversion (possibly made cyclic by suitable boundary conventions), this transformation can be applied to the field vectors without changing their direction or basis vector. It affects  $\hat{\mathbf{r}}$  but not  $\hat{\mathbf{c}}$  and  $\hat{\mathbf{e}}$ .
- d) If the set of equilibrium sites in the basis of a crystal is mapped into itself by a certain symmetry operation this can be applied to the field vectors without changing their direction or lattice vector. It affects  $\hat{\mathbf{c}}$  but not  $\hat{\mathbf{r}}$  and  $\hat{\mathbf{e}}$ .

We shall study the cases a), b) and examples among c) and d). We will show that these cases lead to the introduction of dynamical variables analogous to momentum, angular momentum and parity, and to the splitting of the latter two into orbital and intrinsic parts (like orbital and spin angular momentum)\*.

\* Space group transformations, like e. g. screw translations, which are not combinations of the cases a) to d) will not be considered in detail. They lead to operators which are not analogous to simple operators of ordinary particles.

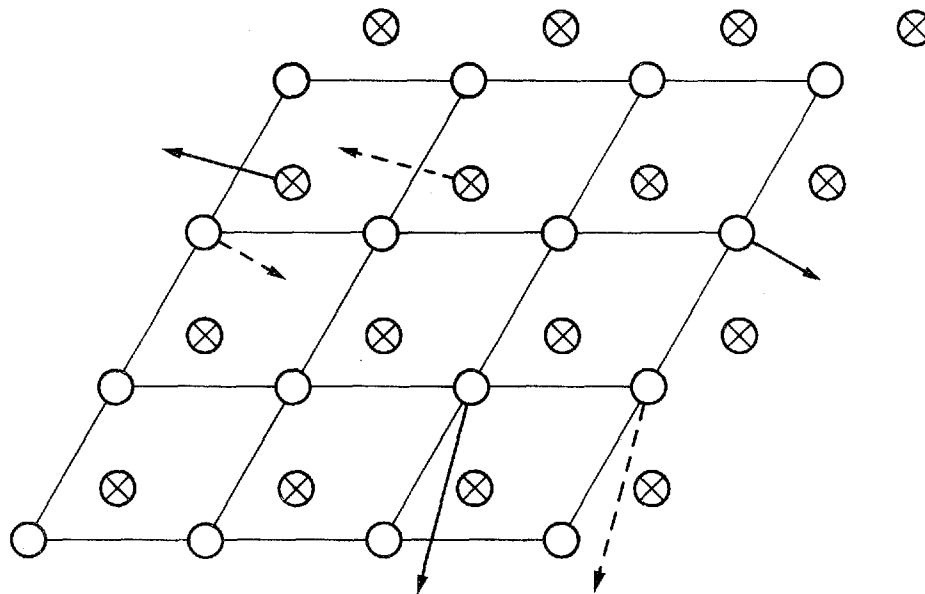


Fig. 1. A cyclic transformation in a finite net. Fully drawn arrows and dashed ones are field vectors before and after the transformation.

We want to emphasize that none of these transformations imply movement of the crystal as a whole. What is, e. g., translated or rotated by the transformations (21) is not the atoms but the patterns of displacements, i. e. the sound field. It is of course also possible to study translation and rotation of the crystal as a whole. This leads to the introduction of the proper momentum and angular momentum operators of the crystal. They are not phonon operators of the type considered here, i. e. they are not of the form (31) (compare <sup>(1)</sup> and <sup>(5)</sup>).

### 5. Translation and Pseudomomentum

A thorough discussion of the concept of pseudomomentum has been given by SÜSSMANN<sup>(1) (2)</sup>. We include a short treatment of this concept in order to see how it fits into the framework presented above.

The transformation group considered is the cyclic shifting of the sound field vectors by integer multiples of the primitive translation vectors of the lattice  $\mathbf{a}_1$ ,  $\mathbf{a}_2$ , and  $\mathbf{a}_3$  (fig. 1). Let the crystal have the form of a parallelepiped with sides  $N_1\mathbf{a}_1$ ,  $N_2\mathbf{a}_2$  and  $N_3\mathbf{a}_3$ , so that it contains  $N = N_1N_2N_3$  unit cells.

The wavevector lattice of the crystal has three primitive translation vectors  $\mathbf{g}_i$  defined by

$$\mathbf{g}_i \cdot \mathbf{a}_j = 2\pi\delta_{ij}. \quad (37)$$

The transformations in question are obtained by repeated application of the three commuting primitive translations

$$\left. \begin{aligned} \mathbf{b}'_{rc} &= \mathbf{b}_{(r-a_i)c} && \text{for } \mathbf{r} - \mathbf{a}_i \text{ inside the crystal} \\ \mathbf{b}'_{rc} &= \mathbf{b}_{(r+(N_i-1)\mathbf{a}_i)c} && \text{for } \mathbf{r} - \mathbf{a}_i \text{ outside the crystal} \\ (i &= 1, 2, 3). \end{aligned} \right\} \quad (38)$$

We now proceed as described in section 3. The three transformation matrices corresponding to (38) have the well known set of simultaneous  $3N\nu$ -component eigenvectors ( $\alpha$  labels three cartesian components).

$$B_{rc\alpha}^{qc'\alpha'} = \frac{1}{\sqrt{N}} \exp(-i\mathbf{q} \cdot \mathbf{r}) \delta_{cc'} \delta_{\alpha\alpha'}. \quad (39)$$

Here the wave vector  $\mathbf{q}$  can take the values

$$\mathbf{q} = \sum_{i=1}^3 \mathbf{g}_i p_i / N_i \quad (40)$$

where  $p_i$  is an integer which can take on  $N_i$  different values, usually chosen so that the possible  $\mathbf{q}$ -vectors are those contained in the first Brillouin zone of the lattice.

The eigenvalues belonging to (39) of the transformation (38) are  $\exp(i\mathbf{q} \cdot \mathbf{a}_i)$ . They are already written in the form required by (29), so we can immediately write down expressions like (31) for the three operators  $\hat{O}_i$  which generate the transformations (38). The result is

$$\hat{O}_i = \sum_{qc} (\mathbf{q} \cdot \mathbf{a}_i) (\mathbf{b}_{qc}^\dagger \cdot \mathbf{b}_{qc}) \quad (41)$$

where the new destruction and creation operators are defined by

$$\left. \begin{aligned} \mathbf{b}_{qc} &= \frac{1}{\sqrt{N}} \sum_{\mathbf{r}} \exp(-i\mathbf{q} \cdot \mathbf{r}) \mathbf{b}_{rc} \\ \mathbf{b}_{qc}^\dagger &= \frac{1}{\sqrt{N}} \sum_{\mathbf{r}} \exp(i\mathbf{q} \cdot \mathbf{r}) \mathbf{b}_{rc}^\dagger. \end{aligned} \right\} \quad (42)$$

Considering the analogy between equation (41) and the definition of momentum proper in terms of operators generating continuous translations of matter, it is now natural to define the vector operator

$$\hbar \hat{\mathbf{q}} = \sum_{\mathbf{qc}} \hbar \mathbf{q} (\mathbf{b}_{\mathbf{qc}}^\dagger \cdot \mathbf{b}_{\mathbf{qc}}) \quad (43)$$

and to call it the pseudomomentum operator. The state  $b_{\mathbf{qc}}^{\alpha\dagger} |0\rangle$  is said to contain one phonon with cell position  $\mathbf{c}$ , polarisation  $\mathbf{e}_\alpha$  and pseudomomentum  $\hbar \mathbf{q}$ . All possible values of the lattice position operator  $\hat{\mathbf{r}}$  have the same probability in such a state, compare (42).

For further details, including a discussion of conservation laws, the reader is referred to <sup>(1)</sup>, <sup>(2)</sup> and <sup>(5)</sup>.

## 6. Local Rotation and Spin

Next we want to consider a common rotation of all field vectors around the equilibrium position of the atoms to which they belong (compare fig. 2). We could of course equally well consider an opposite rotation of the coordinate system  $\mathbf{e}_x$ ,  $\mathbf{e}_y$  and  $\mathbf{e}_z$  used to describe the field vector components.

The rotation in question belongs to the continuous group of vector rotations and is well known. Let us first consider a rotation with angle  $\theta$  around the  $z$ -axis  $\mathbf{e}_z$ . The transformation of the  $\mathbf{b}$ -vectors is then (we leave out the reference to the atoms ( $\mathbf{r}$  and  $\mathbf{c}$ ) the numbering of which is not changed)

$$b^{\alpha'} = L_{\alpha\beta} b^\beta \quad (44)$$

where

$$\{L_{\alpha\beta}\} = \begin{Bmatrix} \cos \theta & -\sin \theta & 0 \\ \sin \theta & \cos \theta & 0 \\ 0 & 0 & 1 \end{Bmatrix}. \quad (45)$$

Eigenvectors of this matrix and the corresponding eigenvalues are

$$\left. \begin{aligned} \{B_\alpha^{+1}\} &= \frac{1}{\sqrt{2}} \begin{Bmatrix} 1 \\ -i \\ 0 \end{Bmatrix}; & \{B_\alpha^0\} &= \begin{Bmatrix} 0 \\ 0 \\ 1 \end{Bmatrix}; & \{B_\alpha^{-1}\} &= \frac{1}{\sqrt{2}} \begin{Bmatrix} 1 \\ i \\ 0 \end{Bmatrix} \\ \lambda_1 &= \exp(i\theta) & \lambda_2 &= 1 & \lambda_3 &= \exp(-i\theta) \\ l_1 &= \theta & l_2 &= 0 & l_3 &= -\theta \end{aligned} \right\} \quad (46)$$

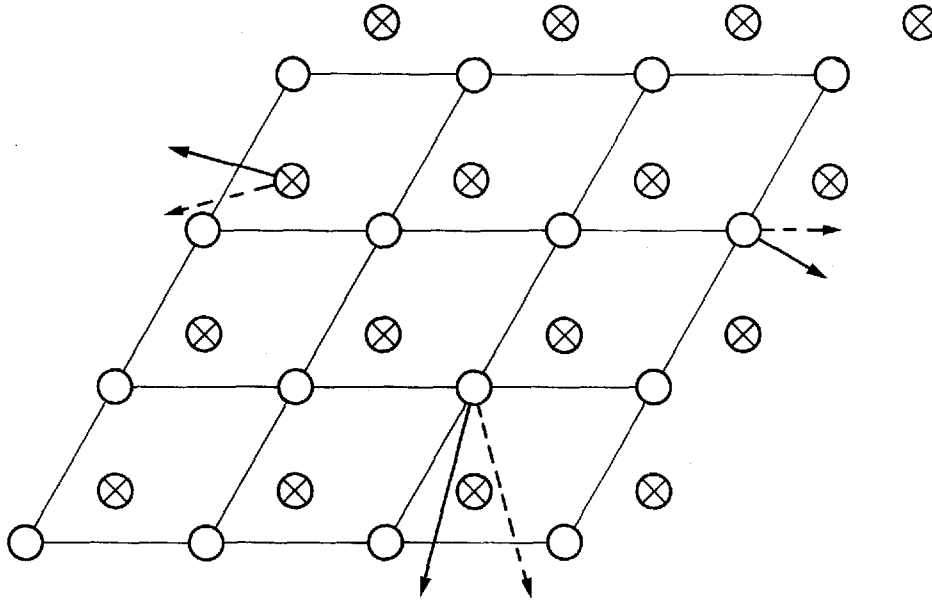


Fig. 2. Local rotation. The fully drawn and the dashed arrows are field vectors before and after the rotation.

Again proceeding as described in section 3 we find from (32) the new basic creation and destruction operators, the latter are:

$$\begin{pmatrix} b^{+1} \\ b^0 \\ b^{-1} \end{pmatrix} = \begin{pmatrix} \frac{1}{\sqrt{2}}(b^x - ib^y) \\ b^z \\ \frac{1}{\sqrt{2}}(b^x + ib^y) \end{pmatrix}. \quad (47)$$

From (46), (47) and (31) the  $\hat{O}$ -operator for this transformation is found to be

$$\hat{O} = \theta((b^{+1})^\dagger b^{+1} - (b^{-1})^\dagger b^{-1}) = -i\theta(\mathbf{b}^\dagger \times \mathbf{b})_z. \quad (48)$$

Combining this with the results for rotations around the two other axes we are led to define the axial vector operator

$$\hat{\mathbf{S}} = \frac{\hbar}{i} \sum_{rc} \mathbf{b}_{rc}^\dagger \times \mathbf{b}_{rc} \quad (49)$$

(we again introduce explicitly the summation over the atoms ( $\mathbf{r}$  and  $\mathbf{c}$ )).

Comparing with the theory of rotations for ordinary particles it is natural to identify  $\hat{\mathbf{S}}$  with an angular momentum, and because it is independent of the origin chosen for the position coordinates of the phonons, it should be called a spin angular momentum.

As the transformation generated by  $\hat{\mathbf{S}}$  is a continuous rotation it is to be expected that  $\hat{\mathbf{S}}$  fulfils the usual commutation relations for angular momentum. A straightforward calculation confirms this, particularly it is found that all one-phonon states are eigenfunctions of

$$\hat{\mathbf{S}}^2 = \hat{S}_x^2 + \hat{S}_y^2 + \hat{S}_z^2$$

with the eigenvalue  $2\hbar^2$ . The one-phonon eigenfunctions of  $\hat{S}_z$  are (leaving out  $\mathbf{r}$  and  $\mathbf{c}$ )

$$(b^{+1})^\dagger |0\rangle = \frac{1}{\sqrt{2}}(bx^\dagger + iby^\dagger) |0\rangle, \quad \text{eigenvalue } +\hbar,$$

$$(b^0)^\dagger |0\rangle = bz^\dagger |0\rangle, \quad \text{eigenvalue } 0,$$

$$(b^{-1})^\dagger |0\rangle = \frac{1}{\sqrt{2}}(bx^\dagger - iby^\dagger) |0\rangle, \quad \text{eigenvalue } -\hbar.$$

The phonon must consequently be said to be a spin one particle in conformity with its vectorial character. The state  $(b_{rc}^{+1})^\dagger |0\rangle$  is said to contain one phonon with  $\hat{S}_z = \hbar$  at the position  $\mathbf{r} + \mathbf{c}$ .

The  $\hat{F}$  operators (compare eq. (33)) generating local rotations of the displacements and momenta are well known from the quantum mechanics of ordinary particles. They are the components of the vector operator

$$\hat{\mathbf{F}} = \frac{\theta}{\hbar} \sum_{rc} (\mathbf{u}_{rc} \times \mathbf{p}_{rc}). \quad (50)$$

This is obtained from (35) or directly verified by using the commutation relations for the displacements and momenta. The operator  $\hat{\mathbf{F}}$  divided by  $\theta/\hbar$  is immediately recognized as a part of the angular momentum proper of the total system. When specialized to the continuum limit, it is identical with the spin-operator derived in ref. (3) by means of NOETHER'S theorem. However, in ref. (3) only the isotropic case is treated. The operators  $\hat{O}$  (48) and  $\hat{F}_z$  (50) are identical if both the kinetic and potential energy are invariant under local rotations. In the anisotropic case  $\hat{\mathbf{F}}$  (50) is not a one-phonon operator, and the term phonon spin operator should be reserved for the quantity (49).



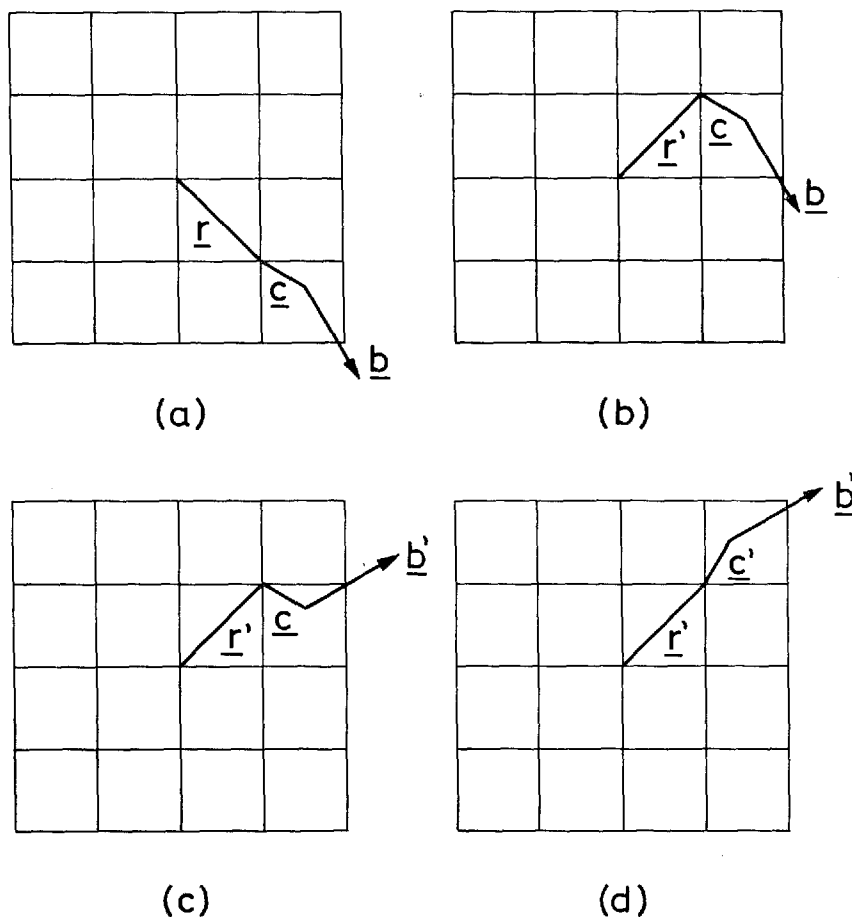


Fig. 3. A  $90^\circ$  rotation decomposed into successive rotations of the lattice vector  $\underline{r}$ , the field vector  $\underline{b}$  and the basis vector  $\underline{c}$ .

### 7. Point Group Rotation and Pseudo Angular Momentum

Let us consider a crystal, the atomic equilibrium positions of which form a structure with an  $n$ -fold axis of symmetry through a lattice point, the axis is called the  $z$ -axis.

We transform the excitation pattern of the crystal by rotating it through an angle  $2\pi/n$  around the  $z$ -axis. This transformation can be split into three commuting operations of the types c), b) and d) of section 4 (compare fig. 3)\*.

\* We only consider an infinite crystal. In order to give meaning to the transformation for a finite crystal we would have to choose a special form of the crystal and a special axis or to impose suitable cyclic boundary conditions on the transformation.

The second operation on fig. 3, which is possible for an arbitrary angle, has already been treated and has led to the introduction of the spin operator (49). The first operation and the third one are cyclic transformations very similar to the transformations which led to the concept of pseudomomentum. They permute in a cyclic way the field vectors belonging to  $n$  equivalent positions and first we need only consider the  $n$   $\mathbf{b}$ -vectors belonging to such a "star" of equivalent points. These points are labelled by the angle  $\theta$  (modulo  $2\pi$ ) between, say, the  $xz$ -plane and the normal from the point to the  $z$ -axis. For a definite "star"  $\theta$  takes values with intervals  $2\pi/n$ .

The transformation in question is then

$$\mathbf{b}'_{\theta} = \mathbf{b}_{\theta - 2\pi/n} \quad (51)$$

with the convention that

$$\mathbf{b}_{\theta} = \mathbf{b}_{\theta + 2\pi}.$$

A complete set of eigenvectors  $B_{\theta}^m$  and eigenvalues  $\lambda^m$  of the transformation (51) is (for one "star" and one direction of the field vectors)

$$B_{\theta}^m = \frac{1}{\sqrt{n}} e^{-im\theta} \quad (52)$$

$$\lambda^m = e^{i2\pi m/n} \quad (53)$$

where  $m$  is an integer which can take on  $n$  consecutive values. Following the scheme of section 3 we find that the  $\hat{O}$ -operator in question is

$$\hat{O} = \frac{2\pi}{n} \sum_m m \mathbf{b}_m^{\dagger} \cdot \mathbf{b}_m \quad (54)$$

where

$$\mathbf{b}_m = \frac{1}{\sqrt{n}} \sum_{\theta} \mathbf{b}_{\theta} e^{-im\theta}. \quad (55)$$

As  $2\pi/n$  in (54) is the angle of rotation and as the analogous rotation operator for an ordinary particle is  $1/\hbar$  times the product of the angle of rotation and the component along the axis of rotation of the orbital angular momentum operator it is natural to call the operator  $\hbar \sum_m m \mathbf{b}_m^{\dagger} \cdot \mathbf{b}_m$  the  $z$ -component of a "pseudo orbital angular momentum". The full expression for this operator becomes

$$\hat{L}_z = \hbar \sum_{ms} m \mathbf{b}_{ms}^{\dagger} \cdot \mathbf{b}_{ms} \quad (56)$$

where the summation index  $s$  runs over all distinct "stars" of  $n$  equivalent sites.

The resemblance of the pseudo orbital angular momentum to the pseudo-momentum is now evident: each "star" corresponds to a one-dimensional cyclic crystal with  $n$  lattice sites.

The full rotation shown on fig. 3 is generated by an operator

$$\exp\left(\frac{i 2\pi}{\hbar n} \hat{J}_z\right) = \exp\left(\frac{i 2\pi}{\hbar n} \hat{L}_z\right) \exp\left(\frac{i 2\pi}{\hbar n} \hat{S}_z\right). \quad (57)$$

The operator  $\hat{J}_z$  may be defined so that the three operators  $\hat{J}_z$ ,  $\hat{L}_z$  and  $\hat{S}_z$  commute and thus have simultaneous eigenfunctions. If their eigenvalues are denoted by  $\hbar m_j$ ,  $\hbar m_l$  and  $\hbar m_s$  respectively, we can only conclude from (57) that

$$m_j = m_l + m_s \text{ modulo } n. \quad (58)$$

A convenient choice of  $m_j$  would be such that it is limited to the same range of values as  $m_l$ .

If, e. g., we consider the case  $n = 4$ , the twelve independent one-phonon eigenstates of  $\hat{S}_z$ ,  $\hat{L}_z$  and  $\hat{J}_z$  of a single star can be classified by means of the following quantum numbers.

$m_s$	-1	0	1	-1	0	1	-1	0	1	-1	0	1
$m_l$	-1	-1	-1	0	0	0	1	1	1	2	2	2
$m_j$	2	-1	0	-1	0	1	0	1	2	1	2	-1

(59)

It is tempting to try to use  $\hbar \hat{r} \times \hat{q}$  (or rather  $\frac{1}{2} \hbar (\hat{r} \times \hat{q} + h.c.)$ ) as a "pseudo orbital angular momentum" but the operator does not generate rotations, not even in the limit of an infinite crystal (compare fig. 4). This is seen in the following way:

The operator  $\hat{r}$  is found to generate translations in the  $q$ -space just as  $\hat{q}$  generates translations in the  $r$ -space. In the limit of an infinite crystal  $\hat{q}$  has a continuum of eigenvalues and in this limit  $\hat{r}$  and  $\hbar \hat{q}$  resemble (in the  $q$ -representation) the usual position and momentum operators (in momentum representation) except for the fact that the eigenvalues of  $\hat{q}$  are limited to, say, the Brillouin zone. Denoting a vector in  $q$ -space by  $\mathbf{Q}$  it is therefore possible to make the identification

$$\hat{r} = i \frac{\partial}{\partial \mathbf{Q}}.$$

In order to stay within the space of functions which are periodic in the  $q$ -space the operator  $\hat{q}$  must be identified by a periodic function of  $\mathbf{Q}$  and not just by  $\mathbf{Q}$ . This explains why  $\hbar \hat{r} \times \hat{q}$  does not generate rotations except within a sphere which does not touch the Brillouin zone boundary and which has its centre at  $\mathbf{Q} = 0$ . The difficulties of  $\hbar \hat{r} \times \hat{q}$  are connected with the fact that the spherical harmonics are not orthogonal functions in the Brillouin zone.

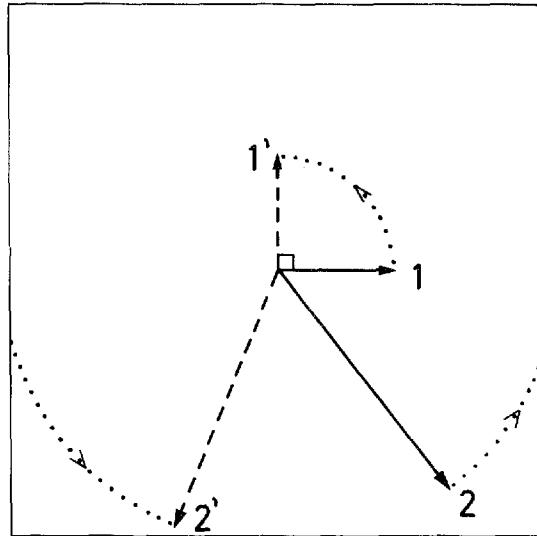


Fig. 4. The fully drawn arrows and the dashed ones show two wave vectors in the Brillouin zone before and after a  $90^\circ$  "rotation" generated by  $\hbar\hat{p} \times \hat{q}$ . Only the wave vector which does not touch the zone boundary during the "rotation" is really rotated.

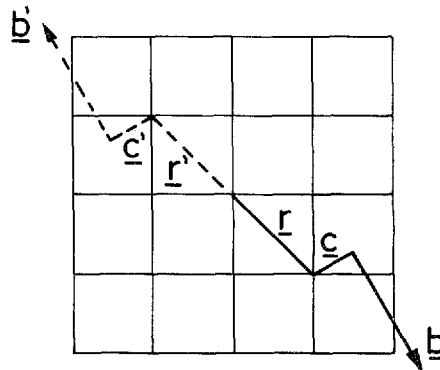


Fig. 5. Inversion of the excitation pattern.

## 8. Inversion and Parity

For a crystal whose atomic sites form a pattern with inversion symmetry, we can consider the following transformation of the excitation vectors (compare fig. 5).

$$\mathbf{b}'_{rc} = -\mathbf{b}_{-r-c}. \quad (60)$$

This can be split into an inversion without changing the direction of the vectors and a local inversion  $\mathbf{b} \rightarrow -\mathbf{b}$ . The latter leads to the definition of the *intrinsic parity* which clearly has the eigenvalue  $-1$  for a single phonon. The full intrinsic parity operator is the trivial operator  $(-1)^{\hat{n}}$ , where  $\hat{n}$  is the particle number operator.

The rest of the transformation (60), i. e.

$$\mathbf{b}'_{rc} = \mathbf{b}_{-r-c}, \quad (61)$$

leads to the definition of the concept of *extrinsic parity*. It is generated by a unitary operator with the eigenvalues  $+1$  and  $-1$  for states which are symmetric and antisymmetric respectively under the inversion (61). Of course one could introduce an  $\hat{O}$ -type operator in this case too in complete analogy with the case of rotation. It does not seem to be useful, however, so we shall not write it down explicitly.

If the crystal—not only the pattern of atomic sites—has inversion symmetry around  $\mathbf{r} = 0$  the total parity is conserved. The intrinsic parity, however, is not conserved when uneven anharmonic terms are present in the Hamiltonian.

## Appendix I

The usual harmonic Hamiltonian may be generalized so as to contain "mixed products" i. e. terms of the form  $p_r G_{rs} u_s$ . For simplicity only the following Hamiltonian will be treated (in this appendix bold face types as  $\mathbf{u}$  stand for a  $1 \times 3N$  column matrix)

$$\left. \begin{aligned} H &= \frac{1}{2}(\tilde{\mathbf{p}} - \tilde{\mathbf{u}}\tilde{A})(\mathbf{p} - A\mathbf{u}) + \frac{1}{2}\tilde{\mathbf{u}}D\mathbf{u} \\ A &= A^* = -\tilde{A} \quad D = D^* = \tilde{D} \\ 3N &\text{ degrees of freedom,} \\ D &\text{ is a positive definite matrix.} \end{aligned} \right\} \text{(I, 1)}$$

The term  $A\mathbf{u}$  may be the result of a homogeneous magnetic field around every single particle or a Coriolis field.

This Hamiltonian will be shown to describe a set of one dimensional harmonic oscillators just as the usual harmonic Hamiltonian. In order to show this the Heisenberg picture will be used.

The equations of motion are now

$$\left. \begin{aligned} \dot{\mathbf{u}} &= \mathbf{p} - A\mathbf{u} \\ \dot{\mathbf{p}} &= -(D - A^2)\mathbf{u} - A\mathbf{p}. \end{aligned} \right\} \text{(I, 2)}$$

This is a system of linear first order differential equations with constant coefficients. In order to show that these equations have a complete set of harmonic solutions new variables are introduced

$$\left. \begin{aligned} \mathbf{r} &= \mathbf{u} & \mathbf{u} &= \mathbf{r} \\ \mathbf{s} &= D^{-1/2}(\mathbf{p} + A\mathbf{u}) & \mathbf{p} &= D^{1/2}\mathbf{s} - A\mathbf{r} \end{aligned} \right\} \text{(I, 3)}$$

$$\begin{pmatrix} \dot{\mathbf{r}} \\ \dot{\mathbf{s}} \end{pmatrix} = \begin{pmatrix} -2A & D^{1/2} \\ -D^{1/2} & 0 \end{pmatrix} \begin{pmatrix} \mathbf{r} \\ \mathbf{s} \end{pmatrix} = \tilde{E} \begin{pmatrix} \mathbf{r} \\ \mathbf{s} \end{pmatrix}. \quad \text{(I, 4)}$$

The Hamiltonian may be expressed by  $\mathbf{r}$  and  $\mathbf{s}$  in the following ways

$$H = \frac{1}{2}(\tilde{\mathbf{r}}, \tilde{\mathbf{s}}) \tilde{\mathcal{E}} \mathcal{E} \begin{Bmatrix} \mathbf{r} \\ \mathbf{s} \end{Bmatrix} = \frac{1}{2}(\tilde{\mathbf{r}}, \tilde{\mathbf{s}}) \begin{Bmatrix} \dot{\mathbf{r}} \\ \dot{\mathbf{s}} \end{Bmatrix}. \quad (\text{I, 5})$$

The matrix  $i\mathcal{E}$  is a  $6N \times 6N$  dimensional and hermitian matrix and can consequently be diagonalized by a unitary matrix

$$\left. \begin{aligned} \mathcal{E}\Gamma &= i\Gamma\Omega & \Gamma^\dagger\Gamma &= \Gamma\Gamma^\dagger = 1 \\ \Omega &= \text{a diagonal matrix, } \Omega = \Omega^*. \end{aligned} \right\} (\text{I, 6})$$

The matrix  $D^{1/2}$  has no eigenvalue equal zero and eq. (I, 4) shows therefore that  $\mathcal{E}$  has no eigenvalue equal zero. It is also immediately seen that if  $(\xi_\nu, i\omega_\nu)$  are corresponding eigenvectors and eigenvalues of  $\mathcal{E}$ , then  $(\xi_\nu^*, -i\omega_\nu)$  are also corresponding eigenvectors and eigenvalues of  $\mathcal{E}$ . It is therefore possible to choose  $\Gamma$  and  $\Omega$  in the following way

$$\Gamma = \begin{Bmatrix} E & E^* \\ F & F^* \end{Bmatrix} \quad \Omega = \begin{Bmatrix} \omega & 0 \\ 0 & -\omega \end{Bmatrix} \quad (\text{I, 7})$$

$\omega$  is a  $3N \times 3N$ , positive definite and diagonal matrix.

In order to separate the Hamiltonian we introduce new variables  $\mathbf{f}$  and  $\mathbf{f}^\dagger$

$$\begin{Bmatrix} \mathbf{r} \\ \mathbf{s} \end{Bmatrix} = \Gamma \begin{Bmatrix} \mathbf{f}^\dagger \\ \mathbf{f} \end{Bmatrix}. \quad (\text{I, 8})$$

The operators  $(\mathbf{f}^\dagger)_\alpha$  and  $(\mathbf{f})_\alpha$  are hermitian conjugate operators because  $(\mathbf{r})_\beta$  and  $(\mathbf{s})_\beta$  are hermitian operators.

If  $\mathbf{u}$  and  $\mathbf{p}$  are replaced by  $\mathbf{f}^\dagger$  and  $\mathbf{f}$  then the Hamiltonian (I, 5) will be

$$H = \frac{1}{2}(\mathbf{f}^\dagger \omega^2 \mathbf{f} + \mathbf{f}^\dagger \omega^2 \mathbf{f}). \quad (\text{I, 9})$$

In order to show that  $H$  really is separated into one-dimensional systems, it is necessary to study the commutation relations of  $\mathbf{f}$  and  $\mathbf{f}^\dagger$ . After some lengthy calculations the following result is found

$$\left. \begin{aligned} [(\mathbf{f}^\dagger)_\alpha, (\mathbf{f}^\dagger)_\beta] &= [(\mathbf{f})_\alpha, (\mathbf{f})_\beta] = 0 \\ [(\mathbf{f})_\alpha, (\mathbf{f}^\dagger)_\beta] &= \hbar(\omega^{-1})_{\alpha\beta}. \end{aligned} \right\} (\text{I, 10})$$

Now destruction operators of phonons in stationary states can immediately be constructed

$$\left. \begin{aligned}
 b_\alpha &= \sqrt{\frac{\omega_\alpha}{\hbar}} (f)_\alpha \\
 H &= \hbar \sum_\alpha \omega_\alpha (b_\alpha^\dagger b_\alpha + \frac{1}{2}) \\
 [b_\alpha, b_\beta] &= 0 \quad [b_\alpha, b_\beta^\dagger] = \delta_{\alpha\beta}.
 \end{aligned} \right\} \text{(I, 11)}$$

In cases where the matrices  $A$  and  $D$  of the Hamiltonian (I, 1) commute it is possible to choose destruction operators for localized phonons of the form (8) where

$$M = (D - A^2)^{1/4} \quad \text{(I, 12)}$$

can be used (note that  $-A^2$  has no negative eigenvalues). In the general case it does not appear very useful to introduce localized phonons, at least not localized with respect to direction.

The whole question of phonons and magnetic fields is not an important one because the most essential result of a magnetic field will be a change of the electronic structure, i. e. a change of the dynamical matrix  $D$ . The reason for this is that the charge-mass ratio is much larger for the electrons than for the nuclei. It may, however, be of some importance to realize that a homogeneous magnetic field itself is not an anharmonic force.

## Appendix II

The most reasonable way to study how phonons are localized is to study how the corresponding excitation energy is localized. This means that the matrix elements of  $p_r p_s$  and  $u_r u_s$  must be studied. In the usual case where the matrix  $T$  (1) of the kinetic energy is a diagonal matrix,  $\pi_r$  and  $v_r$  (2) can evidently be used instead of  $p_r$  and  $u_r$  and this will be done here.

If  $|A\rangle$  is a normalized state vector containing a definite number of localized phonons (8) (9), i. e. if  $|A\rangle$  is an eigenvector of all operators  $b_r^\dagger b_r$ , then a short calculation gives these results:

$$\langle A | \pi_r \pi_s | A \rangle = \hbar \sum_t M_{rt} M_{ts} (\langle A | b_t^\dagger b_t | A \rangle + \frac{1}{2}) \quad \text{(II, 1)}$$

$$\langle A | v_r v_s | A \rangle = \hbar \sum_t (M^{-1})_{rt} (M^{-1})_{ts} (\langle A | b_t^\dagger b_t | A \rangle + \frac{1}{2}) \quad \text{(II, 2)}$$

$$\langle A | H | A \rangle = \hbar \sum_t (M^2)_{tt} (\langle A | b_t^\dagger b_t | A \rangle + \frac{1}{2}) \quad \text{(II, 3)}$$

$$\langle A | H | A \rangle = \sum_t \langle A | \pi_t \pi_t | A \rangle = \sum_{st} \langle A | v_s D_{st} v_t | A \rangle. \quad \text{(II, 4)}$$



It is seen that the energy of  $|A\rangle$  is, as expected, divided into two equal parts, a potential energy part and a kinetic energy part.

The excitation energy of a phonon created by  $b_u^\dagger$  from the state  $|A\rangle$  is now determined by the increment in the matrix elements (II, 1) and (II, 2) when  $b_u^\dagger b_u$  is increased by one. In particular, that part of the kinetic excitation energy which belongs to the  $r$ 'th degree of freedom is (from II, 1)

$$\delta E_{\text{kin}, r} = \delta \frac{1}{2} \langle A | \pi_r^2 | A \rangle = \frac{1}{2} \hbar (M_{ru})^2 \quad (\text{II, 5})$$

Thus the kinetic excitation energy of the phonon created by  $b_u^\dagger$  would be strictly localized if and only if  $M$  were a diagonal matrix; but then  $D = M^4$  would be a diagonal matrix too, and this is clearly not true. The kinetic energy connected with "localized" phonons is therefore not strictly localized, but if  $M_{rs}$  is small when the atoms to which  $r$  and  $s$  belong are rather far apart then it is quite reasonable to call the phonon localized. As a matter of fact in the case of sufficiently large systems  $D_{rs}$  will usually be small when the atoms to which  $r$  and  $s$  belong are sufficiently far apart, and this feature will, more or less, be preserved for functions of  $D$ , e. g.  $M = D^{1/4}$  (reasons for this will be given later).

Another way to study the localization of "localized phonons" would be to study the transitions between different states of localized phonons. This will lead to the study of the matrix elements of the Hamiltonian  $H$  or according to eq. (II, 3) the matrix elements of  $M^2$ .

From the preceding considerations it is seen that the localization of "localized phonons" is connected with functions of  $D$ . We shall not give any real proof but only sketch how one may investigate to what extent it follows that if  $D_{rs}$  is small when the distance between  $r$  and  $s$  is large enough then the same is, more or less, true for functions of  $D$ . We shall limit ourselves to the case where

$$D_{rs} = 0 \text{ when distance } (r, s) > d. \quad (\text{II, 6})$$

It follows immediately that

$$(D^n)_{rs} = 0 \text{ when distance } (r, s) > n \times d, \quad n = 1, 2, 3, \dots \quad (\text{II, 7})$$

Polynomials of  $D$  will therefore have the wanted property.

If the eigenvalues of  $D$  are such that a function  $f(x)$  may be approximated by a polynomial of  $x$  for  $x$  equal to any of the eigenvalues of  $D$ , then  $f(D)$  may be approximated by the same polynomial in  $D$  instead of  $x$ . This is most easily seen in a representation where  $D$  is a diagonal matrix. Approxima-

tion by polynomials gives now an explanation of the said property of functions of  $D$ .

Instead of giving examples using the approximation by polynomials only a few numerical results will be given. For some simple models of crystals it has been calculated how much of the kinetic excitation energy of a localized phonon belongs to the same degree of freedom as the phonon; this means (see II, 5) that  $(M_{rr})^2/(M^2)_{rr}$  has been calculated. The examples are:

1) A harmonic, infinite, one-dimensional and diatomic crystal with nearest neighbour interaction. The masses are  $m_1$  and  $m_2$ , and the phonon is localized at the atom with mass  $m_1$ . Except for  $m_1/m_2 = 1$  numerical integration is necessary.

$m_1/m_2$	0.1	0.5	1	2	10
$(M_{11})^2/(M^2)_{11}$	0.978	0.938	0.914	0.892	0.865

It is seen that a phonon is better localized at a light atom than at a heavy atom, but the kinetic energy part of the excitation energy is in all cases quite well localized.

2) A harmonic, infinite, two-dimensional and hexagonal crystal with one atom in the basis and nearest neighbour interaction.

Numerical calculations have shown that

$$(M_{rr})^2/(M^2)_{rr} \approx 0.97.$$

3) This is a Debye model with a "spherical" Brillouin zone and only one sound velocity. The model is not very realistic but particularly in one and three dimensions many calculations are easily performed.

	1-dim.	2-dim.	3-dim.
$(M_{rr})^2/(M^2)_{rr}$	8/9	24/25	48/49

Although the interaction range of a Debye model is long it is found that the kinetic excitation energy of a localized phonon is well localized. Note that the degree of localization increases with the number of dimensions.

### Appendix III

Below is shown that the operator  $\hat{O}$  (31) generates the transformation (27) of the destruction operators  $b_r$ .

First we define by induction an operator  $[A, B]^{(n)}$  for all integers  $n \geq 0$  ( $A$  and  $B$  are usual operators)

$$\begin{aligned} [A, B]^{(0)} &= B & [A, B]^{(1)} &= AB - BA \\ [A, B]^{(n+1)} &= [A, [A, B]^{(n)}]^{(1)}. \end{aligned}$$

We have now the following identity\*

$$\exp(A)B \exp(-A) = \sum_{n=0}^{\infty} \frac{1}{n!} [A, B]^{(n)} \quad (= \exp([A, B])). \quad (\text{III, 1})$$

Here the last expression is to be understood in a purely formal way.

A proof of the identity is easily constructed using the formula

$$[A, B]^{(n)} = \sum_{\nu=0}^n (-1)^{n+\nu} \binom{n}{\nu} A^{\nu} B A^{n-\nu} \quad (\text{III, 2})$$

which is found by induction.

Using the commutation relations (10) for  $b_{\alpha}$  and  $b_{\alpha}^{\dagger}$  (32) it is found by induction that

$$[b_{\alpha}^{\dagger} b_{\alpha}, b_{\beta}]^{(n)} = (-1)^n b_{\alpha} \delta_{\alpha\beta} \quad n > 0.$$

The transformation of  $b_{\alpha}$  is now immediately accessible

$$\exp(i\hat{O})b_{\alpha} \exp(-i\hat{O}) = \exp(-il_{\alpha})b_{\alpha} = \lambda_{\alpha}^* b_{\alpha}.$$

The transformation of  $b_r$  is consequently

$$\exp(i\hat{O})b_r \exp(-i\hat{O}) = \sum_{\alpha} B_r^{\alpha*} \lambda_{\alpha}^* b_{\alpha} = \sum_{\alpha st} L_{rs} B_s^{\alpha*} B_r^{\alpha} b_t = \sum_s L_{rs} b_s \quad \text{q. e. d.}$$

\* See F. HAUSDORFF (7). A more recent treatment is given by WILHELM MAGNUS et al. (8).

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