

*INELASTIC SCATTERING OF SLOW NEUTRONS IN ARBITRARY CRYSTALS AND THE
GENERAL PROBLEM OF RECONSTRUCTING THE PHONON SPECTRUM*

Yu. KAGAN

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The scattering of slow neutrons in crystals of arbitrary symmetry and with an arbitrary number of atoms per unit cell is treated. A rigorous analysis is given of the inverse problem of reconstructing the phonon spectrum (without using any model). A possibility for separating coherent and incoherent scattering is pointed out. It is found that to determine the frequency distribution of the phonon spectrum one must make measurements of incoherent scattering on crystals of different isotopic compositions, where the number of such different crystals must be no less than the number of different atoms in a unit cell. It is shown that it is possible, from measurements of inelastic coherent scattering, to reconstruct (aside from the dispersion law) the polarization vectors for arbitrary values of the wave vector in the reciprocal lattice.

1. INTRODUCTION

THE development of experimental methods related to the study of inelastic scattering of slow neutrons by macroscopic systems has made practical the study of the dynamics of condensed systems. The question naturally arises as to the maximum information that can be obtained from such experiments. We should state immediately that we are speaking of the rigorous inverse problem, where the passage from the results of measurements of differential cross sections for inelastic scattering of neutrons to information about the condensed system does not require the use of any specific model for describing the interaction between the individual particles of the system.

In the present paper we give a complete analysis of the inverse problem for the phonon branch of excitations in crystals of arbitrary symmetry and with arbitrary structure of the elementary cell. We assume that the conditions are such that one can, with sufficient accuracy, speak of a system of independent excitations (for example, temperatures are restricted so that the interactions between phonons are small, etc.).

In investigating the phonon branch of excitations, one should distinguish the following four types of information (each succeeding description contains the ones which precede it):

1. The distribution function $\psi(\omega)$ of frequencies in the phonon spectrum. The function $\psi(\omega)$ provides complete knowledge for the thermodynamic description of the crystal.

2. The dispersion law $\omega(\mathbf{f}, \alpha)$ for the phonon spectrum for all branches α and all values of \mathbf{f} in one elementary cell of the reciprocal lattice.

3. The dispersion law and a particular type of bilinear combination of projections of polarization vectors $\mathbf{v}_j(\mathbf{f}, \alpha)$ along the direction of the wave vector \mathbf{f} . These bilinear combinations appear in all problems associated with inelastic scattering by phonons when one-phonon processes are predominant. For a wide class of problems (scattering of x rays, electrical conductivity, etc), this type of information is complete.

4. The dispersion law and the value in the general case of the complex polarization vectors $\mathbf{v}_j(\mathbf{f}, \alpha)$ for all values of \mathbf{f} and α and for all atoms (with index j) in the unit cell.* In this case we obviously have a complete description of the phonon branch of excitations.

Thus the problem is to determine the conditions under which a measurement of the differential cross section for inelastic scattering of "cold" neutrons would rigorously determine one of these descriptions of the phonon spectrum of crystals.

2. GENERAL FORMULAS

Let us consider an arbitrary crystal. We write the general expression for the interaction of a neutron with a crystal lattice in the form (cf. [1])

$$V(\mathbf{r}) = \sum_{l,j} [A_{lj}I + B_{lj}(\mathbf{S}\mathbf{K}_{lj})] \delta(\mathbf{r} - \mathbf{R}_{lj}). \quad (1)$$

*Unless otherwise stated specifically, we always deal with values of the wave vector \mathbf{f} restricted to the first cell of the reciprocal lattice.

Here \mathbf{S} is the neutron spin, \mathbf{K}_j is the spin of the j -th nucleus and A_j and B_j are constants which are nuclear parameters. The index l numbers the unit cell and j labels the atoms in a given cell.

The expression for \mathbf{R}_{lj} can be written as

$$\mathbf{R}_{lj} = \mathbf{r}_l + \boldsymbol{\rho}_j + \mathbf{u}_{lj}, \quad (2)$$

where $\mathbf{r}_l + \boldsymbol{\rho}_j$ is the equilibrium position of the j -th nucleus in the l -th unit cell. For the displacement \mathbf{u}_{lj} of an arbitrary nucleus, we have the expression

$$\mathbf{u}_{lj} = \sum_{\mathbf{f}, \alpha} \left(\frac{\hbar}{2M_j \omega(\mathbf{f}, \alpha) N} \right)^{1/2} [\mathbf{v}_j(\mathbf{f}, \alpha) \exp\{i\mathbf{f}\mathbf{r}_l\} a(\mathbf{f}, \alpha) + \mathbf{v}_j^*(\mathbf{f}, \alpha) \exp\{-i\mathbf{f}\mathbf{r}_l\} a^*(\mathbf{f}, \alpha)], \quad (3)$$

where $a(\mathbf{f}, \alpha)$ and $a^*(\mathbf{f}, \alpha)$ are operators for annihilation and creation of a phonon, M_j is the mass of the j -th nucleus and N is the number of unit cells in the crystal. The complex polarization vectors $\mathbf{v}_j(\mathbf{f}, \alpha)$ are restricted by the conditions

$$\sum_{i,j} v_j^i(\mathbf{f}, \alpha) v_j^{i*}(\mathbf{f}, \alpha') = \delta_{\alpha\alpha'}, \quad \sum_{\alpha} v_j^i(\mathbf{f}, \alpha) v_j^{k*}(\mathbf{f}, \alpha) = \delta_{ik} \delta_{ij} \quad (4)$$

(where the superscripts denote projections of the vector).

Let us consider the matrix element of (1) corresponding to the scattering of the neutron with transfer to the lattice of momentum $\boldsymbol{\kappa} = \mathbf{k}_0 - \mathbf{k}'$ and simultaneous excitation of p phonons. We use the standard procedure (cf., for example, [1]) for this purpose, making use of the explicit form of (2) and (3). We average the square of the matrix element over the initial state of the crystal, assume thermal equilibrium and absence of correlations between spins of individual nuclei:

$$\begin{aligned} |M_p|^2 = & \left| \sum_{l,j} A_{lj} e^{-W_j/2} \exp\{i\boldsymbol{\kappa}\boldsymbol{\rho}_j + i\boldsymbol{\kappa}\mathbf{r}_l\} \right. \\ & \times \prod_{\beta=1}^p (\boldsymbol{\kappa}\mathbf{v}_j(\beta)) \left(\frac{\hbar(\bar{n}(\beta) + 1/2 \pm 1/2)}{2M_j \omega(\beta) N} \right)^{1/2} \exp\{\mp i\mathbf{f}_\beta \mathbf{r}_l\} \left. \right|^2 \\ & + \frac{1}{4} \sum_{l,j} K_{l,j} (K_{l,j} + 1) B_{l,j}^2 e^{-W_j} \left(\frac{R_j}{N} \right)^p \prod_{\beta=1}^p \frac{|\mathbf{q}\mathbf{v}_j(\beta)|^2}{\hbar\omega(\beta)} (\bar{n}(\beta) \\ & + 1/2 \pm 1/2), \end{aligned} \quad (5)$$

$$W_j = R_j \sum_{\alpha} \frac{|\mathbf{q}\mathbf{v}_j(\mathbf{f}, \alpha)|^2}{\hbar\omega(\mathbf{f}, \alpha)} (2\bar{n}(\mathbf{f}, \alpha) + 1). \quad (5')$$

We have used the notation: $R_j = \hbar^2 \kappa^2 / 2M_j$ for the recoil energy of a free nucleus of mass M_j , $\mathbf{q} = \boldsymbol{\kappa} / \kappa$. The upper sign corresponds to emission, the lower sign to absorption of the phonon. (A summation has been carried out in (5) over the two spin projections of the neutron after the scattering.)

We average in (5) over the isotopic composition for each of the types of atoms in the unit cell. In

doing this we shall assume that there is a complete absence of correlation between the positions in the crystal of individual isotopes, including also their locations within a unit cell. The result is

$$\begin{aligned} |\overline{M_p}|^2 = & N^2 \left| \sum_j \overline{A_j} \exp\{-W_j/2\} \exp\{i\boldsymbol{\kappa}\boldsymbol{\rho}_j\} \right. \\ & \times \prod_{\beta=1}^p (\boldsymbol{\kappa}\mathbf{v}_j(\beta)) \left(\frac{\hbar(\bar{n}(\beta) + 1/2 \pm 1/2)}{2M_j \omega(\beta) N} \right)^{1/2} \Delta(\boldsymbol{\kappa} \mp \mathbf{f}_1 \mp \dots \mp \mathbf{f}_p \\ & + 2\pi\mathbf{b}) + N \sum_j C_j e^{-W_j} \left(\frac{R_j}{N} \right)^p \prod_{\beta=1}^p \frac{|\mathbf{q}\mathbf{v}_j(\beta)|^2}{\hbar\omega(\beta)} (\bar{n}(\beta) \\ & + 1/2 \pm 1/2), \end{aligned} \quad (6)$$

$$C_j = \frac{1}{4} K_j (K_j + 1) B_j^2 + (\overline{A_j})^2 - (\overline{A_j})^2. \quad (7)$$

We now transform from (6) to the differential scattering cross section referred to a unit cell:

$$\frac{d^2\sigma(\Delta E, \boldsymbol{\kappa})}{d\epsilon d\Omega} = \sum_p \frac{d^2\sigma^p(\Delta E, \boldsymbol{\kappa})}{d\epsilon d\Omega}, \quad (8)$$

$$\begin{aligned} \frac{d^2\sigma^p(\Delta E, \boldsymbol{\kappa})}{d\epsilon d\Omega} = & \frac{m^2 k' N^{p-1}}{4\pi^2 \hbar^4 k_0} \frac{v_0^p}{(2\pi)^{3p}} \frac{1}{\rho!} \mathbf{S} \sum_{\alpha_1, \dots, \alpha_p} \int d^3f_1 \dots d^3f_p \\ & \times |\overline{M_p}|^2 \delta(\Delta E \mp \hbar\omega(\mathbf{f}_1, \alpha_1) \mp \dots \mp \hbar\omega(\mathbf{f}_p, \alpha_p)). \end{aligned} \quad (9)$$

Here v_0 is the volume of the unit cell; \mathbf{S} is a summation over all the different combinations of absorbed and emitted phonons, and $\Delta E = \hbar^2(k_0^2 - k'^2) / 2m$ is the energy transferred to the lattice.

We shall be interested only in lattices containing relatively heavy nuclei. In this case, for suitably low temperatures the contribution to the differential cross section (8) of the terms with $p \geq 2$ is small. Because of this, for $\Delta E \neq 0$ the value of $d^2\sigma/d\epsilon d\Omega$ will be determined mainly by the cross section for one-phonon scattering. We shall write this cross section explicitly, separating the coherent [first term in (6)] and incoherent [second term in (6)] parts of the scattering:

$$\frac{d^2\sigma^1(\Delta E, \boldsymbol{\kappa})}{d\epsilon d\Omega} = \frac{d^2\sigma_{\text{coh}}^1(\Delta E, \boldsymbol{\kappa})}{d\epsilon d\Omega} + \frac{d^2\sigma_{\text{inc}}^1(\Delta E, \boldsymbol{\kappa})}{d\epsilon d\Omega}, \quad (10)$$

$$\begin{aligned} \frac{d^2\sigma_{\text{coh}}^1(\Delta E, \boldsymbol{\kappa})}{d\epsilon d\Omega} = & \frac{m^2 k' \kappa^2}{8\pi^2 \hbar^3 k_0} \sum_{\alpha} \left| \sum_j (A_j / \sqrt{M_j}) e^{-W_j/2} \right. \\ & \times \exp\{i\boldsymbol{\kappa}\boldsymbol{\rho}_j\} \mathbf{q}\mathbf{v}_j(\boldsymbol{\kappa}, \alpha) \left. \right|^2 \\ & \times \omega^{-1}(\boldsymbol{\kappa}, \alpha) [(\bar{n}(\Delta E) + 1) \delta(\Delta E - \hbar\omega(\boldsymbol{\kappa}, \alpha)) \\ & + \bar{n}(\Delta E) \delta(\Delta E + \hbar\omega(\boldsymbol{\kappa}, \alpha))], \end{aligned} \quad (11)$$

$$\begin{aligned} \frac{d^2\sigma_{\text{inc}}^1(\Delta E, \boldsymbol{\kappa})}{d\epsilon d\Omega} = & \frac{m^2 k' \kappa^2}{8\pi^2 \hbar^3 k_0} \frac{v_0}{(2\pi)^3} \sum_j (C_j / M_j) e^{-W_j} \sum_{\alpha} \int d^3f \\ & \times \frac{|\mathbf{q}\mathbf{v}_j(\mathbf{f}, \alpha)|^2}{\hbar\omega(\mathbf{f}, \alpha)} [(\bar{n}(\Delta E) + 1) \delta(\Delta E - \hbar\omega(\mathbf{f}, \alpha)) \\ & + \bar{n}(\Delta E) \delta(\Delta E + \hbar\omega(\mathbf{f}, \alpha))]. \end{aligned} \quad (12)$$

3. SEPARATION OF INCOHERENT AND COHERENT SCATTERING

To solve the general inverse problem, it is very important to be able to separate out consistently the purely incoherent scattering. At first glance it might seem that this is impossible rigorously. In fact, if as is usually done in experiments, one fixes \mathbf{k}_0 and considers the scattering cross section for a given direction as a function of \mathbf{k}' , then letting ΔE run from 0 to $\hbar\omega_{\max}$ (where ω_{\max} is the maximum frequency in the phonon spectrum of the crystal), we unavoidably measure incoherent as well as coherent scattering. This statement is rigorously valid for any direction.

However, the differential cross sections for coherent and incoherent scattering are functions of ΔE and κ , where, most important, in the general case the vector and scalar variables are independent quantities. This being the case, one can fix κ and study the cross section as a function of ΔE .

Let us set $\kappa = 2\pi\mathbf{b}$, where \mathbf{b} is a reciprocal lattice vector. It then follows from (11) that the cross section for coherent one-phonon scattering will be different from zero only when

$$\Delta E = \pm \hbar\omega(2\pi\mathbf{b}, \alpha) = \pm \hbar\omega(0, \alpha). \quad (13)$$

(The last part of the equation is a consequence of the periodicity of $\omega(\mathbf{f}, \alpha)$ in the reciprocal lattice space.) For the acoustic branches, $\omega(0, \alpha) = 0$. Thus in the case of an arbitrary monatomic lattice, by measuring $d^2\sigma^1(2\pi\mathbf{b}, \Delta E)/d\epsilon d\Omega$ for $\Delta E \neq 0$, we measure the cross section for the purely incoherent scattering, i.e., (12). Thus for monatomic lattices, one can carry out a completely rigorous separation.

If there is more than one atom per unit cell, then in addition to the acoustic branches there will also be optical branches, for which $\omega(0, \alpha) \neq 0$. If the minimum value of the frequency in the optical branches is greater than $\omega_{\max}^{\text{ac}}$ for the acoustic branches, then for $\Delta E \leq \omega_{\max}^{\text{ac}}$ the scattering will be purely incoherent. For the interval ΔE itself, which corresponds to the optical branches, there is a superposition of both types of scattering. In the general case one can unfortunately not claim that $\omega_{\max}^{\text{ac}} < \omega_{\max}^{\text{opt}}$, so that a superposition can occur even within an interval ΔE corresponding to the acoustic branches.

However, the coherent scattering corresponding to the frequencies $\omega(0, \alpha)$ of the optical branches will give rise to narrow sharp peaks, especially if one includes the nature of the dependence $\omega(\kappa, \alpha)$ near $\kappa = 0$. Thus, with sufficiently good resolution it should be possible to get a clear

resolution of the coherent scattering at the frequencies $\omega(0, \alpha)$ and thus obtain the incoherent scattering cross section over the whole range of frequencies of the phonon spectrum.

We note that to get the clearest separation of the coherent scattering by the optical frequencies $\omega(0, \alpha)$, one can make measurements simultaneously for $\kappa \neq 2\pi\mathbf{b}$, but $\kappa \parallel \mathbf{b}$. If we then divide the two results by κ^2 , we should have identical curves, except for the coherent scattering bumps, which in general occur at different values of ΔE . From a comparison of the curves one can also directly establish the behavior of the incoherent scattering near the optical frequencies $\omega(0, \alpha)$.

In using the method of separation based on fixing $\kappa = 2\pi\mathbf{b}$,* the interval ΔE of possible changes can be chosen to cover the whole phonon spectrum. Then in order not to work with small scattering angles, it is expedient to work with the minimum \mathbf{b} when $\hbar\omega_{\max} \lesssim \hbar^2 |2\pi\mathbf{b}_{\min}|^2/2m$ and to change to larger values of the reciprocal lattice vector when $\hbar\omega_{\max} \gg \hbar^2 |2\pi\mathbf{b}_{\min}|^2/2m$. As one runs through the region of small ΔE , one can reduce the value of \mathbf{b} and thus avoid problems of small angles while still preserving the separation.

Measurements at fixed κ introduce significant complications into the experiment. Because of this we point out two approximate methods for the separation, which are much simpler.

The first method is measurement at small scattering angles. Without going into detail about this method, we mention only that when observing at sufficiently small angles, the coherent scattering by acoustic phonons may be "shoved" into the region of sound frequencies. Then for ΔE outside this range, the scattering will be completely incoherent if there is a single atom in the unit cell. If, however, the crystal has optical branches, one must also make auxiliary measurements at large angles in order to determine the coherent scattering on optical phonons at frequencies far from $\omega(0, \alpha)$. A comparison of the cross sections as functions of ΔE for both cases enables an approximate separation of the purely incoherent scattering.

The second method is simply the comparison of the $d^2\sigma/d\epsilon d\Omega$ curves for several fixed directions of \mathbf{k}' . Since the coherent scattering will in general occur in different frequency intervals, there is a possibility of separating out the incoherent scattering for the whole frequency range. This

*Experimentally this method corresponds to the "constant κ method" developed by Brockhouse for determining the frequencies of different branches, corresponding to a fixed wave vector.

method is the simplest, but it is also the least accurate. It obviously imposes special requirements on the resolution.

4. INCOHERENT SCATTERING

Let us specifically consider the inverse problem and start with the analysis of the information given by the incoherent scattering cross section, obtained either from a separation of the cross section for scattering by a single crystal, described in the preceding section (which we shall call case 1) or directly, if the incoherent scattering predominates (case 2). In the latter case, as we shall see later, one can use results obtained with either single crystals or polycrystals.

Let us turn to expression (12). As already remarked, in this paper we are considering only lattices consisting of relatively heavy nuclei and at reasonable temperatures. In this case, $W_j \ll 1$ for all f , and we commit a small error if for each group of identical atoms we replace e^{-W_j} by its average value. We introduce the notation

$$\frac{v_0}{(2\pi)^3} \sum_j \sum_\alpha \int d^3f \frac{|q \mathbf{v}_j(\mathbf{f}, \alpha)|^2}{\hbar \omega(\mathbf{f}, \alpha)} [(\bar{n}(\Delta E) + 1) \delta(\Delta E - \hbar \omega(\mathbf{f}, \alpha)) + \bar{n}(\Delta E) \delta(\Delta E + \hbar \omega(\mathbf{f}, \alpha))] = q^i q^k T_\xi^{ik}. \quad (14)$$

The summation extends over all atoms of type ξ . If we denote the factor in front of the summation sign in (12) by β , then (12) can be rewritten as

$$\frac{1}{\beta} \frac{d^2 \sigma_{\text{inc}}^1(\Delta E, \kappa)}{d\epsilon d\Omega} = q^i q^k \sum_\xi \frac{C_\xi}{M_\xi} \overline{\exp\{-W_\xi\}} T_\xi^{ik}(\Delta E). \quad (15)$$

In the same approximation in which we neglect two-phonon processes, we can replace $\exp\{-W_\xi\}$ by its value from any simple model. It then follows from (15) that the determination of $q^i q^k T_\xi^{ik}$ for all ξ requires the knowledge of the cross section for samples with different sets of C_ξ , and that the number of such sets must be the same as the number of different sorts of atoms in the unit cell. Obviously a change in C_ξ can be accomplished by an appropriate change of isotopic composition.

It is not difficult to show that the second-rank tensor

$$T^{ik}(\Delta E) = \sum_\xi T_\xi^{ik}(\Delta E) \quad (16)$$

will have symmetry properties which are completely determined by the symmetry of the crystal. If for simplicity we exclude from consideration crystals of the monoclinic and triclinic systems, then by reducing the tensor (16) to the principal axes we will know both the number of independent components as well as the actual directions of all the principal axes. Then, in case 1 by measuring

the scattering cross section for three values $\kappa = 2\pi\mathbf{b}$, which are not coplanar (for all isotopic compositions), one can find the eigenvalues of the tensor T^{ik} and thus the trace, $\text{Sp}(T)$.

From (16) and (14) it follows that $\text{Sp}(T)$ contains $\sum_j |\mathbf{v}_j(\mathbf{f}, \alpha)|^2$. But according to (4) this quantity is equal to unity for all \mathbf{f} and α . Consequently we find

$$\text{Sp}(T(\Delta E)) = \frac{3}{\hbar} g \psi\left(\frac{\Delta E}{\hbar}\right) [\bar{n}(\Delta E) + 1/2 \pm 1/2], \quad (17)$$

where g is the number of atoms in the unit cell, and $\psi(\omega)$ is the distribution function for the frequencies in the phonon spectrum, normalized to unity.

Thus the determination of $\text{Sp}(T(\Delta E))$ automatically gives a determination of the frequency distribution function for the phonon spectrum of an arbitrary crystal.

For uniaxial crystals the number of independent wave vectors $\kappa = 2\pi\mathbf{b}$ can be reduced to two, while in the case of cubic symmetry one value of the wave vector is sufficient.

In case 2 the problem simplifies considerably. In fact, by measuring the differential cross section (15), at each isotopic composition, with the direction of \mathbf{k}' fixed (but its magnitude varying) for three positions of the single crystal differing by cyclic permutations of the coordinates, one finds immediately (cf., [3])

$$\sum T_\xi^{ik}(\Delta E) q^i q^k = \text{Sp}(T_\xi(\Delta E)) \quad (18)$$

(where the summation is over the three measurements). Equation (18) is valid for any crystal symmetry, including the monoclinic and triclinic systems. Summing (18) over ξ , we get $\text{Sp}(T)$, and thus also $\psi(\Delta E/\hbar)$.

If the sample is polycrystalline, then (neglecting the anisotropy of W_ξ) the averaging in (18) occurs automatically. Thus to determine the frequency distribution function in case 2 it is sufficient to take polycrystalline samples which have different isotopic compositions and measure $d^2\sigma/d\epsilon d\Omega$ as a function of ΔE for just a single direction of scattering.

If there are identical atoms in the unit cell, a single isotopic composition is sufficient for determining $\psi(\omega)$. For case 2 the corresponding problem was treated earlier. [3] The special case of a cubic crystal with one atom in the unit cell was investigated in detail in the well-known paper of Placzek and Van Hove, [4] where they actually established for the first time the connection between the incoherent scattering cross section and the function $\psi(\omega)$.

We note that for fixed \mathbf{q} , it follows from (14) and (5') that

$$q^i q^k \int T_{\xi}^{ik}(\Delta E) d\Delta E = \frac{1}{R_{\xi}} \sum_{j(\xi)} W_j(\mathbf{q}). \quad (19)$$

Since, for $W_j \ll 1$,

$$\overline{\exp\{-W_j\}} \approx \exp\{-\overline{W_j}\},$$

equation (19) provides the possibility of taking account of the different Debye-Waller factors more correctly in the next approximation.

In concluding this section, we should especially emphasize that the use of several isotopic compositions in the case of a unit cell with several types of atoms is important in principle. It is impossible, by playing with angles and using the dependence of the cross section on κ , to make up for the insufficiency of information caused by the presence of incoherent scattering, when one uses only one isotopic composition. This is related to the fact that the square modulus of the polarization vector for each atom in the unit cell depends essentially on \mathbf{f} and α , and as we see from (4) such a dependence disappears only for the sum $\sum_j |\mathbf{v}_j|^2$. The question

of the relation between the polarization vectors cannot be solved in any way without introducing some definite model. Thus in particular, from measurements of the incoherent scattering for a crystal with a complex unit cell at a single isotopic composition one cannot in general draw any unique conclusions concerning the frequency distribution function.

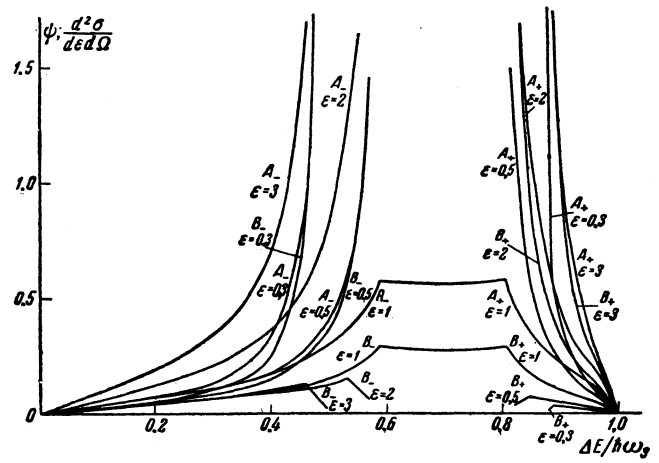
In order to make this assertion particularly clear, let us consider a primitive cubic lattice with two atoms in the unit cell, and include nearest-neighbor interactions of both central and non-central type. Such a simplified type of lattice was described in detail in [5].

Suppose that the incoherent scattering from one of the atoms in the unit cell is small compared to that from the other (we denote the latter by 1). Then we have [cf. Eq. (15)]:

$$\frac{M_1}{\beta C_1} e^{W_1} \frac{d^2 \sigma_{\text{inc}}^1}{d\epsilon d\Omega} = \frac{v_0}{(2\pi)^3} \sum_{\alpha=1}^6 \int d^3 f \frac{|\mathbf{q}\mathbf{v}_1(\mathbf{f}, \alpha)|^2}{\hbar\omega(\mathbf{f}, \alpha)} \overline{n\delta}(\Delta E + \hbar\omega(\mathbf{f}, \alpha)) \quad (20)$$

(where, for simplicity, we have taken the case of $\Delta E < 0$). For such a crystal, the three acoustic branches and the three optical branches coincide, and the polarization vectors are along the cubic axes. As a result the right side of (20) simplifies and reduces to

$$\frac{v_0}{(2\pi)^3} \sum_{\alpha=1,2} \int d^3 f \frac{|\mathbf{v}_1(\mathbf{f}, \alpha)|^2}{\hbar\omega(\mathbf{f}, \alpha)} \overline{n\delta}(\Delta E + \hbar\omega(\mathbf{f}, \alpha')) \quad (21)$$



A_- and A_+ are frequency distribution functions for the acoustic and optical branches, respectively. B_- and B_+ are the differential scattering cross sections corresponding to the acoustic and optical branches (the right side of (23)).

where $\alpha' = 1$ corresponds to the acoustic, and $\alpha' = 2$ to the optical branches.

One can show (cf. [6]) that on this model $|v_1|^2$ for each branch depends only on ω^2 and is equal to

$$|v_1|^2 = \frac{(\omega^2/\omega_3^2)(1 + \epsilon) - 1}{2(\omega^2/\omega_3^2)(1 + \epsilon) - (1 + \epsilon)}, \quad (22)$$

where $\epsilon = M_2/M_1$, $\omega_3^2 = 2(\gamma_1 + 2\gamma_2)(1/M_1 + 1/M_2)$ (where γ_1 and γ_2 are the force constants for the central and noncentral interactions, respectively).

Going over to the frequency distribution function $\psi_{\alpha'}(\omega)$ for the separate branches, we find from (21) and (22),

$$\frac{\hbar M_1}{C_1 \beta} e^{W_1} \frac{d^2 \sigma_{\text{inc}}^1}{d\epsilon d\Omega} = \sum_{\alpha'=1,2} \psi_{\alpha'}\left(\frac{\Delta E}{t}\right) \frac{((\Delta E)^2/\hbar^2\omega_3^2)(1 + \epsilon) - 1}{2((\Delta E)^2/\hbar^2\omega_3^2)(1 + \epsilon) - (1 + \epsilon)}. \quad (23)$$

Here

$$\begin{aligned} \psi_1(\omega) &\neq 0 \quad \text{for} \quad 0 \leq \omega \leq \omega_1, \\ \psi_2(\omega) &\neq 0 \quad \text{for} \quad \omega_2 \leq \omega \leq \omega_3, \end{aligned}$$

where $\omega_{1,2} = \sqrt{2(\gamma_1 + 2\gamma_2)/M_{1,2}}$.

The computation of $\psi_{\alpha'}(\omega)$ for different values of ϵ and $\xi = \gamma_2/\gamma_1$ can be done in the same way as in [5,6].

The figure shows curves of $\psi_{\alpha'}(\omega)$ for both branches for different values of ϵ ($\xi = 1$), and also the right hand side of (23). One sees that for $\epsilon < 0.5$ the incoherent scattering cross section already hardly affects the optical branch, while for $\epsilon > 2$ it has no effect on the acoustic branch.

5. COHERENT SCATTERING

1. Let us now consider the question of the information which can be gotten if one knows the

differential cross section for coherent scattering. From the general expression (11) it follows that for an arbitrary crystal the coherent scattering peaks correspond to values of ΔE and κ for which the relation

$$\Delta E = \hbar\omega(\kappa, \alpha)$$

holds. Since in general ΔE and κ are freely variable, it is easy to see that in principle one can from the coherent scattering reconstruct the dispersion law for all of phase space and for all branches of the phonon spectrum. Thus the second type of information (see the introduction) can be obtained for an entirely arbitrary single crystal.

2. It can be shown in general that the matrix element for the coherent scattering on phonons of any particles describable by plane waves is given by an expression identical with (5) and with A_{1j} dependent on κ . If single-phonon processes predominate in the inelastic coherent scattering, we find for the corresponding cross section an expression of the type of (11) with $\bar{A}_j(\kappa)$. Consequently, for describing such a scattering process one must know bilinear combinations of the form $D_j(\kappa, \alpha) D_j^*(\kappa, \alpha)$, where

$$D_j(\kappa, \alpha) = (M_j)^{-1/2} \exp\{-W_j/2\} \exp\{i\kappa\rho_j\} \mathbf{q}\mathbf{v}_j(\kappa, \alpha). \quad (24)$$

Suppose that cross section measurements of coherent neutron scattering are made each time for fixed κ as a function of ΔE . Then the cross section peaks will occur at $\Delta E = \hbar\omega(\kappa, \alpha)$ for all branches of the spectrum. If we determine the area under each peak and divide it by $\bar{n}(\hbar\omega(\kappa, \alpha))/\omega(\kappa, \alpha)$ (where we assume, to be specific, that $\Delta E < 0$), then to within a constant factor the corresponding quantity will be

$$\sum_{j, j'} \bar{A}_j \bar{A}_{j'} D_j(\kappa, \alpha) D_{j'}^*(\kappa, \alpha). \quad (25)$$

We sum this expression over all α . Then, remembering the second relation in (4), we find

$$\sum_{\alpha} \sum_{j, j'} \bar{A}_j \bar{A}_{j'} D_j(\kappa, \alpha) D_{j'}^*(\kappa, \alpha) = \sum_j A_j^2 M_j^{-1} e^{-W_j}. \quad (26)$$

Thus the normalization (26) holds for the coherent scattering peaks corresponding to a fixed κ . Again assuming $W_j(\kappa) \ll 1$, we can once more compute W_j on the basis of any simple model, and consequently regard the right hand side of (26) as a known quantity. Then one must consider the question of the absolute value of the cross section.

Let us consider some individual cases.

a) There is one atom in the unit cell. In this case the relative values of the cross sections for the three branches immediately determine $D_j(\kappa, \alpha)$.

b) There are several identical atoms in the unit cell. From measurements of each branch, one determines the quantity

$$\left| \sum_j D_j(\kappa, \alpha) \right|^2.$$

But this is precisely the quantity which appears in the cross section for the one-phonon coherent scattering in the general case.

c) There are s types of atoms in the unit cell. The relative value of the cross section for each branch can be written in this case in the form [cf. (25)]

$$\sum_{\xi, \xi'} F_{\xi\xi'} \bar{A}_\xi \bar{A}_{\xi'},$$

where

$$F_{\xi\xi'} = \frac{1}{2} \sum_{j(\xi)} \sum_{j'(\xi')} (D_j D_{j'}^* + D_{j'}^* D_j). \quad (27)$$

To determine the tensor $F_{\xi\xi'}$, it is sufficient to make measurements at fixed κ on $s(s+1)/2$ samples of different isotopic compositions. For example, in the case of two types of atoms we need to have three isotopic compositions. For larger numbers of types, the value $s(s+1)/2$ may in general be lowered, especially when each atom in the lattice is a center of symmetry, so that \mathbf{v}_j is a real vector. We shall not consider this question in detail.

Thus, by having several isotopic compositions one can determine the tensor $F_{\xi\xi'}(\kappa, \alpha)$ in (27) for all values of κ and α , which together with the dispersion law completely solves the problem of obtaining information of the third type.

We note that for a whole variety of problems, in addition to values of $\kappa = \mathbf{f}$ lying in the first cell of the reciprocal lattice, one needs to know values of κ in the neighboring zone ($\kappa = \mathbf{f} + 2\pi\mathbf{b}$). This complicates the problem but gives rise to no difficulties in principle.

3. Let us now turn to the analysis of the fourth type of information, i.e., the question of the possibility of determining polarization vectors for individual phonons. We shall discuss in detail the case of a monatomic lattice.

Suppose that one knows the peaks in the coherent scattering cross section corresponding, for fixed κ , to the three frequencies $\omega(\kappa, \alpha)$ for the three branches of the spectrum. It is not difficult to show that in this case the normalization condition (26) reduces to the simple relation

$$\sum_{\alpha} |\mathbf{q}\mathbf{v}(\mathbf{f}, \alpha)|^2 = 1. \quad (28)$$

Thus from experiments one can directly deter-

mine the value of $(\mathbf{q} \cdot \mathbf{v}(\boldsymbol{\kappa}, \alpha))^2$ for each branch without making use of absolute cross section measurements.

Since $(\mathbf{v}(\boldsymbol{\kappa}, \alpha))^2 = 1$, the measurements actually determine a cone on whose surface the unit polarization vector lies. Let us now consider the vector $\boldsymbol{\kappa}_1$, which differs from $\boldsymbol{\kappa}$ by a vector of the reciprocal lattice: $\boldsymbol{\kappa}_1 = \boldsymbol{\kappa} + 2\pi\mathbf{b}_1$. Then the coherent scattering gives rise to the same three phonons, since

$$\omega(\boldsymbol{\kappa} + 2\pi\mathbf{b}_1, \alpha) = \omega(\boldsymbol{\kappa}, \alpha), \quad \mathbf{v}(\boldsymbol{\kappa} + 2\pi\mathbf{b}_1, \alpha) = \mathbf{v}(\boldsymbol{\kappa}, \alpha),$$

but with different values of the projections of the polarization vectors $(\mathbf{q}_1 \cdot \mathbf{v}(\boldsymbol{\kappa}, \alpha))^2$. We then get a new conical surface for each unit vector; its intersection with the surface of the cone around \mathbf{q} determines the direction of the polarization vector (more precisely the line along which the polarization vector is directed). However the use of two directions \mathbf{q} and \mathbf{q}_1 leaves an ambiguity in the solution—one cannot distinguish the two systems of polarization vectors which transform into one another under reflection in the plane formed by the vectors \mathbf{q} and \mathbf{q}_1 . This ambiguity is removed if one makes a third measurement for fixed $\boldsymbol{\kappa}_2 = \boldsymbol{\kappa} + 2\pi\mathbf{b}_2$ not lying in this plane.

To determine the polarization vectors in the case of a monatomic lattice it is thus necessary and sufficient to make measurements each time of the coherent scattering cross section for three noncoplanar values of $\boldsymbol{\kappa}$, which differ from one another by reciprocal lattice vectors.

Now let us consider briefly crystals with an arbitrary lattice. If there are s atoms in the unit cell, to solve the general problem one must determine s polarization vectors (for each $\boldsymbol{\kappa}$ and α), which are complex, in general. These vectors are subject to the single condition

$$\sum_j |\mathbf{v}_j(\boldsymbol{\kappa}, \alpha)|^2 = 1.$$

Consequently, in the general case one must determine $6s - 1$ independent quantities. If each atom in the lattice is a center of symmetry, the number of independent quantities reduces to $3s - 1$.

By measuring the coherent scattering cross section for a fixed value of $\boldsymbol{\kappa}$, we determine the value of (25) for each branch. If the coefficient of $(\mathbf{q} \cdot \mathbf{v}_j)$ in (24) were independent of $\boldsymbol{\kappa}$, one could not determine \mathbf{v}_j if there were identical atoms in the unit cell, while for different atoms one would

have to use a set of single crystals with different isotopic compositions. However the presence of the factor $\exp\{i\boldsymbol{\kappa} \cdot \boldsymbol{\rho}_j\}$ simplifies the problem somewhat.

Let us consider a lattice with two different atoms in the unit cell, these atoms also being centers of symmetry. Then for each normal mode one must find two real polarization vectors. Let us take two isotopic compositions and, for each single crystal, make measurements for three noncoplanar values $\boldsymbol{\kappa}_i = \boldsymbol{\kappa} + 2\pi\mathbf{b}_i$. We then get six equations for determining the five independent quantities. We note that one can also try to get the six equations for \mathbf{v}_1 and \mathbf{v}_2 for a single isotopic composition, taking into account the factor $\exp\{i\boldsymbol{\kappa} \cdot \boldsymbol{\rho}_j\}$ and using measurements for six values of $\boldsymbol{\kappa}_i = \boldsymbol{\kappa} + 2\pi\mathbf{b}_i$.

For $s > 2$, the problem of reconstructing the polarization vectors becomes very difficult in practice, because of the unavoidable accumulation of errors in using a large number of equations based on experimental data. Some special difficulties also occur in the case of $s = 2$ if one of the atoms is not a center of inversion. In this case to determine the complex polarization vectors requires the determination of eleven (!) independent quantities.

Thus one can, in principle, state the conditions of an experiment which will enable one to determine the polarization vectors and the dispersion law for an arbitrary crystal. However it seems that such a problem can be solved practically only for monatomic lattices or for diatomic lattices for which each site is a center of inversion.

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