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Fig. 1

Figure 1: Fig. 1

Abstract

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PHYSICS

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THE INFLUENCE OF FORCED OSCILLATIONS OF A CRYSTAL ON THE SPECTRUM OF SCATTERED γ-QUANTA

(Presented by Academician N. N. Bogolyubov, 19 II 1962)

It is known that from the spectrum of scattered γ-quanta or neutrons one can reconstruct the dispersion law for the frequencies of normal lattice vibrations ([1–3]). The difficulty in measuring the spectrum of scattered γ-quanta is that sources of monochromatic γ-quanta (Mössbauer sources) are not sufficiently intense to make it possible to accumulate good statistics in determining a one-phonon peak in the spectrum of scattered γ-quanta. On the other hand, it is known that in experiments on resonant absorption, the oscillation of the source as a whole, produced by ultrasound, leads to the appearance in the emission spectrum of additional peaks whose intensities are comparable to, or even greater than, the intensity of the Mössbauer line ([4]). These peaks are located at distances $n\omega_q$ ($n = \pm1, \pm2, \dots$; ω_q is the ultrasound frequency) from the main Mössbauer line. The same will occur if oscillations are excited in the emitter in such a way that its center of gravity does not move. Such experiments make it possible to measure the frequencies ω_q of normal vibrations, but not their wave vector. To measure the wave vector $\mathbf{q}$ of a vibration, and thereby to determine the section of the isoenergetic surface $\omega(\mathbf{q})$, it is necessary to use data from experiments on resonant or Rayleigh scattering in crystals in which forced oscillations are excited, for example, by means of a quartz ultrasound generator ([5]). In this case, as will be shown below, a considerable enhancement of the one-phonon peaks in scattering can be achieved.

Fig. 1

Let us consider, for definiteness, Rayleigh scattering. From the point of view of perturbation theory with respect to the electromagnetic interaction, Rayleigh scattering is a second-order process. It is obvious that when the energy of the γ-quantum does not coincide with the excitation energy of any of the atomic or nuclear levels, the greatest contribution to scattering is made by those virtual states of the atom and nucleus whose excitation energy is close to the energy of the incident γ-quanta. The system crystal + electromagnetic field is described by the Hamiltonian

$$\mathbf{H} = \mathbf{H}_0 + \mathbf{H}_a + \mathbf{H}_e + \mathbf{H}_1 + \mathbf{H}_2, \tag{1}$$

where $\mathbf{H}_0$ is the Hamiltonian describing lattice vibrations in the harmonic approximation (it is expressed in terms of the displacements of nuclei from their equilibrium positions $\mathbf{u}_{lj}$, where l is the number of the unit cell, j is the number of the atom in the cell ([6])); $\mathbf{H}_a$ is the Hamiltonian of the internal state of the atom and nucleus; $\mathbf{H}_e$ is the Hamiltonian of the free electromagnetic field; $\mathbf{H}_1 = \sum_{lj} \mathbf{F}_{lj}(t)\mathbf{u}_{lj}$ is the Hamiltonian describing the interaction of the crystal with an external source ($\mathbf{F}_{lj}(t)$ is the force acting on the lj-th atom); $\mathbf{H}_2 = \sum_{lj\lambda\sigma} L^{\mathbf{k}\lambda}_{\sigma j} e^{i\mathbf{k}_\lambda \mathbf{R}_{lj}} a_{\mathbf{k}\lambda\sigma} +$ c.c. is the Hamiltonian of the electromagnetic interaction; $a_{\mathbf{k}\lambda\sigma}$ is an ope-

operator for annihilation of a photon with wave vector $\mathbf{k}_\lambda$ and polarization σ; $L^{\mathbf{k}_\lambda}_{\sigma j}$ is that part of the electromagnetic-interaction operator which depends only on the coordinates of the electrons and nucleons; $\mathbf{R}_{lj} = \mathbf{R}^0_{lj} + \mathbf{u}_{lj}$; $\mathbf{R}^0_{lj} = \mathbf{l} + \mathbf{r}_j$ is the radius vector of the equilibrium position of an atom in the lattice; $\mathbf{r}_j$ is the radius vector of the j-th atom in the cell.

To calculate the scattering probability we shall use the S-matrix formalism. We shall assume that $\mathbf{H}_1 + \mathbf{H}_2$ is switched on adiabatically at $t = -\infty$ and switched off at $t = \infty$. Then the total scattering probability, calculated to second order in $\mathbf{H}_2$ (and to all orders in $\mathbf{H}_1$), is written in the form

$$\begin{aligned} w(\mathbf{k}_\lambda, \mathbf{k}_{\lambda'}) = \frac{N_{\mathbf{k}_\lambda}(N_{\mathbf{k}_{\lambda'}} + 1)}{\hbar^4} \sum_{\mathbf{n},\mathbf{n}',b,b'} e^{-i\mathbf{k}(\mathbf{R}^0_{\mathbf{n}'} - \mathbf{R}^0_{\mathbf{n}})} Q^{bb'}_{jj'}(\mathbf{k}_\lambda, \mathbf{k}_{\lambda'}) \int_{-\infty}^{\infty} dt \int_{-\infty}^{t} dt' \int_{-\infty}^{\infty} d\tau \int_{-\infty}^{\tau} d\tau' \\ \times \left\langle T \exp\left\{ -i \sum_{\mathbf{n}''} \int_c dz \left[\mathbf{f}_{\mathbf{n}''}(z)\mathbf{u}_{\mathbf{n}''}(z) + \frac{1}{\hbar}\mathbf{F}_{\mathbf{n}''}(z)\mathbf{u}_{\mathbf{n}''}(z) \right] \right\} \right\rangle \\ \times \exp\{-i(\omega_{ba} - \omega_\lambda)t + i(\omega_{ba} - \omega_\lambda)t' + i(\omega_{b'a} - \omega_{\lambda'})\tau - (\omega_{b'a} - \omega_\lambda)\tau'\}. \end{aligned} \tag{2}$$

Here a and b are the quantum numbers, respectively, of the ground and intermediate states of the nucleus and the atomic electrons; $\mathbf{k} = \mathbf{k}_{\lambda'} - \mathbf{k}_\lambda$; $\omega_{ba} = E_{ba}/\hbar$; E_{ba} is the excitation energy of the intermediate state; $(\omega_\lambda, \mathbf{k}_\lambda)$ and $(\omega_{\lambda'}, \mathbf{k}_{\lambda'})$ are, respectively, the frequencies and wave vectors of the incident and scattered γ-quanta; $N_{\mathbf{k}_\lambda}$ are the occupation numbers of the electromagnetic field; $\langle\cdots\rangle$ denotes statistical averaging over the vibrational states of the lattice; $\mathbf{n} = (l, j)$;

$$Q_{jj'}^{bb'}(\mathbf{k}_\lambda, \mathbf{k}_{\lambda'}) = (L_j^{\mathbf{k}_\lambda})^*_{ab}(L_j^{\mathbf{k}_{\lambda'}})^*_{ba}(L_{j'}^{\mathbf{k}_{\lambda'}})_{ab'}(L_{j'}^{\mathbf{k}_\lambda})_{b'a},$$

$$\mathbf{f}_{\mathbf{n}''}(z) = \{\mathbf{k}_\lambda\delta(z-\tau') - \mathbf{k}_{\lambda'}\delta(z-\tau)\}\delta_{\mathbf{n}''\mathbf{n}} + \{\mathbf{k}_{\lambda'}\delta(z-t) - \mathbf{k}_\lambda\delta(z-t')\}\delta_{\mathbf{n}''\mathbf{n}'},$$

T-ordering is carried out along the contour c.

The average of the type appearing in expression (2),

$$\left\langle T\exp\left\{-i\sum_{\mathbf{n}''}\int_c \mathbf{I}_{\mathbf{n}''}(z)\mathbf{u}_{\mathbf{n}''}(z)\,dz\right\}\right\rangle, \tag{3}$$

where $\mathbf{I}_{\mathbf{n}''}(z)$ is a c-function, in the harmonic approximation can be represented in the form

$$\exp\left\{-\frac{1}{2}\int_c dz\int_c dz' \sum_{\mathbf{n}''\mathbf{n}'''} I^\alpha_{\mathbf{n}''}(z)D^{\alpha\beta}_{\mathbf{n}''\mathbf{n}'''}(z,z')I^\beta_{\mathbf{n}'''}(z')\right\}, \tag{4}$$

where

$$D^{\alpha\beta}_{\mathbf{n}''\mathbf{n}'''}(z,z') = \langle Tu^\alpha_{\mathbf{n}''}(z)u^\alpha_{\mathbf{n}'''}(z')\rangle$$

is the phonon Green' s function in the harmonic approximation ($\alpha, \beta = 1, 2, 3$). Using (4), the expression for the scattering probability of γ-quanta can be represented in the form

$$\begin{aligned}
w(\mathbf{k}_\lambda, \mathbf{k}_{\lambda'}) = \frac{N_{\mathbf{k}_\lambda}(N_{\mathbf{k}_{\lambda'}}+1)}{\hbar^4} A \sum_{\mathbf{n},\mathbf{n}',b,b'} e^{-i\mathbf{k}(\mathbf{R}^0_{\mathbf{n}'}-\mathbf{R}^0_{\mathbf{n}})} Q^{bb'}_{jj'}(\mathbf{k}_\lambda,\mathbf{k}_{\lambda'}) \int_{-\infty}^{\infty} dt \int_{-\infty}^{t} dt' \\
\times \int_{-\infty}^{\infty} d\tau \int_{-\infty}^{\tau} d\tau' \exp\left\{-\frac{1}{2}\left[B_{\mathbf{nn}}(\tau,\tau') + B_{\mathbf{n}'\mathbf{n}'}(t,t')\right] - C_{\mathbf{nn}'}(\tau,\tau';t,t')\right\} \\
\times \exp\{-i\mathbf{k}_\lambda\mathbf{u}_\mathbf{n}(\tau') + i\mathbf{k}_{\lambda'}\mathbf{u}_\mathbf{n}(\tau) - i\mathbf{k}_{\lambda'}\mathbf{u}_{\mathbf{n}'}(t) + i\mathbf{k}_\lambda\mathbf{u}_{\mathbf{n}'}(t')\} \\
\times \exp\{-i(\omega_{ba}-\omega_\lambda)t + i(\omega_{ba}-\omega_\lambda)t' + i(\omega_{b'a}-\omega_{\lambda'})\tau - i(\omega_{b'a}-\omega_\lambda)\tau'\},
\end{aligned}$$

where

$$B_{\mathbf{nn}}(\tau,\tau') = k^\alpha_\lambda D^{\alpha\beta}_{\mathbf{nn}}(0,+0)k^\beta_\lambda + k^\alpha_{\lambda'}D^{\alpha\beta}_{\mathbf{nn}}(0,+0)k^\beta_{\lambda'} - 2k^\alpha_{\lambda'}D^{\alpha\beta}_{\mathbf{nn}}(\tau,\tau')k^\beta_\lambda,$$

$$C_{\mathbf{nn}'}(\tau,\tau';t,t') = k^{\alpha}_{\lambda'} D^{\alpha\beta}_{\mathbf{nn}'}(\tau',t) k^{\beta}_{\lambda'} + k^{\alpha}_{\lambda} D^{\alpha\beta}_{\mathbf{nn}'}(\tau,t') k^{\beta}_{\lambda} - k^{\alpha}_{\lambda'} D^{\alpha\beta}_{\mathbf{nn}'}(\tau,t) k^{\beta}_{\lambda'} - k^{\alpha}_{\lambda} D^{\alpha\beta}_{\mathbf{nn}'}(\tau',t') k^{\beta}_{\lambda}.$$

$$A = \exp\left\{ -\frac{1}{2\hbar^2} \int_c dz \int_c dz' \sum_{n,\mathbf{n}'} F^{\alpha}_{n}(z) D^{\alpha\beta}_{n\mathbf{n}'}(z,z') F^{\beta}_{n'}(z') \right\},$$

$$\overline{u^{\alpha}_{n}}(t) = -\frac{i}{\hbar} \sum_{n'} \int_{-\infty}^{t} \left\langle \left[u^{\alpha}_{n}(t), u^{\beta}_{n'}(\tau) \right] \right\rangle F^{\beta}_{n'}(\tau)\, d\tau$$

is the average displacement of atoms caused by the external force F^{α}_{n}; $\mathbf{n} = (lj)$.

In what follows we shall assume that $\overline{\mathbf{u}}_n(t)$ can be represented in the form $\overline{\mathbf{u}}_n(t) = \mathbf{a}\cos(\mathbf{q}\mathbf{R}^0_n - \omega_q t)$, where ω_q is the frequency of one of the normal vibrations of the scattering crystal; $\mathbf{a}$ is the amplitude of the vibration, and we shall calculate the shape of the spectrum of scattered γ-quanta in the case of small thermal broadening ([7]).

Taking into account the expression for $\overline{\mathbf{u}}_n(t)$ and using the relation

$$\exp\{i\alpha\cos\varphi\} = \sum_{m=-\infty}^{\infty} i^m J_m(\alpha) e^{im\varphi},$$

where $J_m(\alpha)$ is the cylindrical Bessel function of the m-th order with real argument, the probability of scattering per second for $|(\mathbf{k}_\lambda \cdot \mathbf{a})| < |(\mathbf{k}_{\lambda'} \cdot \mathbf{a})|$ can be represented in the form

$$W(\mathbf{k}_\lambda, \mathbf{k}_{\lambda'}) = N_{k_\lambda}(N_{k_{\lambda'}}+1) \frac{2\pi}{\hbar^2} \sum_{m=-\infty}^{\infty} \left| \sum_{l} e^{-i(\mathbf{k}-m\mathbf{q})l} \right|^2 \left| \sum_{j} C_j e^{-i(\mathbf{k}-m\mathbf{q})\mathbf{r}_j - G_j} \right|^2 \times \times J^2_m(\mathbf{k}\cdot\mathbf{a})\, \delta(\omega_\lambda - \omega_{\lambda'} - m\omega_q). \tag{5}$$

Here

$$C_j = \sum_b \frac{(L^{k_\lambda}_i)_{ab} (L^{k_{\lambda'}}_j)_{ba}}{\hbar(\omega_{ba} - \omega_\lambda)}$$

is the scattering form factor;

$$G_j = \frac{1}{2}\{ k^{\alpha}_{\lambda} D^{\alpha\beta}_{n\mathbf{n}}(0,+0) k^{\beta}_{\lambda} + k^{\alpha}_{\lambda'} D^{\alpha\beta}_{n\mathbf{n}}(0,+0) k^{\beta}_{\lambda'} \}.$$

It follows from (5) that if an energetically narrow spectrum, in which the Mössbauer line is present, is incident on the scatterer, then on observation at the Bragg angle, which is determined from the condition $\omega_\lambda - \omega_{\lambda'} = 0$, $\mathbf{k}_\lambda - \mathbf{k}_{\lambda'} = \mathbf{K}$, where ω_λ is the frequency of the Mössbauer γ-quanta and $\mathbf{K}$ is the reciprocal-lattice vector, a Mössbauer line with relative intensity $J_0^2(\mathbf{k}\cdot\mathbf{a})$ will be observed in the scattering spectrum. On observation at angles different from the Bragg angles ($\mathbf{k}_{\lambda'} - \mathbf{k}_\lambda - m\mathbf{q} = \mathbf{K}$), narrow lines with frequency $\omega_{\lambda'} = \omega_\lambda \pm m\omega_q$ will be observed in the spectrum of the scattered γ-quanta; their relative intensity, if the thermal background is neglected, will be $J_m^2(\mathbf{k}\cdot\mathbf{a})$. The ratio of the intensities of the lines of the scattered γ-quanta is determined by the quantity $2\pi|\mathbf{a}|/d$, where d is the lattice constant of the scatterer. For example, for $|\mathbf{a}| = 0.1d$ the relative intensities of the lines with $m = 0$ and $m = 1$ are 0.81 and 0.09 (so that all harmonics with $m \geqslant 2$ account for 0.01 of the number of scattered γ-quanta); for $|\mathbf{a}| = 0.13d$ the relative intensities are, respectively, 0.70 and 0.13 (the higher harmonics account for 0.04 of the total number of scattered γ-quanta).

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Note: Figure translations are in progress. See original paper for figures.