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INELASTIC NEUTRON SCATTERING FROM FERROELECTRICS

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Using the method of double-time thermal Green's function, an expression is derived for one-phonon differential cross section for the coherent inelastic scattering of thermal neutrons by a ferroelectric crystal from a modification of Silverman and Joseph Hamiltonian augmented with dominant fourth- and fifth-order anharmonic terms in the lattice energy. The scattering function is shown to have a Lorentzian form due to anharmonic terms. Expressions for the frequency shift and the width of the maximum of the peak are obtained.

1. Introduction

The study of finer characteristics of energy spectrum of neutrons inelastically scattered by a crystal is quite useful from the point of view of dynamical properties of crystals [1]. It is found that in the scattering of slow neutrons from a crystal, the coherent inelastic part consists of a series of peaks representing the absorption or emission of a number of phonons. In the harmonic approximation, these peaks are infinitely narrow and are described by δ-functions centred at phonon frequencies. The elastic term gives rise to peaks in the energy spectrum of scattered nautrons at the Bragg angle [2]. For a real crystal, the presence of anharmonic forces leads to the diffusion of peaks and the peaks are broadened due to the finite lifetime of phonons on account of anharmonic interactions between normal modes of vibrations and are shifted relative to those predicted by the harmonic approximation. A study of shape, shift of the maximum and the width of the peaks is obviously of considerable interest for several different problems in solid state physics. The observed width of the peaks can give a direct measure of the phonon mean lifetime being the inverse of it.

Weinstock [3] in a fundamental paper first gave a basic theory of neutron scattering by crystals by considering processes in which a neutron either creates or absorbs a single phonon, i.e., one-phonon processes. It has been amplified in various aspects by a number

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of workers [4]. Within the last few years, the coherent scattering of thermal neutrons by anharmonic crystals has been theoretically studied by several authors [5–7] using different approaches. Van Hove [8] used time-independent perturbation theory at zero temperature. Kokkedee [9] has extended this work to finite temperatures using time-dependent perturbation theory. Baym [10] gave a different treatment for neutron scattering from an anharmonic crystal which involves the evaluation of the Fourier transform in space and time of the time-relaxed displacement correlation function, the phonon propagator.

Silverman and Joseph [11] have propounded a Hamiltonian for displacive ferroelectric crystals in the paraelectric phase by making use of available information about the temperature dependence of lowest transverse optic mode. Jaiswal and Sharma [12] have used this Hamiltonian to investigate the electric field dependence of the Curie temperature. Nettleton [13] has modified the Silverman-Joseph Hamiltonian to include effects due to fourth-order anharmonic terms in the lattice potential.

In this paper we report a theoretical study of one-phonon differential cross section for coherent inelastic scattering of neutrons from a ferroelectric crystal using the method of double-time Green's function [14] by considering fifth-order anharmonism of the lattice vibrations. In Section 2 we give a general formalism of the scattering cross-section. Section 3 describes a modification of the Silverman–Joseph Hamiltonian with allowance for fifth-order anharmonicities and presents evaluation of double-time Green's functions by the equation of motion method. In Section 4 expressions for differential cross section for scattering by acoustical and optical phonons are obtained.

2. General formulation

From the well known Fermi scattering theory, the expression for the differential cross section per unit solid angle and per unit interval of outgoing energy ε of the scattered neutron for coherent scattering can be written [4, 15]

$$\frac{d^2\sigma_{\mathrm{coh}}}{d\Omega d\varepsilon} = \frac{1}{\hbar}\,\frac{q_1}{q_0}\,S(Q, \omega), \tag{1}$$

where the scattering factor $S(Q, \omega)$ is given by

$$S(Q, \omega) = \frac{1}{2\pi}\sum_{ss'}\sum_{KK'} b_K b^*_{K'} e^{-i\boldsymbol{Q}\cdot[\boldsymbol{R}(sK)-\boldsymbol{R}(s'K')]} \times \int_{-\infty}^{\infty} dt e^{i\omega t}\langle e^{-i\boldsymbol{Q}\cdot\boldsymbol{u}(sK;t)} e^{i\boldsymbol{Q}\cdot\boldsymbol{u}(s'K';0)}\rangle. \tag{2}$$

In these equations, $\boldsymbol{q}_0$ and $\boldsymbol{q}_1$ are the wave vectors of the neutron before and after scattering, $\boldsymbol{Q} = \boldsymbol{q}_0 - \boldsymbol{q}_1$ is the scattering vector, $\hbar\omega$ is the energy transferred from the neutron to the crystal, b_K is the scattering length of the nucleus K, $\boldsymbol{R}(sK)$ is the position vector of the mean position of the K^{th} atom in the s^{th} unit cell and $\boldsymbol{u}(sK)$ is the displacement from the equilibrium position of that atom. The position vector $\boldsymbol{R}(sK)$ can be written as

$$\boldsymbol{R}(sK) = \boldsymbol{x}(s) + \boldsymbol{r}(K), \tag{3}$$

where $\boldsymbol{x}(s)$ is a crystal translation vector and the vector $\boldsymbol{r}(K)$ describes the position of atom K in the unit cell, $\boldsymbol{u}(sK; t)$ is the operator of displacement in the Heisenberg representation

$$\boldsymbol{u}(sK; t) = e^{i(t/\hbar)H}\boldsymbol{u}(sK; 0)e^{-i(t/\hbar)H}, \tag{4}$$

H being the Hamiltonian of the system and the angular brackets $\langle \ldots \rangle$ represent the canonical ensemble average of the expectation value of an operator, namely

$$\langle 0 \rangle = \mathrm{Tr}\,(e^{-\beta H}0)/\mathrm{Tr}\,(e^{-\beta H}), \tag{5}$$

where Tr denotes the trace of the expression and $\beta = 1/k_{\mathrm{B}}T$, k_{B} being the Boltzmann constant and T the absolute temperature.

The correlation function in Eq. (2) can be converted into a form involving simpler correlation functions by following the approach elaborated by Maradudin and Fein [6] and Baym [10]. The result is

$$\langle e^{-i\boldsymbol{Q}\cdot\boldsymbol{u}(sK;t)}e^{i\boldsymbol{Q}\cdot\boldsymbol{u}(s'K';0)}\rangle = e^{-[W(K)+W(K')]}e^{\langle \boldsymbol{Q}\cdot\boldsymbol{u}(sK;t)\boldsymbol{Q}\cdot\boldsymbol{u}(s'K';0)\rangle} + \ldots \tag{6}$$

where $W(K)$ is the Debye–Waller factor exponent corrected for anharmonic effects and the dots represent the time-relaxed correlation functions arising from anharmonic terms in the potential energy of the crystal. Expressions for them are given in Maradudin and Fein [6]. The contributions from these terms are at least two orders of magnitude smaller than the anharmonic contributions to the average $\langle \boldsymbol{Q}\cdot\boldsymbol{u}(sK; t)\,\boldsymbol{Q}\cdot\boldsymbol{u}(s'K', 0)\rangle$. To the lowest nonvanishing order in the anharmonic interactions, the dotted terms in Eq. (6) can be neglected and the scattering function $S(\boldsymbol{Q}, \omega)$ can be expanded in power of atomic displacements. This gives

$$S(\boldsymbol{Q}, \omega) = S_0(\boldsymbol{Q}, \omega) + S_1(\boldsymbol{Q}, \omega) + \ldots, \tag{7}$$

where

$$S_0(\boldsymbol{Q}, \omega) = N^2\delta(\omega)\Delta(\boldsymbol{Q})\,|\sum_K b_K e^{-W(K)}e^{-i\boldsymbol{Q}\cdot\boldsymbol{r}(K)}|^2 \tag{8a}$$

and

$$S_1(\boldsymbol{Q}, \omega) = \frac{1}{2\pi}\sum_{sK}\sum_{s'K'} b_K b_{K'}^{*} e^{-[W(K)+W(K')]}e^{-i\boldsymbol{Q}\cdot[\boldsymbol{R}(sK)-\boldsymbol{R}(s'K')]} \times \int_{-\infty}^{\infty} dt e^{i\omega t}\langle \boldsymbol{Q}\cdot\boldsymbol{u}(sK; t)\boldsymbol{Q}\cdot\boldsymbol{u}(s'K'; 0)\rangle. \tag{8b}$$

$\Delta(\boldsymbol{Q})$ is equal to unity if $\boldsymbol{Q}$ equals a translation vector of the reciprocal lattice and vanishes otherwise. The first term in the expansion (7) represents coherent elastic scattering from a crystal involving no phonons, because of the factors $\Delta(\omega)$ and $\Delta(\boldsymbol{Q})$. The second term describes the coherent inelastic scattering of neutrons from the crystal by one phonon processes. For temperatures well below the Debye temperature, the one-phonon processes give dominant contribution to scattering cross-section. It is with the evaluation of $S_1(Q, \omega)$ that we will be concerned in this paper.

The atomic displacement $\boldsymbol{u}(sK; 0) = \boldsymbol{u}(sK)$ can be expressed in terms of normal modes of vibration as

$$\boldsymbol{u}(sK) = \sum_{kj} \left(\frac{\hbar}{2M_K N \omega_{kj}}\right)^{1/2} \boldsymbol{e}(K; kj) A_{kj} e^{i\boldsymbol{k}\cdot\boldsymbol{R}(sK)}, \tag{9}$$

where N is the number of unit cells in the crystal, ω_{kj} is the frequency of the normal mode described by the wave vector $\boldsymbol{k}$ and polarization index j, $\boldsymbol{e}(K, kj)$ is the polarization vector and $A_{kj} = a_{kj} + a^{+}_{-kj}$, a_{kj} and a^{+}_{kj} being the phonon annihilation and creation operators. The scattering function then becomes

$$S_1(Q, \omega) = \frac{\hbar}{2\pi N} \sum_{sK} \sum_{s'K'} \sum_{kj} \sum_{k'j'} b_K b^{*}_{K'} e^{-[W(K)+W(K')]}$$
$$\times \left(\frac{1}{M_K M_{K'}}\right)^{1/2} \left(\frac{1}{4\omega_{kj}\omega_{k'j'}}\right)^{1/2} [\{Q \cdot \boldsymbol{e}(K, kj)\}\{Q \cdot \boldsymbol{e}^{*}(K', k'j')\}]$$
$$\times e^{-i(Q-k)\cdot \boldsymbol{R}(sK) + i(Q+k')\cdot \boldsymbol{R}(s'K')} \int_{-\infty}^{\infty} dt e^{i\omega t} \langle A_{kj}(t) A^{+}_{k'j'}(0)\rangle. \tag{10}$$

For convenience in what follows we use only one index k for pair of indices $\boldsymbol{kj}$. From the cyclic boundary conditions, Eq. (10) can be written as

$$S_1(Q, \omega) = \frac{N}{2\pi} \sum_{kk'} \Delta(Q-k)\Delta(Q+k') F(Q, k) F^{*}(Q, -k')$$
$$\times \left(\frac{1}{4\omega_k \omega_{k'}}\right)^{1/2} \int_{-\infty}^{\infty} dt e^{i\omega t} \langle A_k(t) A^{+}_{k'}(0)\rangle, \tag{11}$$

where

$$F(Q, k) = \sum_{K} b_K \left(\frac{\hbar}{M_K}\right)^{1/2} [Q \cdot \boldsymbol{e}(K, k)] e^{-W(K)} e^{-i[(Q-k)\cdot \boldsymbol{R}(K)]}. \tag{12}$$

Equation (11) shows that the problem of finding cross-section becomes one of evaluating the Fourier coefficients of the correlation function $\langle A_k(t) A^{+}_{k'}(0)\rangle$. These are determined by the dynamic properties of the system, i.e., by its Hamiltonian and can be evaluated by several techniques. Here we use the method of double-time Green's function as elaborated by Zubarev [14].

3. *The Hamiltonian and Green's function*

In contrast to other systems, in ferroelectrics the frequency corresponding to transverse optic mode with zero wave vector is imaginary in the harmonic approximation and a lattice instability results. The stabilization of this mode can only be brought about by taking into consideration of anharmonic interactions between the ions. It has been shown [11] that the lowest-order anharmonic interaction that can stabilize the soft ferroelectric mode frequency is of the fourth-order. In the present work, we include all lowest-order effects of anharmonic interactions through the fifth-order of anharmonicity.

The Hamiltonian of the ferroelectric crystal is constructed from the crystal model proposed by Silverman and Joseph [11] by augmenting it with fourth- and fifth-order anharmonic interaction terms involving ferroelectric optic modes of lowest wave vector. These processes are selected because the soft modes of lowest wave number, due to their large occupation number, should cause appreciable scattering of neutrons. The harmonic contribution to the Hamiltonian H_{h} can be written as

$$H_{\mathrm{h}} = \sum_k \hbar\omega_k^{\mathrm{a}}(a_k^{\mathrm{a}+}a_k^{\mathrm{a}}+\tfrac{1}{2}) + \sum_k \hbar\omega_k^{\mathrm{o}}(a_k^{\mathrm{o}+}a_k^{\mathrm{o}}+\tfrac{1}{2}) + \tfrac{1}{2}\left[(p_0^{\mathrm{o}})^2-(\omega_0^{\mathrm{o}}q_0^{\mathrm{o}})^2\right], \tag{13}$$

where superscripts o and a denote the optical and acoustical modes, respectively. All the long wavelength soft optic modes which become unstable in the harmonic approximation are lumped together into a single mode of zero wave vector with normal coordinate q_0^{o} and conjugate momentum p_0^{o} and are assigned as imaginary frequency $i\omega_0^{\mathrm{o}}$.

In the problem considered here the main role is played by the anharmonic interactions. Using Szigeti's [16] theorem which asserts that for a crystal in which each atom is at a centre of inversion symmetry, the coordinates of the optical modes come in pairs, the anharmonic Hamiltonian H_{a} which includes dominant third, fourth and fifth-order anharmonic terms in the expansion of the lattice potential energy in power of ionic displacements can be written as

$$\begin{aligned} H_{\mathrm{a}} = {} & \sum_k \frac{\hbar}{(\omega_k^{\mathrm{o}}\omega_k^{\mathrm{a}})^{1/2}}\frac{q_0^{\mathrm{o}}}{\sqrt{N}} F(k)A_k^{\mathrm{a}}A_k^{\mathrm{o}+} + \sum_k \frac{\hbar}{\omega_k^{\mathrm{o}}}\frac{(q_0^{\mathrm{o}})^2}{N} G^{\mathrm{o}}(k)A_k^{\mathrm{o}}A_k^{\mathrm{o}+} \\ & + \sum_k \frac{\hbar}{\omega_k^{\mathrm{a}}}\frac{(q_0^{\mathrm{o}})^2}{N} G^{\mathrm{a}}(k)A_k^{\mathrm{a}}A_k^{\mathrm{a}+} + \sum_{k_1,k_2,k_3} \frac{\hbar^{3/2}}{(\omega_{k_1}^{\mathrm{o}}\omega_{k_2}^{\mathrm{a}}\omega_{k_3}^{\mathrm{a}})^{1/2}}\frac{q_0^{\mathrm{o}}}{\sqrt{N}} \Phi(k_1,k_2,k_3)A_{k_1}^{\mathrm{o}}A_{k_2}^{\mathrm{a}}A_{k_3}^{\mathrm{a}} \\ & + \sum_{k_1,k_2,k_3} \frac{\hbar^{3/2}}{(\omega_{k_1}^{\mathrm{o}}\omega_{k_2}^{\mathrm{o}}\omega_{k_3}^{\mathrm{o}})^{1/2}}\frac{q_0^{\mathrm{o}}}{\sqrt{N}} \psi(k_1,k_2,k_3)A_{k_1}^{\mathrm{o}}A_{k_2}^{\mathrm{o}}A_{k_3}^{\mathrm{o}} \\ & + \sum_{k_1,k_2,k_3,k_4} \frac{\hbar^2}{(\omega_{k_1}^{\mathrm{a}}\omega_{k_2}^{\mathrm{o}}\omega_{k_3}^{\mathrm{o}}\omega_{k_4}^{\mathrm{o}})^{1/2}}\frac{q_0^{\mathrm{o}}}{\sqrt{N}} \xi(k_1,k_2,k_3,k_4)A_{k_1}^{\mathrm{a}}A_{k_2}^{\mathrm{o}}A_{k_3}^{\mathrm{o}}A_{k_4}^{\mathrm{o}} \\ & + \sum_{k_1,k_2,k_3,k_4} \frac{\hbar^2}{(\omega_{k_1}^{\mathrm{o}}\omega_{k_2}^{\mathrm{a}}\omega_{k_3}^{\mathrm{a}}\omega_{k_4}^{\mathrm{a}})^{1/2}}\frac{q_0^{\mathrm{o}}}{\sqrt{N}} \eta(k_1,k_2,k_3,k_4)A_{k_1}^{\mathrm{o}}A_{k_2}^{\mathrm{a}}A_{k_3}^{\mathrm{a}}A_{k_4}^{\mathrm{a}}. \end{aligned} \tag{14}$$

Here the quantities $F(k)$, $G^{o}(k)$, $G^{a}(k)$, $\Phi(k_1, k_2, k_3)$, $\psi(k_1, k_2, k_3)$, $\xi(k_1, k_2, k_3, k_4)$ and $\eta(k_1, k_2, k_3, k_4)$ are Fourier transformed third, fourth and fifth-order anharmonic coupling constants. These terms generate three and four-phonon processes and make significant contribution to the lifetime of optical as well as acoustical phonons. From Eqs. (13) and (14) the Hamiltonian of the system is given by

$$H = H_{\mathrm{h}} + H_{\mathrm{a}}. \tag{15}$$

We now introduce one-particle retarded double-time Green's functions [14] of the system for the acoustical and optical phonons as

$$G^{a}_{kk'}(t) = \langle\langle A^{a}_{k}(t); A^{a}_{k'}(0)\rangle\rangle = -i\theta(t)\langle[A^{a}_{k}(t), A^{a+}_{k'}(0)]\rangle, \tag{16a}$$

and

$$G^{o}_{kk'}(t) = \langle\langle A^{o}_{k}(t); A^{o+}_{k'}(0)\rangle\rangle = -i\theta(t)\langle[A^{o}_{k}(t), A^{o+}_{k'}(0)]\rangle, \tag{16b}$$

where $\theta(t)$ is the usual unit step function: $\theta(t) = 1$ for $t > 0$, $\theta(t) = 0$ for $t < 0$. The correlation function can be expressed as [14]

$$f_{kk'}(t) = \langle A_k(t)A^{+}_{k'}(0)\rangle = \int_{-\infty}^{\infty} d\omega e^{\beta\hbar\omega} J_{kk'}(\omega) e^{-i\omega t}, \tag{17}$$

where $J_{kk'}(\omega)$ is the spectral density function of the related Green's function and is given by

$$J_{kk'}(\omega) = \lim_{\varepsilon\to 0} \frac{i}{(e^{\beta\hbar\omega}-1)} [G_{kk'}(\omega+i\varepsilon) - G_{kk'}(\omega-i\varepsilon)], \tag{18}$$

$G_{kk'}(\omega)$ being the Fourier transform of the double-time Green's function.

Differentiating expression (16a) with respect to time t, the equation of motion for Green's function $G^{a}_{kk'}(t)$ is

$$i\hbar \frac{d}{dt} G^{a}_{kk'}(t) = \hbar\delta(t)\langle[A^{a}_{k}(t), A^{a+}_{k'}(0)]\rangle + \langle\langle[A^{a}_{k}(t), H]; A^{a+}_{k'}(0)\rangle\rangle. \tag{19}$$

For the Hamiltonian (15), using the commutation property of the operators, the above equation can be written as

$$i\hbar \frac{d}{dt} G^{a}_{kk'}(t) = \hbar\omega^{a}_{k}\langle\langle B^{a}_{k}(t); A^{a+}_{k'}(0)\rangle\rangle, \tag{20}$$

where $B_k = a_k - a^{+}_{-k}$. Differentiating Eq. (20) again with respect to time argument t, we obtain

$$-\hbar \frac{d^2}{dt^2} G^{a}_{kk'}(t) = \hbar\omega^{a}_{k}\delta(t)\langle[B^{a}_{k}(t), A^{a+}_{k'}(0)\rangle + \omega^{a}_{k}\langle\langle[B^{a}_{k}(t), H]; A^{a+}_{k'}(0)\rangle\rangle. \tag{21}$$

The operators A_k and B_k satisfy the commutation relation

$$[B_k, A_{k'}] = 2\delta_{k,-k'}, \tag{22}$$

which follows from the commutation relations for creation and annihilation operators. This gives

$$-\frac{d^2}{dt^2}G^{a}_{kk'}(t) = 2\omega^{a}_{k}\delta(t)\delta_{k,-k'}+(\omega^{a}_{k})^2G^{a}_{kk'}(t)+\frac{4}{N}(q^{\circ}_{0})^2G^{a}(k)G^{a}_{kk'}(t)$$

$$-i\theta(t)\frac{4}{\sqrt{N}}\sum_{k_1,k_2}q^{\circ}_{0}\Phi(k_1,k_2,-k)\left(\frac{\hbar\omega^{a}_{k}}{\omega^{\circ}_{k_1}\omega^{a}_{k_2}}\right)^{1/2}\langle[A^{\circ}_{k_1}(t)A^{a}_{k_2}(t),A^{a+}_{k'}(0)]\rangle$$

$$+\frac{6}{\sqrt{N}}\sum_{k_1,k_2,k_3}q^{\circ}_{0}\eta(k_1,k_2,k_3,-k)\left(\frac{\hbar^2\omega^{a}_{k}}{\omega^{\circ}_{k_1}\omega^{a}_{k_2}\omega^{a}_{k_3}}\right)^{1/2}\langle\langle A^{\circ}_{k_1}A^{a}_{k_2}A^{a}_{k_3};A^{a+}_{k'}(0)\rangle\rangle. \tag{23}$$

We now factor the expectation values of acoustical and optical operators so that the above equation may be written as

$$-\frac{d^2}{dt^2}G^{a}_{kk'}(t) = 2\omega^{a}_{k}\delta(t)\delta_{k,-k'}+(\omega^{a}_{k})^2G^{a}_{kk'}(t)+\frac{4}{N}(q^{\circ}_{0})^2G^{a}(k)G^{a}_{kk'}(t)$$

$$-i\theta(t)\frac{4}{\sqrt{N}}\sum_{k_1,k_2}q^{\circ}_{0}\Phi(k_1,k_2,-k)\left(\frac{\hbar\omega^{a}_{k}}{\omega^{\circ}_{k_1}\omega^{a}_{k_2}}\right)^{1/2}\langle A^{\circ}_{k_1}(t)\rangle\langle[A^{a}_{k_2}(t),A^{a+}_{k'}(0)]\rangle$$

$$+\frac{6}{\sqrt{N}}\sum_{k_1,k_2,k_3}q^{\circ}_{0}\eta(k_1,k_2,k_3,-k)\left(\frac{\hbar^2\omega^{a}_{k}}{\omega^{\circ}_{k_1}\omega^{a}_{k_2}\omega^{a}_{k_3}}\right)^{1/2}\langle A^{\circ}_{k_1}(t)\rangle\langle\langle A^{a}_{k_2}(t)A^{a}_{k_3}(t);A^{a+}_{k'}(0)\rangle\rangle. \tag{24}$$

The Green's function

$$G^{(1)}_{k_2k_3k'} = \langle\langle A^{a}_{k_2}A^{a}_{k_3};A^{a+}_{k'}(0)\rangle\rangle \tag{25a}$$

appearing in the last term is a third-order Green's function. To evaluate it we introduce the following retarded Green's functions

$$G^{(2)}_{k_2k_3k'} = \langle\langle B^{a}_{k_2}A^{a}_{k_3};A^{a+}_{k'}(0)\rangle\rangle, \tag{25b}$$

$$G^{(3)}_{k_2k_3k'} = \langle\langle A^{a}_{k_2}B^{a}_{k_3};A^{a+}_{k'}(0)\rangle\rangle, \tag{25c}$$

and

$$G^{(4)}_{k_2k_3k'} = \langle\langle B^{a}_{k_2}B^{a}_{k_3};A^{a+}_{k'}(0)\rangle\rangle. \tag{25d}$$

The equations of motion for these Green's functions are

$$i\hbar\frac{d}{dt}G^{(1)}_{k_2k_3k'} = \hbar\omega^{a}_{k_2}G^{(2)}_{k_2k_3k'}+\hbar\omega^{a}_{k_3}G^{(3)}_{k_2k_3k'}, \tag{26a}$$

$$i\hbar\frac{d}{dt}G^{(2)}_{k_2k_3k'} = \hbar\omega^{a}_{k_3}G^{(4)}_{k_2k_3k'}+\hbar\omega^{a}_{k_2}G^{(1)}_{k_2k_3k'}+\frac{4}{N}(q^{\circ}_{0})^2G^{a}(k_2)\frac{\hbar}{\omega^{a}_{k_2}}G^{(1)}_{k_2k_3k'}$$

$$+\frac{4}{\sqrt{N}}\sum_{k_1',k_2'} q_0^{\rm o}\Phi(k_1',k_2',-k_2)\left(\frac{\hbar\omega_k^{\rm a}}{\omega_{k_1'}^{\rm o}\omega_{k_2'}^{\rm a}}\right)^{1/2}\langle A_{k_1}^{\rm o}(t)\rangle G_{k_2k_3k'}^{(1)}$$

$$+\frac{6}{\sqrt{N}}\sum_{k_1',k_2',k_3'} q_0^{\rm o}\eta(k_1',k_2',k_3',-k_3)\frac{\hbar(\omega_k^{\rm a})^{1/2}}{(\omega_{k_1'}^{\rm o}\omega_{k_2'}^{\rm a}\omega_{k_3'}^{\rm a})^{1/2}}\langle A_{k_1}^{\rm o}(t)\rangle \langle\langle A_{k_2'}^{\rm a}A_{k_3'}^{\rm a}A_{k_3}^{\rm a}; A_{k'}^{\rm a+}(0)\rangle\rangle, \tag{26b}$$

$$i\hbar\frac{d}{dt}G_{k_2k_3k'}^{(3)} = \hbar\omega_{k_2}^{\rm a}G_{k_2k_3k'}^{(4)}+\hbar\omega_{k_3}^{\rm a}G_{k_2k_3k'}^{(1)}+\frac{4}{N}(q_0^{\rm o})^2G^{\rm a}(k_3)\frac{\hbar}{\omega_{k_3}^{\rm a}}G_{k_2k_3k'}^{(1)}$$

$$+\frac{4}{\sqrt{N}}\sum_{k_1',k_2'} q_0^{\rm o}\Phi(k_1',k_2',-k_2)\left(\frac{\hbar\omega_k^{\rm a}}{\omega_{k_1'}^{\rm o}\omega_{k_2'}^{\rm a}}\right)^{1/2}\langle A_{k_1}^{\rm o}(t)\rangle G_{k_2k_3k'}^{(1)}$$

$$+\frac{6}{\sqrt{N}}\sum_{k_1',k_2',k_3'} q_0^{\rm o}\eta(k_1',k_2',k_3',-k_3)\frac{\hbar(\omega_k^{\rm a})^{1/2}}{(\omega_{k_1'}^{\rm a}\omega_{k_2}^{\rm a}\omega_{k_3'}^{\rm a})^{1/2}}\langle A_{k_1}^{\rm o}(t)\rangle \langle\langle A_{k_2'}^{\rm a}A_{k_3'}^{\rm a}A_{k_2}^{\rm a}; A_{k'}^{\rm a+}(0)\rangle\rangle, \tag{26c}$$

$$i\hbar\frac{d}{dt}G_{k_2k_3k'}^{(4)} = \hbar\omega_{k_3}^{\rm a}G_{k_2k_3k'}^{(2)}+\hbar\omega_{k_2}^{\rm a}G_{k_2k_3k'}^{(3)}$$

$$+\frac{4}{N}(q_0^{\rm o})^2G^{\rm a}(k_3)\frac{\hbar}{\omega_{k_3}^{\rm a}}G_{k_2k_3k'}^{(2)}+\frac{4}{N}(q_0^{\rm o})^2G^{a}(k_2)\frac{\hbar}{\omega_{k_2}^{\rm a}}G_{k_2k_3k'}^{(3)}$$

$$+\frac{4}{\sqrt{N}}\sum_{k_1',k_2'} q_0^{\rm o}\Phi(k_1',k_2',-k_2)\left(\frac{\hbar\omega_k^{\rm a}}{\omega_{k_1'}^{\rm o}\omega_{k_2'}^{\rm a}}\right)^{1/2}\langle A_{k_1}^{\rm o}(t)\rangle G_{k_2k_3k'}^{(2)}$$

$$+\frac{4}{\sqrt{N}}\sum_{k_1',k_2'} q_0^{\rm o}\Phi(k_1',k_2',-k_1)\left(\frac{\hbar\omega_k^{\rm a}}{\omega_{k_1'}^{\rm o}\omega_{k_2'}^{\rm a}}\right)^{1/2}\langle A_{k_1}^{\rm o}(t)\rangle G_{k_2k_3k'}^{(3)}$$

$$+\frac{6}{\sqrt{N}}\sum_{k_1',k_2',k_3'} q_0^{\rm o}\eta(k_1',k_2',k_3',-k_3)\frac{\hbar(\omega_k^{\rm a})^{1/2}}{(\omega_{k_1'}^{\rm o}\omega_{k_2'}^{\rm a}\omega_{k_3'}^{\rm a})^{1/2}}\langle A_{k_1}^{\rm o}(t)\rangle \langle\langle A_{k_2'}^{\rm a}A_{k_3'}^{\rm a}B_{k_3}^{\rm a}; A_{k'}^{\rm a+}(0)\rangle\rangle$$

$$+\frac{6}{\sqrt{N}}\sum_{k_1',k_2',k_3'} q_0^{\rm o}\eta(k_1',k_2',k_3',-k_2)\frac{\hbar(\omega_k^{\rm a})^{1/2}}{(\omega_{k_1'}^{\rm o}\omega_{k_2'}^{\rm a}\omega_{k_3'}^{\rm a})^{1/2}}\langle A_{k_1}^{\rm o}(t)\rangle \langle\langle B_{k_2}^{\rm a}A_{k_2'}^{\rm a}A_{k_3'}^{\rm a}; A_{k'}^{\rm a+}(0)\rangle\rangle. \tag{26d}$$

The fourth-order Green's functions in the above equations can be decoupled into sum of products of second-order Green's functions [17, 18]. When such a decoupling has been done, transformation to a Fourier representation gives

$$\left[\omega^2-(\omega_k^{\rm a})^2-\frac{2}{N}(q_0^{\rm o})^2G^{\rm a}(k)\right]G_{kk'}^{\rm a}(\omega)=\frac{\omega_k^{\rm a}\delta_{k,-k'}}{\pi}$$

$$+\frac{4}{\sqrt{N}}\sum_{k_1,k_2}q_0^{\rm o}\Phi(k_1,k_2,-k)\left(\frac{\hbar\omega_k^{\rm a}}{\omega_{k_1}^{\rm o}\omega_{k_2}^{\rm a}}\right)^{1/2}\langle A_{k_1}^{\rm o}\rangle G_{k_2k'}^{\rm a}(\omega)$$

$$+\frac{6}{\sqrt{N}}\sum_{k_1,k_2,k_3}q_0^{\rm o}\eta(k_1,k_2,k_3,-k)\frac{\hbar(\omega_k^{\rm a})^{1/2}}{(\omega_{k_1}^{\rm o}\omega_{k_2}^{\rm a}\omega_{k_3}^{\rm a})^{1/2}}\langle A_{k_1}^{\rm o}\rangle G_{k_2k_3k'}^{(1)}(\omega), \tag{27}$$

where

$$G_{k_2k_3k'}^{(1)}(\omega)=\frac{F(k_2,k_3,\omega)}{\sqrt{N}}\sum_{k_1',k_2'}q_0^{\rm o}\eta(-k_1,-k_2,-k_3,k_1')\frac{\hbar}{(\omega_{k_1}^{\rm o}\omega_{k_2}^{\rm a}\omega_{k_3}^{\rm a}\omega_{k_1'}^{\rm a})^{1/2}}$$

$$\times\langle\langle A_{k_2'}^{\rm a}(t);A_{k'}^{\rm a+}(0)\rangle\rangle_\omega, \tag{28}$$

with

$$F(k_2,k_3,\omega)=6[\langle A_{k_2}^{\rm a+}A_{k_2}^{\rm a}\rangle+\langle A_{k_3}^{\rm a+}A_{k_3}^{\rm a}\rangle]\frac{\omega_{k_2}^{\rm a}+\omega_{k_3}^{\rm a}}{\omega^2-(\omega_{k_2}^{\rm a}+\omega_{k_3}^{\rm a})^2}$$

$$+6[\langle A_{k_3}^{\rm a+}A_{k_3}^{\rm a}\rangle-\langle A_{k_2}^{\rm a+}A_{k_2}^{\rm a}\rangle]\frac{\omega_{k_2}^{\rm a}-\omega_{k_3}^{\rm a}}{\omega^2-(\omega_{k_2}^{\rm a}-\omega_{k_3}^{\rm a})^2}$$

$$+6[\langle B_{k_2}^{\rm a}A_{k_2}^{\rm a+}\rangle+\langle A_{k_3}^{\rm a+}B_{k_3}^{\rm a}\rangle]\left\{\frac{\omega}{\omega^2-(\omega_{k_2}^{\rm a}+\omega_{k_3}^{\rm a})^2}-\frac{\omega}{\omega^2-(\omega_{k_2}^{\rm a}-\omega_{k_3}^{\rm a})^2}\right\}. \tag{29}$$

If we substitute Eq. (28) into Eq. (27), we get

$$G_{kk'}^{\rm a}(\omega)$$

$$=\frac{\omega_k^{\rm a}\delta_{k,-k'}/\pi}{\left[\omega^2-(\omega_k^{\rm a})^2-\frac{4}{N}(q_0^{\rm o})^2G^{\rm a}(k)-\frac{4}{\sqrt{N}}\sum_{k_1}q_0^{\rm o}\Phi(k_1,k,-k)\left(\frac{\hbar}{\omega_{k_1}^{\rm o}}\right)^{1/2}\langle A_{k_1}^{\rm o}\rangle-2\omega_k^{\rm a}M_k^{\rm a}(\omega)\right]}, \tag{30}$$

where

$$M_k^{\rm a}(\omega)=\frac{3}{N}\sum_{k_1,k_2,k_3}(q_0^{\rm o})^2|\eta(k_1,k_2,k_3,-k)|^2\frac{\hbar^2}{(\omega_{k_1}^{\rm o}\omega_{k_2}^{\rm a}\omega_{k_3}^{\rm a})}\langle A_{k_1}^{\rm o}\rangle F(k_2,k_3,\omega). \tag{31}$$

The explicit expression for $M_k^{a}(\omega)$ can be obtained by writing

$$M_k^{a}(\omega+i\varepsilon) = \Delta_k^{a}(\omega)-i\Gamma_k^{a}(\omega). \tag{32}$$

The real part $\Delta_k^{a}(\omega)$ represents the shift of the frequency, while the imaginary part $\Gamma_k^{a}(\omega)$ gives the half-width of the response function of a mode. From Eqs. (31) and (32), we finally obtain

$$\begin{aligned}\Delta_k^{a}(\omega) = \frac{18P}{N}\sum_{k_1,k_2,k_3} (q_0^{o})^2|\eta(k_1, k_2, k_3, -k)|^2 \frac{\hbar^2(\omega_k^{a})}{(\omega_{k_1}^{o}\omega_{k_2}^{a}\omega_{k_3}^{a})}\langle A_{k_1}^{o}\rangle \\ \times\left\{[\langle A_{k_2}^{a+}A_{k_2}^{a}\rangle+\langle A_{k_3}^{a+}A_{k_3}^{a}\rangle]\frac{\omega_{k_2}^{a}+\omega_{k_3}^{a}}{\omega^2-(\omega_{k_2}^{a}+\omega_{k_3}^{a})^2}\right. \\ \left.+[\langle A_{k_3}^{a+}A_{k_3}^{a}\rangle-\langle A_{k_2}^{a+}A_{k_2}^{a}\rangle]\frac{\omega_{k_2}^{a}-\omega_{k_3}^{a}}{\omega^2-(\omega_{k_2}^{a}-\omega_{k_3}^{a})^2}\right\},\end{aligned} \tag{33}$$

and

$$\begin{aligned}\Gamma_k^{a}(\omega) = \frac{18\pi}{N}\varepsilon(\omega)\sum_{k_1,k_2,k_3} (q_0^{o})^2|\eta(k_1, k_2, k_3, -k)|^2 \frac{\hbar^2(\omega_k^{a})}{(\omega_{k_1}^{o}\omega_{k_2}^{a}\omega_{k_3}^{a})}\langle A_{k_1}^{o}\rangle \\ \times\{[\langle A_{k_2}^{a+}A_{k_2}^{a}\rangle+\langle A_{k_3}^{a+}A_{k_3}^{a}\rangle](\omega_{k_2}^{a}+\omega_{k_3}^{a})\delta[\omega^2-(\omega_{k_2}^{a}+\omega_{k_3}^{a})^2] \\ +[\langle A_{k_3}^{a+}A_{k_3}^{a}\rangle-\langle A_{k_2}^{a+}A_{k_2}^{a}\rangle](\omega_{k_2}^{a}-\omega_{k_3}^{a})\delta[\omega^2-(\omega_{k_2}^{a}-\omega_{k_3}^{a})^2]\},\end{aligned} \tag{34}$$

in which P stands for the principal part and $\varepsilon(\omega) = 1$ for $\omega > 0$, $\varepsilon(\omega) = -1$ for $\omega < 0$.

Similarly if we proceed with the equation of motion of Green's function (16b) for optical mode and follow the procedure as used above, we obtain the following equation for the determination of Fourier transform of the Green's function

$$\begin{aligned}\left[\omega^2-(\omega_k^{o})^2-\frac{4}{N}(q_0^{o})^2G^{o}(k)\right]G_{kk'}^{o}(\omega) = \frac{\omega_k^{o}\delta_{k,-k'}}{\pi} \\ +\frac{6\omega_k^{o}}{\sqrt{N}}\sum_{k_1,k_2} q_0^{o}\psi(k_1, k_2, -k)\frac{\hbar^{1/2}}{(\omega_{k_1}^{o}\omega_{k_2}^{o}\omega_k^{o})^{1/2}}G_{k_1k_2k'}^{(1)}(\omega) \\ +\frac{6}{\sqrt{N}}\sum_{k_1,k_2} q_0^{o}\xi(k_1, k_2, -k_2, -k)\frac{\hbar}{(\omega_{k_1}^{a}\omega_{k_2}^{o})^{1/2}}G_{kk'}^{o}(\omega)\langle A_{k_1}^{o+}A_{k_1}^{o}\rangle,\end{aligned} \tag{35}$$

where $G_{k_1k_2k'}^{(1)}(\omega)$ is the Fourier transform of the third-order Green's function

$$G_{k_1k_2k'}^{(1)}(t) = \langle\langle A_{k_1}^{o}A_{k_2}^{o}; A_{k'}^{o+}(0)\rangle\rangle, \tag{36}$$

and is given by

$$G^{(1)}_{k_1k_2k'}(\omega) = \frac{F(k_1, k_2, \omega)}{\sqrt{N}} \sum_{k_1'} q_0^{\circ}\psi(-k_1, -k_2, k_1') \frac{\hbar^{1/2}}{(\omega^{\circ}_{k_1}\omega^{\circ}_{k_2}\omega^{\circ}_{k_1'})^{1/2}} \langle\langle A^{\circ}_{k_1'}(t); A^{\circ +}_{k'}(0)\rangle\rangle_{\omega}, \tag{37}$$

with

$$F(k_1, k_2, \omega) = 6[\langle A^{\circ +}_{k_1} A^{\circ}_{k_1}\rangle + \langle A^{\circ +}_{k_2} A^{\circ}_{k_2}\rangle] \frac{\omega^{\circ}_{k_1}+\omega^{\circ}_{k_2}}{\omega^2-(\omega^{\circ}_{k_1}+\omega^{\circ}_{k_2})^2}$$
$$+6[\langle A^{\circ +}_{k_2} A^{\circ}_{k_2}\rangle - \langle A^{\circ +}_{k_1} A^{\circ}_{k_1}\rangle] \frac{\omega^{\circ}_{k_1}-\omega^{\circ}_{k_2}}{\omega^2-(\omega^{\circ}_{k_1}-\omega^{\circ}_{k_2})^2}$$
$$+6[\langle B^{\circ}_{k_1} A^{\circ +}_{k_1}\rangle + \langle A^{\circ +}_{k_2} B^{\circ}_{k_2}\rangle] \left\{\frac{\omega}{\omega^2-(\omega^{\circ}_{k_1}+\omega^{\circ}_{k_2})^2} - \frac{\omega}{\omega^2-(\omega^{\circ}_{k_1}-\omega^{\circ}_{k_2})^2}\right\}. \tag{38}$$

Combining Eqs. (35) and (37), we find an expression similar to Eq. (30) for the acoustical phonons

$$G^{\circ}_{kk'}(\omega) = \frac{\omega^{\circ}_k \delta_{k,-k'}/\pi}{\left[\omega^2-(\omega^{\circ}_k)^2 - \frac{4}{N}(q^{\circ}_0)^2 G^{\circ}(k) - \frac{6}{\sqrt{N}} \sum_{k_1,k_2} q^{\circ}_0 \xi(k_1, k_2, -k_2, -k) \times \frac{\hbar}{(\omega^{a}_{k_1}\omega^{\circ}_{k_2})^{1/2}} \langle A^{\circ +}_{k_1} A^{\circ}_{k_1}\rangle - 2\omega^{\circ}_k M^{\circ}_k(\omega)\right]}, \tag{39}$$

where

$$M^{\circ}_k(\omega) = \frac{3}{N} \sum_{k_1,k_2} (q^{\circ}_0)^2 |\psi(-k, k_1, k_2)|^2 \frac{\hbar}{(\omega^{\circ}_k\omega^{\circ}_{k_1}\omega^{\circ}_{k_2})} F(k_1, k_2, \omega). \tag{40}$$

In this case the shift of the frequency $\Delta^{\circ}_k(\omega)$ and the half-width $\Gamma^{\circ}_k(\omega)$ of the response function are given by

$$\Delta^{\circ}_k(\omega) = \frac{18P}{N} \sum_{k_1,k_2} (q^{\circ}_0)^2 |\psi(-k, k_1, k_2)|^2 \frac{\hbar}{(\omega^{\circ}_k\omega^{\circ}_{k_1}\omega^{\circ}_2)}$$
$$\times \left\{[\langle A^{\circ +}_{k_1} A^{\circ}_{k_1}\rangle + \langle A^{\circ +}_{k_2} A^{\circ}_{k_2}\rangle] \frac{\omega^{\circ}_{k_1}+\omega^{\circ}_{k_2}}{\omega^2-(\omega^{\circ}_{k_1}+\omega^{\circ}_{k_2})^2}\right.$$
$$\left.+[\langle A^{\circ +}_{k_2} A^{\circ}_{k_2}\rangle - \langle A^{\circ +}_{k_1} A^{\circ}_{k_1}\rangle] \frac{\omega^{\circ}_{k_1}-\omega^{\circ}_{k_2}}{\omega^2-(\omega^{\circ}_{k_1}-\omega^{\circ}_{k_2})^2}\right\}, \tag{41}$$

and

$$\Gamma_k^{\circ}(\omega) = \frac{18\pi}{N}\,\varepsilon(\omega)\sum_{k_1,k_2}(q_0^{\circ})^2|\psi(-k,k_1,k_2)|^2\,\frac{\hbar}{(\omega_k^{\circ}\omega_{k_1}^{\circ}\omega_{k_2}^{\circ})}$$

$$\times[\langle A_{k_1}^{\circ+}A_{k_1}^{\circ}\rangle+\langle A_{k_2}^{\circ+}A_{k_2}^{\circ}\rangle]\,(\omega_{k_1}^{\circ}+\omega_{k_2}^{\circ})\delta[\omega^2-(\omega_{k_1}^{\circ}+\omega_{k_2}^{\circ})^2]$$

$$+[\langle A_{k_2}^{\circ+}A_{k_2}^{\circ}\rangle-\langle A_{k_1}^{\circ+}A_{k_1}^{\circ}\rangle]\,(\omega_{k_1}^{\circ}-\omega_{k_2}^{\circ})\delta[\omega^2-(\omega_{k_1}^{\circ}-\omega_{k_2}^{\circ})^2]. \tag{42}$$

Having formulated the Green's function, we can easily obtain the correlation function using the prescription (17) and hence the expression for the differential scattering cross section.

4. *Inelastic neutron scattering cross section*

We evaluate separately the acoustical and optical phonon contributions to the scattering cross-section. From Eqs. (17) and (30) the correlation function corresponding to the acoustical mode is given by

$$\langle A_k^{a}(t)A_{k'}^{a+}(0)\rangle = -\frac{2\omega_k^{a}\delta_{k,-k'}}{\pi}\,\mathrm{Im}\int d\omega\,\frac{e^{\beta\hbar\omega}}{(e^{\beta\hbar\omega}-1)}$$

$$\times\frac{e^{-i\omega t}}{\left[\omega^2-(\omega_k^{a})^2-\frac{4}{N}(q_0^{\circ})^2G^{a}(k)-\frac{4}{\sqrt{N}}\sum_{k_1,k_2}q_0^{\circ}\phi(k_1,k_2,-k)\left(\frac{\hbar\omega_k^{a}}{\omega_{k_1}^{\circ}\omega_{k_2}^{a}}\right)^{1/2}\langle A_{k_1}^{\circ}\rangle-2\omega_k^{a}\Delta_k^{a}(\omega)+2i\omega_k^{a}\Gamma_k^{a}(\omega)\right]}$$

$$=\frac{4(\omega_k^{a})^2\delta_{k,-k'}}{\pi}\int_{-\infty}^{\infty}d\omega\,\frac{e^{\beta\hbar\omega}}{(e^{\beta\hbar\omega}-1)}$$

$$\times\frac{e^{-i\omega t}\Gamma_k^{a}(\omega)}{\left\{\left[\omega^2-(\omega_k^{a})^2-\frac{4}{N}(q_0^{\circ})^2G^{a}(k)-\frac{4}{\sqrt{N}}\sum_{k_1,k_2}q_0^{\circ}\phi(k_1,k_2,-k)\left(\frac{\hbar\omega_k^{a}}{\omega_{k_1}^{\circ}\omega_{k_2}^{a}}\right)^{1/2}\langle A_{k_1}^{\circ}\rangle-2\omega_k^{a}\Delta_k^{a}(\omega)\right]^2+4(\omega_k^{a})^2\Gamma_k^{a2}(\omega)\right\}}, \tag{43}$$

where $\Delta_k^a(\omega)$ and $\Gamma_k^a(\omega)$ are the shift and width of frequency and are given by expressions (33) and (34) respectively. Substitution of Eq. (43) into Eq. (11) gives the scattering factor $S_1(Q, \omega)$ as

$$S_1(\boldsymbol{Q}, \omega) = \frac{N}{\pi^2} \sum_{k} \Delta(\boldsymbol{Q}-\boldsymbol{k}) \, |F(\boldsymbol{Q}, \boldsymbol{k})|^2 \omega_k^a \int_{-\infty}^{\infty} d\omega \frac{e^{\beta\hbar\omega}}{(e^{\beta\hbar\omega}-1)}$$

$$\times \frac{\Gamma_k^a(\omega)}{\left\{\left[\omega^2-(\omega_k^a)^2-\frac{4}{N}(q_0^o)^2 G^a(k)-\frac{4}{\sqrt{N}}\sum_{k_1,k_2} q_0^o\phi(k_1, k_2, -k)\left(\frac{\hbar\omega_k^a}{\omega_{k_1}^o\omega_{k_2}^a}\right)^{1/2}\langle A_{k_1}^o\rangle - 2\omega_k^a\Delta_k^a(\omega)\right]^2 + 4(\omega_k^a)^2\Gamma_k^{a2}(\omega)\right\}} . \quad (44)$$

With this result, the one-phonon differential scattering cross section becomes

$$\frac{d^2\sigma_{coh}^a}{d\Omega d\varepsilon} = \frac{N}{\pi^2\hbar}\frac{q_1}{q_0}\sum_{kj} |F(\boldsymbol{Q}, \boldsymbol{k})|^2 \omega_{k,j}^a \int_{-\infty}^{\infty} d\omega \frac{e^{\beta\hbar\omega}}{(e^{\beta\hbar\omega}-1)}$$

$$\times \frac{\Gamma_{kj}^a(\omega)}{\left\{\left[\omega^2-(\omega_{kj}^a)^2-\frac{4}{N}(q_0^o)^2 G^a(k, j)-\frac{4}{\sqrt{N}}\sum_{k_1j_1,k_2j_2} q_0^o\phi(k_1j_1, k_2j_2, -kj) \times \left(\frac{\hbar\omega_{kj}^a}{\omega_{k_1j_1}^o\omega_{k_2j_2}^a}\right)^{1/2}\langle A_{k_1j_1}^o\rangle - 2\omega_{kj}^a\Delta_{kj}^a(\omega)\right]^2 + 4(\omega_{kj}^a)^2\Gamma_{kj}^{a2}(\omega)\right\}} , \quad (45)$$

where the vector $\boldsymbol{k}$ is related to the vector $\boldsymbol{Q}$ by

$$\boldsymbol{k} = \boldsymbol{Q}+\boldsymbol{g}, \quad (46)$$

$\boldsymbol{g}$ being the translation vector of the reciprocal lattice.

In a similar way the one-phonon differential scattering cross section for the optical mode is obtained as

$$\frac{d^2\sigma_{coh}^o}{d\Omega d\varepsilon} = \frac{N}{\pi^2\hbar}\frac{q_1}{q_0}\sum_{kj} |F(\boldsymbol{Q}, \boldsymbol{k})|^2 \omega_{kj}^o \int_{-\infty}^{\infty} d\omega \frac{e^{\beta\hbar\omega}}{(e^{\beta\hbar\omega}-1)}$$

$$\times \frac{\Gamma_{kj}^o(\omega)}{\left\{\left[\omega^2-(\omega_{kj}^o)^2-\frac{4}{N}(q_0^o)G^o(kj)-\frac{6}{\sqrt{N}}\sum_{k_1j_1,k_2j_2} q_0^o\xi(k_1j_1, k_2j_2, -k_2j_2, -kj) \times \frac{\hbar}{(\omega_{k_1j_1}^a\omega_{k_2j_2}^o)^{1/2}}\langle A_{k_1j_1}^{o+}A_{k_1j_1}^o\rangle - 2\omega_{kj}^o\Delta_{kj}^o(\omega)\right]^2 + 4(\omega_{kj}^o)^2\Gamma_{kj}^{o2}(\omega)\right\}} . \quad (47)$$

Expressions (45) and (47) for sufficiently small values of $\Delta_{kj}(\omega)$ and $\Gamma_{kj}(\omega)$ compared with ω_{kj} give a number of peaks in the energy distribution of scattered neutrons. As a result of anharmonic interactions, the one-phonon peaks are broadened and their positions are shifted from the unperturbed phonon frequencies. The shape of the peaks is of Lorentzian form of width $\Gamma_{kj}(\omega)$ centered about the point $\pm(\omega_{kj}+\Delta_{kj})$. However, if the dependence of $\Gamma_{kj}(\omega)$ and $\Delta_{kj}(\omega)$ on ω_{kj} is considerable, there will be deviations in the line shape from that described by a Lorentzian function; in particular, asymmetry may appear in the energy distribution of the scattered neutrons. Equations (33), (41) and (34), (42) give expressions for the shifts and widths of phonon frequency which are frequency dependent and are obtained here as a direct consequence of the choice of the Hamiltonian. If the anharmonic forces are set equal to zero, the peak reduce to familiar two-delta functions, corresponding to neutron scattering with energy loss and energy gain respectively.

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