DETERMINATION OF THE FREQUENCY SPECTRUM OF PHONONS IN CRYSTALS

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For crystals of arbitrary symmetry, we consider the problem of determining the phonon frequency spectrum, in connection with the investigation of one-phonon transitions in the Mössbauer effect and incoherent scattering of "cold" neutrons.

INTRODUCTION

A knowledge of the distribution function $\psi(\omega)$ for the phonon frequency spectrum is of great importance for the study of the properties of solids. Since the work of Placzek and Van Hove,[1] it is known that incoherent one-phonon scattering of neutrons enables us to establish the frequency spectrum in the case of cubic crystals with a single atom in the unit cell. Recently Visscher[2] pointed out that analogous information concerning the phonon spectrum of an isotropic body can be gotten by studying the resonance absorption of γ rays in a crystal (Mössbauer effect) with emission of one phonon (where the energy change is compensated by the Doppler effect from an appropriate motion of the source). Using the results of a paper of Lamb,[3] Visscher gave a detailed analysis, within the framework of the isotropic approximation, of the conditions under which one can determine $\psi(\omega)$ experimentally.

For both methods, a very important question is the possibility of determining the frequency distribution function for crystals of arbitrary symmetry with any number of atoms in the unit cell. The present paper gives a treatment of this problem. As applied to "cold" neutrons, this question was first treated by Oskot-skiĭ.[4]

The two processes, which at first glance appear to be different in character, permit of analogous descriptions for one- and multiphonon transitions. This is related to the fact that in the resonance absorption we will be interested only in those transitions for which $\hbar\omega \gg \Gamma$ (where $\hbar\omega$ is the energy of the phonon and Γ is the width of the resonance level).

1. THE MÖSSBAUER EFFECT

Suppose a nucleus located at a lattice site makes a transition from an excited state A* to the state A, with emission of a γ quantum. If we take into account a possible change in the phonon distribution, we can write the matrix element for such a transition in the form

$$M_\beta = \langle \mathbf{k},\ A,\ \{n'\}\,|\,H_1\,|\,A^*,\ \{n\}\rangle = M_{\text{nuc}}\,\langle\{n'\}\,|\exp\{i\mathbf{k}\mathbf{R}_{m\beta}\}\,|\,\{n\}\rangle. \quad (1)$$

Here $\{n\}$, $\{n'\}$ are the sets of phonon occupation numbers before and after decay of the nucleus, M_{nuc} is the matrix element corresponding to the decay of a free nucleus, $\mathbf{k}$ is the wave vector of the γ quantum, $\mathbf{R}_{m\beta}$ is the coordinate of the nucleus labelled β in the m-th unit cell, at the moment of decay.

The expression for $\mathbf{R}_{m\beta}$ has the form

$$\mathbf{R}_{m\beta} = \mathbf{R}^0_{m\beta} + \mathbf{u}_{m\beta}, \qquad \mathbf{R}^0_{m\beta} = \mathbf{r}_m + \rho_\beta, \quad (2)$$

where $\mathbf{r}_m$ and ρ_β denote, respectively, the equilibrium position of a selected site which characterizes the elementary cell, and the position of the β-th atom relative to this site.

The displacement of an arbitrary atom is conveniently represented in the form

$$\mathbf{u}_{m\beta} = \sum_{\mathbf{f},\alpha} (\hbar / 2\mu_\beta \omega_\alpha(\mathbf{f}) N)^{1/2} [\mathbf{V}_{\beta\alpha}(\mathbf{f}) \exp\{i\mathbf{f}\mathbf{r}_m\} a_\alpha(\mathbf{f}) + \mathbf{V}^*_{\beta\alpha}(\mathbf{f}) \exp\{-i\mathbf{f}\mathbf{r}_m\} a^+_\alpha(\mathbf{f})], \quad (3)$$

where $\mathbf{f}$ and ω_α are the wave vector and frequency of the phonon, α is the number of the branch, a and a^+ are the operators for annihilation and creation of the phonon, N is the number of elementary cells in the crystal, and μ_β is the mass of the β-th atom. The complex $\mathbf{V}_{\beta\alpha}$ are orthonormal:

$$\sum_\beta \mathbf{V}_{\beta\alpha}(\mathbf{f})\,\mathbf{V}^*_{\beta\alpha'}(\mathbf{f}) = \delta_{\alpha\alpha'}. \quad (4)$$

Without loss of generality, we set $\mathbf{r}_m = 0$. In accordance with the usual procedure, considering (3), we represent the exponential in (1) as a product of exponentials corresponding to the individual normal vibrations. We expand these exponentials

in series, keeping the first three terms (the remaining terms give a contribution which tends to zero when $N \to \infty$). From (3) it follows that only the second term gives a transition which is not diagonal in the occupation numbers.

We average the square modulus of the matrix element (1) over the initial equilibrium distribution, taking account of the independence of the individual oscillators. After transformation, we find for a process in which s phonons participate,

$$|M_\beta^s|^2 = |M_{\text{nuc}}|^2 \left(\frac{R_\beta}{N}\right)^s \exp\left\{-\frac{R_\beta}{N}\sum_{\alpha,\mathbf{f}}{}' \frac{|\mathbf{q}V_{\beta\alpha}(\mathbf{f})|^2}{\hbar\omega_\alpha(\mathbf{f})}[2\bar{n}_\alpha(\mathbf{f})+1]\right\}$$

$$\times \prod_{i=1}^{s} |\mathbf{q}V_{\beta\alpha_i}(\mathbf{f}_i)|^2 [\hbar\omega_{\alpha_i}(\mathbf{f}_i)]^{-1}\left[\bar{n}_{\alpha_i}(\mathbf{f}_i)+\tfrac{1}{2}\pm\tfrac{1}{2}\right]. \quad (5)$$

Here R_β denotes the recoil energy for an isolated nucleus,

$$R_\beta = \hbar^2 k^2 / 2\mu_\beta, \quad (6)$$

and $\mathbf{q}$ is a unit vector in the direction of $\mathbf{k}$; $\bar{n}$ is the equilibrium value corresponding to the crystal temperature. The upper sign inside the brackets in the product in (5) corresponds to phonon emission and the lower to phonon absorption. The prime on the sum in the exponential in (5) means that the s terms with $(\alpha, \mathbf{f}) = (\alpha_i, \mathbf{f}_i)$ have been omitted. For finite s, the contribution of these terms to the sum is negligible, and from now on we shall omit the prime.

Now let us determine the probability of emission of a γ quantum in the direction $\mathbf{q}$ with energy $E\gamma$ for an s-phonon process, where we take the probability per unit energy range and per unit solid angle.

Using the standard formulas of perturbation theory and changing from summation to an integration over phase space, we find (cf. (5)):

$$W_\beta^s(E_\gamma, \mathbf{q}) = W_0 e^{-Z_\beta}(R_\beta)^s \frac{(v_0)^s}{(2\pi)^{3s}}\frac{1}{s!} S \sum_{\alpha_1\ldots\alpha_s}\int d^3f_1\ldots d^3f_s$$

$$\times \prod_{i=1}^{s} |\mathbf{q}V_{\beta\alpha_i}(\mathbf{f}_i)|^2 [\hbar\omega_{\alpha_i}(\mathbf{f}_i)]^{-1}(\bar{n}_{\alpha_i}+\tfrac{1}{2}\pm\tfrac{1}{2})\,\delta$$

$$\times(E_\gamma - E_0 \pm \hbar\omega_{\alpha_1}\pm\ldots\pm\hbar\omega_{\alpha_s}) \equiv W_0 F_\beta^s(E_\gamma, \mathbf{q}). \quad (7)$$

Here E_0 is the energy of the excited state, W_0 is the probability of γ decay for the free nucleus, v_0 is the volume of the elementary cell, S is a sum over the s + 1 different combinations of absorbed and emitted phonons, and

$$Z_\beta = R_\beta \frac{v_0}{(2\pi)^3}\sum_\alpha \int d^3f \frac{|\mathbf{q}V_{\beta\alpha}(\mathbf{f})|^2}{\hbar\omega_\alpha(\mathbf{f})}(2\bar{n}_\alpha(\mathbf{f})+1) \quad (8)$$

(The energy transfer to the lattice is small compared with the energy of the γ quantum.)

It is not difficult to show that the relation

$$\sum_{s=0}\int F_\beta^s(E_\gamma, \mathbf{q})\,dE_\gamma = 1 \quad (9)$$

holds for the F_β^s.

For resonance absorption of a γ quantum $(E_\gamma, \mathbf{q})$, the probability of an s'-phonon process is given by the same expression (7), except that E_0 and E_γ interchange places in the argument of the δ function. If the source moves with velocity $\mathbf{u}$ and the target is sufficiently thin, the cross section for resonance absorption accompanied by an s-phonon process in the source and by an s'-phonon process in the target is (compare the analogous expression in reference 2):

$$\sigma^{ss'}(\Delta E, \mathbf{q}) = Q\frac{1}{gg'}\sum_{\beta,\lambda}\int F_\beta^s(E_\gamma, \mathbf{q})\,F_\lambda'^{s'}(E_\gamma+\Delta E, \mathbf{q})\,dE_\gamma. \quad (10)$$

In this expression Q is the cross section for resonance absorption by a free nucleus, integrated over energy; g and g' are the numbers of identical nuclei, having the resonance level, in the elementary cells of source and target [the summation in (10) extends only over these nuclei]. From now on, primed quantities refer to the target.

For the Doppler shift ΔE we have the relation $\Delta E = E_\gamma \mathbf{u}\cdot\mathbf{q}/c$.

The expressions we have found enable us to analyze completely the general case. Suppose that one-phonon processes play the prime role in the range $\Gamma \ll \Delta E < \hbar\omega_{\max}$ (however, see later). Then in this interval the resonance absorption cross section reduces to $(\Delta E > 0)$

$$\sigma^1(\Delta E, \mathbf{q}) \equiv \sigma^{01}(\Delta E, \mathbf{q}) + \sigma^{10}(\Delta E, \mathbf{q}) = Q\frac{v_0}{(2\pi)^3}R\frac{1}{gg'}q^i q^k$$

$$\times\sum_{\beta,\lambda}\exp\{-Z_\beta - Z_\lambda'\}\left\{\sum_\alpha\int d^3f \frac{V_{\beta\alpha}^i(\mathbf{f})V_{\beta\alpha}^{k*}(\mathbf{f})}{\hbar\omega_\alpha(\mathbf{f})}(\bar{n}_\alpha(\mathbf{f})+1)\,\delta\right.$$

$$\times(\Delta E - \hbar\omega_\alpha) + \sum_\xi\int d^3f \frac{V_{\lambda\xi}'^i(\mathbf{f})V_{\lambda\xi}'^{k*}(\mathbf{f})}{\hbar\omega_\xi'(\mathbf{f})}(\bar{n}_\xi'(\mathbf{f})+1)\,\delta$$

$$\left.\times(\Delta E - \hbar\omega_\xi')\right\} \quad (11)$$

(where we sum over repeated Latin superscripts).

For the further analysis it is useful to determine the cross section for the true Mössbauer effect, $\sigma^{00}(0, \mathbf{q})$. Expressions (10) and (7) cannot be used for this directly, since (10) contains a product of two δ functions. This result is a consequence of neglecting the width Γ of the excited state. Even when this is valid for transitions involving phonons $(\hbar\omega \gg \Gamma)$, in computing σ^{00} we must consider the fact that Γ is finite. We introduce in place of $\delta(E_\gamma = E_0)$ an expression of the form

$$\frac{1}{\pi}\frac{\Gamma/2}{(E_\gamma - E_0)^2 + \Gamma^2/4}.$$

We then find for σ^{00},

$$\sigma^{00}(0,\mathbf{q}) \equiv \sigma^0(\mathbf{q}) = Q\,\frac{1}{\pi\Gamma}\,\frac{1}{gg'}\sum_{\beta,\lambda}\exp\{-Z_\beta - Z'_\lambda\}. \quad (12)$$

We note that σ^0, like σ', can in general have a sizeable anisotropy. This will especially manifest itself in strongly anisotropic structures, for example in layer lattices. In such cases there are, a priori, directions of the wave vector of the γ quantum for which Z will be especially small.

Let us consider a single crystal of an arbitrary crystal system, having one atom per unit cell. Each of the terms in the curly brackets in (11) is a symmetric tensor of second rank, having the symmetry of the crystal. When it is brought to principal axes, such a tensor has in general three independent components. We determine the ratio

$$\sigma^1(\Delta E, \mathbf{q})/\sigma^0(\mathbf{q})$$

for three mutually perpendicular directions of the vector $\mathbf{q}$ relative to a fixed orientation of source and target, and sum them. If we make use of the condition $\mathbf{V}_\alpha \cdot \mathbf{V}^*_\alpha = 1$, it is easy to see that the result will not contain the polarization vectors. In the integrands in (11) we are then left with only a dependence on the phonon energy. Separating explicitly the integration over ω_α, and introducing the frequency distribution $\psi(\omega)$ for the phonon spectrum normalized in accordance with the relation

$$\tfrac{1}{3}v_0(2\pi)^{-3}\sum_\alpha \int_{\Delta\omega_\alpha = d\omega} d^3f = \psi(\omega)\,d\omega, \quad (13)$$

we get

$$\tfrac{1}{3}\sum_l^3 \sigma^1(\Delta E,\mathbf{q}_l)/\sigma^0(\mathbf{q}_l) = \frac{\pi\Gamma}{\hbar}\,\frac{R}{\Delta E}\,\psi\left(\frac{\Delta E}{\hbar}\right) e^{\Delta E/\varkappa T}\,[e^{\Delta E/\varkappa T} - 1]^{-1}$$
$$+ \psi'\left(\frac{\Delta E}{\hbar}\right) e^{\Delta E/\varkappa T'}\,[e^{\Delta E/\varkappa T'} - 1]^{-1}. \quad (14)$$

If the lattices of source and target are the same and T = T′, then (14) uniquely determines $\psi(\Delta E/\hbar)$. Thus measurement of the cross sections $\sigma^1(\Delta E, \mathbf{q})$ and $\sigma^0(\mathbf{q})$ for three directions enables us to find the frequency distribution for a crystal of abritrary symmetry.

In the case of uniaxial crystals, the symmetric tensor of second rank has two independent components and it is consequently sufficient to make two measurements, first directing the vector $\mathbf{q}$ along the symmetry axis of the crystal and then along any direction in the plane perpendicular to this axis.

In a cubic crystal, the second rank tensor degenerates into the unit tensor, so that one measurement in an arbitrary direction suffices. The isotropic case, treated by Visscher,[2] is actually equivalent to the case of a cubic crystal.

The presence of the temperature dependence in both terms in (14) opens the attractive possibility (for T ≠ T′) of separately determining ψ and ψ'. [We note that here it may be useful to reverse the direction of motion of the source ($\Delta E < 0$), which results in the replacement of $\bar{n}$ by $\bar{n} + 1$ in (14).] The realization of this possibility would enable us, on the one hand, to determine the spectrum for the undistorted lattice of the target and, on the other hand, to obtain valuable information concerning changes in the frequency spectrum resulting from defects in the source lattice, in particular concerning the nature of the vibrations of an atom in an interstitial position.

We note that this separation will involve some experimental difficulties, since the necessary increase in temperature of one of the crystals results in a reduction of σ^0 and σ^1. This imposes an obvious limitation on the value of $(Z + Z')_{T=0}$. [See (11) and (12).]

Now we consider crystals having an arbitrary number of atoms in the unit cell. In this case we make the assumption that the factors $\exp\{-Z_\beta - Z'_\lambda\}$, which depend on the integral properties of the spectrum, differ only slightly for any pair β, λ. This will be valid in any case for Z_β, $Z'_\lambda \ll 1$.

Again we determine the sum of the ratios σ^1/σ^0 for three mutually perpendicular directions:

$$\tfrac{1}{3}\sum_l^3 \sigma^1(\Delta E,\mathbf{q}_l)/\sigma^0(\mathbf{q}_l) = \tfrac{1}{3}\pi\Gamma R v_0 (2\pi)^{-3}$$
$$\times\left\{\frac{1}{g}\sum_{\alpha,\beta}\int d^3f V_{\beta\alpha}(\mathbf{f})\,V^*_{\beta\alpha}(\mathbf{f})\,[\hbar\omega_\alpha(f)]^{-1}(\bar{n}_\alpha + 1)\,\delta(\Delta E - \hbar\omega_\alpha)\right.$$
$$\left.+\frac{1}{g'}\sum_{\xi,\lambda}\int d^3f V_{\lambda\xi}(\mathbf{f})\,V'^*_{\lambda\xi}(\mathbf{f})\,[\hbar\omega'_\xi(f)]^{-1}(\bar{n}'_\xi + 1)\,\delta(\Delta E - \hbar\omega'_\xi)\right\}. \quad (15)$$

If we use condition (4) and relation (13) (where the left side of (13) should now contain an additional factor 1/g), we again arrive at (14). Thus in this case also, when the assumptions made above are valid, it becomes possible to determine the frequency distribution function for a crystal of arbitrary symmetry.

In the general case, when there are atoms of different sorts in the unit cell, $\psi(\omega)$ cannot be determined from the differential cross section of the one-phonon process. This is easily seen by analyzing the general expression (11).

A few words about multiphonon processes. We know that as a rule the frequency distribution function will have at least two maxima. If these maxima are sufficiently sharp, and if twice the smaller critical frequency is greater than the maximum value ω_c, then in the range of values of ΔE from 0 to $\hbar\omega_{max}$ the role of two-phonon and, a fortiori, of multiphonon processes will be markedly reduced.

However, even when the multiphonon processes make a significant contribution to the total cross section $\sigma(\Delta E, \mathbf{q})$, if we use the isotropic approximation for $s + s' \geq 2$ we can consistently include all transitions involving more than one phonon. In fact, for sufficiently low values of $\Delta E (\ll \hbar\omega_{max})$ the contribution of the multiphonon processes is negligible ($\psi(\omega \to 0) \to 0$) and consequently they do not disturb the determination of the frequency distribution in this region. Moreover, for sufficiently low frequencies, $\psi(\omega)$ can be computed directly if we know the elastic constants of the crystal. For an arbitrary ΔE, in multiphonon processes there are created practically no phonons with energies in a sufficiently narrow interval around $\hbar\omega = \Delta E$ (again because $\psi(\omega \to 0) \to 0$). Because of this, the quantities σ^{ss} (for $s + s' \geq 2$) can be expressed in terms of the function $\psi(\omega)$ as determined for the region $\omega < \Delta E/\hbar$. From this it is clear that the knowledge of $\psi(\omega)$ for the initial part of the frequency range enables us, once we know the dependence of $\sigma(\Delta E, \mathbf{q})$, to find $\sigma^1(\Delta E, \mathbf{q})$ and with it $\psi(\omega)$ for the whole range of frequencies of the phonon spectrum.

2. "COLD" NEUTRONS

The results presented in the preceding section can be carried over without difficulty to the case of incoherent scattering of "cold" neutrons. The square of the matrix element describing the inelastic scattering of a neutron will have the form (5) with the appropriate value of M_{nuc}, and with $\mathbf{k} = \mathbf{k}_1 - \mathbf{k}_0$, where $\mathbf{k}_0$ and $\mathbf{k}_1$ are the wave vectors of the neutron before and after scattering.

Let us consider the case where the crystal consists of identical atoms. The differential scattering cross section is expressed (for fixed $\mathbf{k}_0$) as

$$d\sigma(\mathbf{k}_1)/d\mathbf{k}_1 = \sum_s d\sigma^s(\mathbf{k}_1)/d\mathbf{k}_1,$$

$$d\sigma^s(\mathbf{k}_1)/d\mathbf{k}_1 = \sigma_\infty(\hbar^2/4\pi k_0 mg)\sum_\beta F^s_\beta(E_1, \mathbf{q}), \qquad (16)$$

where $F^s_\beta(E_1, \mathbf{q})$ is defined in accordance with (7), with E_γ and E_0 replaced by the final (E_1) and the initial (E_0) energy of the neutron. (We mention that Eq. (9) will no longer be satisfied, since the quantity R_β cannot be treated as constant for varying E_1.) In (16) we use the following notation: σ_∞ is the total incoherent scattering cross section for a rigidly bound nucleus, and m is the mass of the neutron.

Let us consider a single crystal with arbitrary symmetry. Considering the case of a neutron with energy E_0 less than the Debye energy, and assuming that $T_{cryst} \lesssim \Theta_D$ and $\mu \gg m$, we find for Z_β a value much less than unity. Because of this we will disregard any possible weak dependence of the Debye-Waller factor on β. We then have for the one-phonon process,

$$d\sigma^1(\mathbf{k}_1)/d\mathbf{k}_1 = \sigma^0(\mathbf{k})(\hbar^2 R/mk_0 g)\, q^i q^h v_0 (2\pi)^{-3}$$
$$\times \sum_{\alpha,\beta}\int d^3f\, v^i_{\beta\alpha}(\mathbf{f})\, v^{k*}_{\beta\alpha}(\mathbf{f})\,[\hbar\omega_\alpha(\mathbf{f})]^{-1}[(\bar{n}_\alpha + 1)\,\delta(E_0 - E_1 - \hbar\omega_\alpha)$$
$$+ \bar{n}_\alpha \delta(E_0 - E_1 + \hbar\omega_\alpha)], \qquad (17)$$

where $\sigma^0(\mathbf{k})$ is the differential cross section for elastic scattering per unit solid angle.

In the case of the Mössbauer effect there was a simplifying circumstance that was equivalent to the absence of any difference between the directions of the vectors $\mathbf{q}$ and $\mathbf{k}_1$. After fixing the scattering direction in the neutron case (as is done in the experiment), to find the function $\psi(\omega)$ for an arbitrary crystal we must proceed somewhat differently than in the preceding section. Namely, we must determine $d\sigma^1(\mathbf{k}_1)/d\mathbf{k}_1$ and $\sigma^0(\mathbf{k})$ for three positions of the single crystal which differ from one another by a cyclic permutation of the coordinates.

Let us represent (17) in the form

$$(1/\sigma^0(\mathbf{k}))\, d\sigma^1(\mathbf{k}_1)/d\mathbf{k}_1 = q^i q^k T^{ik}. \qquad (18)$$

We denote the direction of the principal axes in the first position of the crystal by x, y, z. Then the right side of (18) gives

$$T^{xx}(q^x)^2 + T^{yy}(q^y)^2 + T^{zz}(q^z)^2.$$

For the other two positions of the crystal we have, respectively,

$$T^{zz}(q^x)^2 + T^{xx}(q^y)^2 + T^{yy}(q^z)^2,$$

$$T^{yy}(q^x)^2 + T^{zz}(q^y)^2 + T^{xx}(q^z)^2.$$

Summing the three equations, we find $q^2 T^{ii} = T^{ii}$.

Comparing (18) with (17) and using (4) and (13), we arrive at the relation

$$\frac{1}{3}\sum_l^3 \frac{1}{\sigma^0(\mathbf{k})}\frac{d\sigma^1(\mathbf{k}_1)}{d\mathbf{k}_1}\Big|_l = \frac{\hbar^3}{2mk_0\mu}\frac{k^2}{\Delta E}\psi\left(\frac{\Delta E}{\hbar}\right)[e^{\Delta E/\varkappa T} - 1]^{-1}, \qquad (19)$$

where

$$\Delta E = E_1 - E_0 > 0.$$

Thus, by using three positions of the crystal and determining the cross section for one-phonon incoherent scattering $d\sigma^1(\mathbf{k}_1)/d\mathbf{k}_1$ as a function of E_1, and the elastic scattering cross section $\sigma^0(\mathbf{k})$ for the corresponding values of $\mathbf{k}$, we can find the phonon spectrum of a crystal of arbitrary symmetry having any number of identical atoms in the unit cell.

In the case of uniaxial crystals, two positions of the crystal are sufficient, but it is necessary to know exactly the direction of the symmetry axis. In a cubic crystal a single crystal position is sufficient, and we arrive at the result of Placzek and Van Hove.[1]

If $Z_\beta \ll 1$, which is often the case when working with "cold" neutrons, it is obviously sufficient to measure only $d\sigma^1(\mathbf{k}_1)/d\mathbf{k}_1$.

The arguments concerning the inclusion of multiphonon processes which were presented at the end of the previous section remain valid also for the present case.

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