Assuming that the molecules are rigid, the width and displacement of a peak for coherent single-phonon scattering of neutrons by an anharmonic molecular crystal is determined.

As is well known, the thermal properties of a molecular crystal are mainly determined by the vibrations of the molecules as a whole. Assuming that the molecules are rigid simplifies the analysis of molecular crystals, but this analysis differs from that for atomic crystals because oriented vibrations are present. This feature has a definite effect on the attenuation of the phonons. Below we determine the width of the peak of single-phonon inelastic coherent scattering of neutrons by a molecular crystal. The molecules are assumed to be rigid.

Consider a crystal whose elementary cell contains one molecule. Its properties are defined by the Hamiltonian

$$H = \sum_{j=1}^{6} \sum_{\alpha} \hbar \omega(\mathbf{k}) (\mathbf{a}_{j,\alpha}^{\dagger} \mathbf{a}_{j,\alpha} + \frac{1}{2}) +$$

$$+ \sum_{\mathbf{k}, \mathbf{j}, \mathbf{j'}, \mathbf{j''}, \mathbf{i}, \mathbf{f}, \mathbf{f'}} V^{(3)}(\mathbf{k}, \mathbf{j}, \mathbf{j'}, \mathbf{j''}, \mathbf{i}, \mathbf{f}, \mathbf{f'}) \mathbf{A}_{\mathbf{i}, \mathbf{f}, \mathbf{f'}} \mathbf{A}_{\mathbf{j}, \mathbf{j'}, \mathbf{j''}} +$$

$$+ \sum_{\mathbf{k}, \mathbf{j}, \mathbf{j'}, \mathbf{j''}, \mathbf{i}, \mathbf{f}, \mathbf{f'}} V^{(4)}(\mathbf{k}, \mathbf{j}, \mathbf{j'}, \mathbf{j''}, \mathbf{i}, \mathbf{f}, \mathbf{f'}) \mathbf{A}_{\mathbf{i}, \mathbf{f}, \mathbf{f'}} \mathbf{A}_{\mathbf{j}, \mathbf{j'}, \mathbf{j''}}. \tag{1}$$

Here $\omega(\mathbf{k})$ is the frequency of the mode of the wave vector $\mathbf{k}$ and the polarization $j$; $\mathbf{a}_{-\mathbf{k}, j}$ and $\mathbf{a}_{\mathbf{k}, j}$ are the creation and annihilation operators;

$$V^{(3)}(\mathbf{k}, \mathbf{j}, \mathbf{j'}, \mathbf{j''}, \mathbf{i}, \mathbf{f}, \mathbf{f'})$$

is the third-order Fourier transform of the molecular force constants:

$$V^{(3)}(\mathbf{k}, \mathbf{j}, \mathbf{j'}, \mathbf{j''}, \mathbf{i}, \mathbf{f}, \mathbf{f'}) = \frac{\hbar^3}{2^{\frac{3}{2}} \cdot 6 \cdot N^\frac{3}{2}} \Phi(\mathbf{k}, \mathbf{j}, \mathbf{j'}, \mathbf{j''}, \mathbf{i}, \mathbf{f}, \mathbf{f'}) \times$$

$$\times [a(\mathbf{k}, \mathbf{j}) a(\mathbf{k}, \mathbf{j'}) a(\mathbf{k}, \mathbf{j''})]^{-\frac{1}{2}},$$

where

$$\Phi(\mathbf{k}, \mathbf{j}, \mathbf{j'}, \mathbf{j''}, \mathbf{i}, \mathbf{f}, \mathbf{f'}) = \sum_{\alpha, \beta = 1}^{2} \sum_{\mathbf{I}, \mathbf{J}} \Phi_{\alpha, \beta, \mathbf{I}, \mathbf{J}} (\mathbf{0}, \mathbf{I}) \times$$

$$\times [M_{\alpha, 1} M_{\beta, 3} M_{\alpha, 1}^{-\frac{1}{2}} \mathbf{e}_{\alpha} (\mathbf{I}) \mathbf{e}_{\beta} (\mathbf{I}) \mathbf{e}_{\alpha} (\mathbf{I})] \times$$

$$\times \exp [i \mathbf{Q} \mathbf{x}(\mathbf{I}) + i \mathbf{k} \mathbf{x}(\mathbf{I})];$$

in which $\Phi_{\alpha, \beta, \mathbf{I}, \mathbf{J}} (\mathbf{0}, \mathbf{I})$ is the third-order molecular force constant introduced in [1]. Here $\alpha = 1, 2$ indicates the connection between the force constant and the $i$-th component of the linear displacement, and $\alpha = 1, 2$ indicates the connection with the corresponding component of the angular displacement; $M_{\alpha, 1} M_{\alpha, 1}^{-\frac{1}{2}} = \Theta_1 \mathbf{I}$ is the moment of inertia of the molecule about the $i$-th axis. Similar expressions also exist for $V^{(4)}(\mathbf{k}, \mathbf{j}, \mathbf{j'}, \mathbf{j''}, \mathbf{i}, \mathbf{f}, \mathbf{f'}, \mathbf{f''}).$ The cross section of single-phonon coherent scattering can be written in terms of the correlation function of the displacement [2], assuming that the molecules are rigid:

$$\sigma_{\text{coh}}^{(1)} = \sum_{i} \exp [i \mathbf{Q} \mathbf{x}(\mathbf{I})] \sum_{\mathbf{v}, \mathbf{v'}} F_{\mathbf{v}, \mathbf{v'}} \times$$

$$\times \int_{-\infty}^{\infty} dt \exp (i \omega t) \langle \{ \mathbf{Q} \mathbf{S}_{\mathbf{v}}^{(0)} \} \{ \mathbf{Q} \mathbf{S}_{\mathbf{v'}}^{(t)} \} \rangle. \tag{4}$$
We have here introduced the following notation: \(Q\) is the momentum transmitted by the neutron, \(t\) is the time, \(x(l)\) is the radius vector of the nodal point \(l\);

\[
S_{\mu}^t = \sum_{\mu} \sum_{j=1}^{g} \xi_{\mu}(\kappa_j) \exp [i \kappa x(l)] A_{\kappa_j}
\]

(5)
is the displacement operator of the \(\mu\)-th atom of the \(l\)-th molecule; \(S_{\mu}^t(t)\) is the same displacement operator in the Heisenberg representation;

\[
\xi_{\mu}(\kappa_j) = \xi_{\mu j} \sqrt{\frac{\hbar}{2MN^2(\kappa_j)}} \left[ \frac{\epsilon_j^{(\kappa)}}{j} \right] +
\]

(6)

\(N\) is the number of cells in the crystal; \(q_j^\mu\) is the radius vector of the \(\mu\)-th atom relative to the center of gravity of the molecule; \(\mu_{kj}\) and \(\xi_{kj}\) denote the contribution of the rotational and translational displacement of the molecule to the displacement of the \(\mu\)-th atom \((\xi_{kj}^\mu + \xi_{kj}^\mu = 1)\);

\[
F_{\mu'\mu} = V \sigma_{\mu'\mu} \exp [i Q(a_{\mu'} - a_{\mu})] \times
\]

(7)

\[
\times \exp \left[ - \frac{1}{2} \left\{ \langle QS_{\mu'}^*(0) \rangle^2 - \frac{1}{2} \right\} \right];
\]

\(\sigma_{\mu'\mu}\) is the cross section for coherent scattering of neutrons by the \(\mu\)-th atom.

The form of the peak obtained in scattering is determined by the correlation function of the displacement in expression (4). Substituting (5) and (6) into (4) we have

\[
q_{(1)}^{\text{coh}} = \sum_{\mu, \mu'} \int_{-\infty}^{\infty} dt \exp (i\omega t) \sum_{\mu, j} \xi_{\mu j} \xi_{\mu' j} F_{\mu'\mu} \times \left\langle A_{\mu}^*(0) A_{\mu'}(t) \right\rangle.
\]

(8)

If we ignore the factors which do not depend on time, this expression is the Fourier transform of the correlation function, which can be expressed in terms of the Green's temperature function \(G(\kappa j', \omega)\) as follows [3]:

\[
\lim_{\epsilon \to 0^+} \frac{\beta h}{2\pi} \left[ \frac{G(\kappa j', \omega \pm i\epsilon) - G(\kappa j', \omega \pm i\epsilon)}{\exp (\beta h\omega) - 1} \right].
\]

(9)

The first term within the braces corresponds to the transfer of neutron energy \(\hbar \omega\) to the crystal, and in the second term corresponds to absorption of energy by the neutron. In this case