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Neutron Scattering in Impure Anharmonic Solids

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The Debye-Waller factor and the coherent inelastic differential scattering cross section for the one- and two-phonon band modes have been investigated for an anharmonic crystal containing randomly distributed substitutional impurities of low concentration. The method adopted in the evaluation is that of double time temperature dependent Green's functions. The quantities thus evaluated have been separated into diagonal and nondiagonal terms. The diagonal terms are further separated in three contributions, namely the contributions from defects, anharmonicity, and simultaneous involvement of impurity and anharmonicity. A new mode, the "impurity-anharmonicity interference" mode, is the salient featue of the theory, and arises due to the interaction between local and anharmonic fields.

1. INTRODUCTION

Experiments involving inelastic coherent and incoherent scattering of long-wavelength thermal neutrons (energies of 1 eV or less) provide one of the most powerful means for investigating the lattice vibrational modes of a crystal (Shull and Wollan, 1956; Kagan, 1962; Kascheev and Krivoglaz, 1961; Stewart and Brockhouse, 1958; Chernoplekov *et al.*, 1963; Haas *et al.*, 1963; Yamada and Shirane, 1969; Cowley, 1962, 1964; Shirane and Yamada, 1967, 1969). The incoherent scattering of thermal neutrons provides detailed information about the frequency spectrum of a crystal in which the scattering is isotropic and somewhat less directly in crystals of lower symmetry. The coherent scattering experiments in which the energy and momentum delivered to the crystal by thermal neutrons are measured, on the other hand, provide information about the phonon dispersion curves (Kascheev and Krivoglaz, 1961; Cowley, 1964; Shirane and Yamada, 1967; Van Hove, 1954; Kittel, 1963). The neutron scattering experiments are

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extremely helpful in the understanding of lattice dynamical properties of crystals (Marshall and Lovesey, 1971; Maradudin and Fein, 1962; Maradudin *et al.*, 1971; Weinstock, 1944).

The inelastic scattering of neutrons was first investigated theoretically by Weinstock (1944) for the one-phonon processes and the theory was subsequently extended by Sjolander (1954; Placzek and Van Hove, 1954) to multiphonon processes. Although the one-phonon processes are quite interesting and important from the standpoint of the information they yield about the dynamical properties of crystals, it appears to us that the multiphonon processes are not negligible and do not appear as a manageable correction to the theory. In the harmonic approximation (Maradudin *et al.*, 1971; Weinstock, 1944) the coherent one-phonon inelastic neutron scattering spectrum consists of a number of delta-function peaks centered around the central frequency. The number of peaks in the cross section is determined by the energy and momentum conservation conditions which govern the scattering process (Maradudin and Fein, 1962).

However, no crystal is perfectly harmonic and pure. The theory of the harmonic approximation applied to real crystals suffers severe drawbacks in investigating a large number of crystal properties. If we go beyond the harmonic approximation and retain cubic and quartic terms in the potential energy expansion of the crystal, we find that the infinitely long-lived normal modes of vibration (in the harmonic approximation) no longer remain exact eigenstates of the crystal Hamiltonian and their lifetimes are considerably reduced to a finite value. This happens as a result of the coupling of normal modes (noninteracting in the harmonic approximation), which, in turn, give rise to the phonon interaction and this situation essentially necessitates the solution of a many-body problem (Maradudin and Fein, 1962; Elliott and Maradudin, 1965; Shukla and Muller, 1980).

The presence of defects in a crystal, on the other hand, considerably modifies the frequency spectrum of the crystal, which drastically changes all the frequency-dependent properties of crystals (Kittel, 1963; Elliott and Maradudin, 1965; Tani, 1974; Sahu and Sharma, 1985). The presence of substitutional isotopic impurities in the host crystal leads to new phonon fields localized around the defect site which give rise to impurity modes. The impurity modes may be localized, gap, or resonance modes; the frequencies of the former two modes lie in the ranges forbidden in the host crystal and the frequencies of the later mode lie in the allowed frequency band of the host crystal. The local and gap modes are nonpropagating and the resonance modes are propagating modes. The changes in the phonon frequency spectrum caused as a result of substitutional impurities are substantially observed in a large number of neutron experiments (Kagan, 1962; Elliott and Maradudin, 1965; Lakatos and Krumhansl, 1968, 1969).

Extensive literature exists explaining the neutron scattering events (Maradudin and Fein, 1962; Weinstock, 1944; Sjölander, 1954; Placzek and Van Hove, 1954; Elliott and Maradudin, 1965; Tani, 1969, 1970, 1974; Tani and Tsuda, 1969; Takemura and Tani, 1971; Gairola and Semwal, 1977; Shukla and Muller, 1982; Sahu and Sharma, 1985; Acharya *et al.*, 1983; Kothari and Singwi, 1955; Maradudin, 1966). When the harmonic approximation is considered, some authors include the anharmonic effects and some consider the defect contributions only. One rarely comes across work in which impurity and anharmonicity effects are considered simultaneously.

In the present paper we study the problem of inelastic neutron scattering in an isotropically disordered anharmonic crystal. As a consequence of anharmonic forces and localized phonon interactions, the inelastic neutron scattering spectrum exhibit finite delta-function peaks and the frequencies are shifted relative to the frequencies of the harmonic approximation. The phonon frequency shifts and linewidths which are independent of temperature in the harmonic approximation are found to depend on temperature, impurity concentration, and force constant changes caused by substitutional impurities which show up in the experiments. Simultaneous inclusion of defects and anharmonicities results in a new type of phonon interaction, which we call the impurity-anharmonicity interaction. The phonons present in the anharmonic force field and those in the localized field of the impurity interact with each other, giving rise to this new scattering process. These processes are of significant magnitude and cannot be neglected in comparison to other events (Indu, 1990; Painuli *et al.*, 1990). In the case of inelastic neutron scattering we take into account the defects, anharmonicities, and their interfering events to investigate the differential cross section of neutrons and the Debye-Waller factor. The differential cross section is evaluated for the one- and two-phonon processes. The separation of these quantities into diagonal and nondiagonal parts gives the additional feature of the theory. The individual contributions of defects, anharmonicities, and of the simultaneous involvement of defects and anharmonicities are also treated.

The general formulation of the problem is given in Section 2, and the defect-induced anharmonic Hamiltonian is given in Section 3; this section also includes the evaluation of multiphonon double-time thermodynamic Green's functions. Section 4 is devoted to the derivation of the Debye-Waller factor and Section 5 gives the detailed evaluation of one- and two-phonon differential scattering cross sections; discussions are given in Section 6.

2. FORMULATION OF THE PROBLEM

The differential scattering cross section per unit solid angle $d\Omega$ and per unit interval of outgoing energy $d\varepsilon$ of scattered neutrons in the first

Born approximation for coherent scattering is given by (Van Hove, 1954)

$$\frac{d^2\sigma_{\text{coh}}}{d\Omega\, d\varepsilon} = \frac{a^2|K_f|}{\hbar|K_i|} S(\mathbf{K}, \omega) \tag{1}$$

where $\mathbf{K}_i$ and $\mathbf{K}_f$ are the wave vectors of the incident and scattered neutrons, respectively, $\hbar\mathbf{K} = \hbar(\mathbf{K}_i - \mathbf{K}_f)$ is the momentum transfer from the neutrons to the crystal, and $\hbar\omega = (\hbar/2m)(K_i^2 - K_f^2)$ is the energy transfer from neutrons to the crystal, a describes the scattering length of the nuclei, and (Maradudin and Fein, 1962; Shukla and Muller, 1980)

$$S(\mathbf{K}, \omega) = [\exp(-2W)/2\pi N] \sum_{ll'} \exp\{-i\mathbf{K}\cdot[\mathbf{x}(l) - \mathbf{x}(l')]\}$$
$$\times \int_{-\infty}^{\infty} dt \exp(i\omega t)$$
$$\times \exp\{\langle[\mathbf{K}\cdot\mathbf{u}(l, t)][\mathbf{K}\cdot\mathbf{u}(l', 0)]\rangle\} \tag{2}$$

Here, the position vector $x(l)$ of the mean position of the lth atom is related to the displacement $\mathbf{u}(l, t)$ of the lth atom from its mean position and the instantaneous position vector $\mathbf{R}(l)$ of the lth atom in the crystal by $\mathbf{R}(l) = \mathbf{x}(l) + \mathbf{u}(l, t)$. The quantity $2W$ is known as the Debye-Waller factor and is given by (Maradudin and Fein, 1962; Shukla and Muller, 1980)

$$2W = -(MN)^{-1} \sum_{kk'} \frac{[\mathbf{K}\cdot\mathbf{e}(k)][\mathbf{K}\cdot\mathbf{e}(k')]}{(\omega_k\omega_{k'})^{1/2}} \exp[2\pi i(\mathbf{k} + \mathbf{k}')\cdot\mathbf{x}(l)]$$
$$\times \lim_{\varepsilon\to 0} \int_{-\infty}^{\infty} n(\omega)\, d\omega\ \text{Im}\ G_{kk'}(\omega + i\varepsilon) \tag{3}$$

where $G_{kk'}(\omega)$ defines the Fourier-transformed double-time thermodynamic Green's function and $n(\omega)$ is Planck's distribution function.

The dynamical structure factor $S(\mathbf{K}, \omega)$ can be expanded in powers of the atomic displacement under the integral sign of equation (2) in the form

$$S(\mathbf{K}, \omega) = S_0(\mathbf{K}, \omega) + S_1(\mathbf{K}, \omega) + S_2(\mathbf{K}, \omega) + \cdots \tag{4}$$

where $S_0(\mathbf{K}, \omega)$ represent coherent elastic (zero-phonon process) scattering of neutrons and is given by (Maradudin and Fein, 1962)

$$S_0(\mathbf{K}, \omega) = N^2 e^{-2W} \delta(\omega)\, \Delta(\mathbf{K}/2\pi) \tag{5}$$

$S_0(\mathbf{K}, \omega)$ is elastic because of the factor $\delta(\omega)$ and is coherent because of $\Delta(\mathbf{K}/2\pi)$, which expresses the Bragg condition. The terms $S_1(\mathbf{K}, \omega)$ and $S_2(\boldsymbol{K}, \omega)$ describe the situation of present interest to us, coherent inelastic

scattering of neutrons by one-phonon and two-phonon processes, respectively, and are given by (Shukla and Muller, 1980)

$$S_n(\mathbf{K}, \omega) = (e^{-2W}/n!\,2\pi N) \sum_{ll'} \exp\{-i\mathbf{K}\cdot[\mathbf{x}(l) - \mathbf{x}(l')]\} \times \int_{-\infty}^{\infty} dt \exp(i\omega t) \times \langle [\mathbf{K}\cdot\mathbf{u}(l, t)][\mathbf{K}\cdot\mathbf{u}(l', t)]\rangle^n; \qquad n = 1, 2, \ldots \tag{6}$$

The quantity defined in angular brackets $\langle \cdots \rangle$ can be expressed in terms of the spectral distribution function, which is readily obtained from the Green's functions of the specific problem. Thus, $S(\mathbf{K}, \omega)$ includes all the solid-state physics of the problem and yields the multiphonon series via equation (6). We shall now investigate in detail the nature of the dynamical structure factor $S(\mathbf{K}, \omega)$ for an impure anharmonic crystal.

3. CRYSTAL HAMILTONIAN AND GREEN'S FUNCTIONS

Let us consider a three-dimensional Bravais crystal in which $N-n$ lattice sites are occupied by atoms of the host crystal with the mass of each atom M, and the rest of the n sites are occupied by isotropically substituted impurity atoms each of mass M'. The impurity concentration is kept very low so that the impurity–impurity interaction can be ignored. The Hamiltonian of this problem can be written as

$$H = H_0 + H_A + H_D \tag{7}$$

where H_0, H_A, and H_D describe the unperturbed, anharmonic, and defect-induced parts of the Hamiltonian, respectively, and can be expressed in second-quantized form as

$$H_0 = (1/4) \sum_k \hbar\omega_k (A_k^* A_k + B_k^* B_k) \tag{8a}$$

$$H_A = \hbar \sum_{S\geq 3} \sum_{k_1, k_2, k_3} V_s(\mathbf{k}_1, \mathbf{k}_2, \ldots, \mathbf{k}_s) A_{k_1} A_{k_2}, \ldots, A_{k_s} \tag{8b}$$

$$H_D = -\hbar \sum_{k_1, k_2} [C(\mathbf{k}_1, \mathbf{k}_2) B_{k_1} B_{k_2} - D(\mathbf{k}_1, \mathbf{k}_2) A_{k_1} A_{k_2}] \tag{8c}$$

In equation (8b), $V_s(k_1, k_2, \ldots, k_s)$ is the anharmonic coupling coefficient, the order of which is given by the subscript s, and $C(\mathbf{k}_1, \mathbf{k}_2)$ and $D(k_1, k_2)$ are the mass difference and force constant change parameters, respectively. These parameters are well defined (Sahu and Sharma, 1985; Indu, 1990; Painuli, 1990) and need no further description.

The double-time temperature-dependent Green's function

$$G_{kk'}(t - t') = \langle\langle A_k(t); A_{k'}^*(t)\rangle\rangle \tag{9}$$

can be obtained in the form

$$G_{kk'}(\omega) = \omega_k \eta_{kk'} / \pi[\omega^2 - \tilde{\omega}_k^2 - 2\omega_k P(kk', \omega)] \tag{10}$$

We have used the quantum dynamics of second-quantized operators and Dyson's equation method to obtain equation (10); the details of this method are well described elsewhere (Indu, 1990; Painuli *et al.*, 1990). The notation is defined as

$$\eta_{kk'} = \delta_{kk'} + 4C(\mathbf{k}, -\mathbf{k})/\omega_k \tag{11}$$

$$\begin{aligned}\tilde{\omega}_k^2 = \omega_k^2 &+ (\omega_k/2\pi)\{\langle[F_k(t), B_{k'}^*]^0\rangle + (\omega/\omega_k)\langle[F_k(t), A_{k'}^*]^0\rangle \\ &+ (4/\omega_k)\sum_{k_1} C(-\mathbf{k}, \mathbf{k}_1)\langle[F_k(t), B_{k'}^*]^0\rangle \\ &+ 8(\omega^2 - \omega_k^2)C(-\mathbf{k}, \mathbf{k}')/\omega_k\}_{t=t'}\end{aligned} \tag{12}$$

and

$$P(kk', \omega) = (2\pi)^{-1}\langle\langle F_k(t); F_{k'}^*(t')\rangle\rangle_\omega \tag{13}$$

with

$$F_k(t) = F_k^{(1)}(t) + F_k^{(2)}(t) + F_k^{(3)}(t) + F_k^{(4)}(t) \tag{14}$$

$$F_k^{(1)}(t) = 4\pi \sum_{k_1} [D(-\mathbf{k}, \mathbf{k}_1) + (\omega_{k_1}/\omega_k)C(-\mathbf{k}, \mathbf{k}_1)]A_{k_1} \tag{15a}$$

$$F_k^{(2)}(t) = 16\pi \sum_{k_1, k_2} [C(-\mathbf{k}, \mathbf{k}_1)D(-\mathbf{k}_2, \mathbf{k}_1)/\omega_k]A_{k_1} \tag{15b}$$

$$\begin{aligned}F_k^{(3)}(t) = 2\pi \sum_{s\geq 3} \sum_{k_1, k_2 \cdots k_{s-1}} &sV_s(\mathbf{k}_1, \mathbf{k}_2 \cdots \mathbf{k}_{s-1}, -\mathbf{k}) \\ &\times A_{k_1} A_{k_2} \cdots A_{k_{s-1}}\end{aligned} \tag{15c}$$

$$F_k^{(4)}(t) = 4 \sum_{k_1} [C(-\mathbf{k}, \mathbf{k}_1)/\omega_k] F_{k_1}^{(3)}(t) \tag{15d}$$

$F_{k_1}^{(3)}(t)$ can be obtained from $F_{k_1}^{(3)}(t)$ after replacing $\mathbf{k}$ by $\mathbf{k}_1$.

The phonon excitation spectrum described by $P(kk', \omega)$ can be obtained with the help of the zeroth-order renormalized Hamiltonian

$$H_{\text{ren}}^{(0)} = (\hbar/4) \sum_k [(\tilde{\omega}_k^2/\omega_k) A_k^* A_k + \omega_k B_k^* B_k] \tag{16}$$

This Hamiltonian is extremely useful for obtaining the multiphonon Green's function appearing in $P(kk', \omega)$ $[\equiv P(k, \omega)]$ with the help of the equation of motion method (Painuli *et al.*, 1990). $P(k, \omega)$ can also be expressed in the most widely used form as

$$P(k, \omega + i\varepsilon) = \Delta_k(\omega) - i\Gamma_k(\omega); \qquad \varepsilon \to 0^+ \tag{17}$$

where the real part of $P(k, \omega)$ (phonon self-energy) $\Delta_k(\omega)$ is the shift in the phonon frequency of the perturbed mode and the imaginary part $\Gamma_k(\omega)$ is the phonon frequency linewidth at the half-maximum of the phonon frequency peak. The new form of $G_{kk'}(\omega)$ can be expressed as

$$G_{kk'}(\omega) = \omega_k \eta_{kk'} / \pi[\omega^2 - \tilde{\tilde{\omega}}_k^2 + 2i\omega_k \Gamma_k(\omega)] \tag{18}$$

where $\tilde{\tilde{\omega}}_k$ is the perturbed mode frequency and can be expressed in terms of the renormalized mode frequency $\tilde{\omega}_k$ as

$$\tilde{\tilde{\omega}}_k^2 = \tilde{\omega}_k^2 + 2\omega_k \Delta_k(\omega) \tag{19}$$

The phonon frequency shifts and widths can be obtained as (Indu, 1990)

$$\Delta_k(\omega) = \Delta_k^D(\omega) + \Delta_k^A(\omega) + \Delta_k^{AD}(\omega) \tag{20}$$

and

$$\Gamma_k(\omega) = \Gamma_k^D(\omega) + \Gamma_k^A(\omega) + \Gamma_k^{AD}(\omega) \tag{21}$$

where the superscripts D, A, and AD stand for the contributions due to defects, anharmonicities, and impurity-anharmonicity interactions, respectively. The various terms in equations (20) and (21) are readily obtainable as (Indu, 1990)

$$\Delta_k^D(\omega) = 8 \sum_{k_1} R(-\mathbf{k}, \mathbf{k}_1) R^*(-\mathbf{k}, \mathbf{k}_1) \omega_{k_1} / (\omega^2 - \tilde{\omega}_{k_1}^2) \tag{22a}$$

$$\Delta_k^A(\omega) = \Delta_k^{3A}(\omega) + \Delta_k^{4A}(\omega) \tag{22b}$$

$$\Delta_k^{AD}(\omega) = 16 \sum_{k_1} |C(-\mathbf{k}, \mathbf{k}_1)|^2 \omega_k^{-2} \Delta_{k_1}^A(\omega) \tag{22c}$$

$$\Delta_k^{3A}(\omega) = 18 \sum_{k_1, k_2} |V_3(\mathbf{k}_1, \mathbf{k}_2, -\mathbf{k})|^2 \eta_1 \times [S_{+1} \omega_{+\alpha} (\omega^2 - \omega_{+\alpha}^2)^{-1} + S_{-1} \omega_{-\alpha} (\omega^2 - \omega_{-\alpha}^2)^{-1}] \tag{22d}$$

$$\Delta_k^{4A}(\omega) = 48 \sum_{k_1, k_2, k_3} |V_4(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3, -\mathbf{k})|^2 \eta_2 \times [S_{+2} \omega_{+\beta} (\omega^2 - \omega_{+\beta}^2)^{-1} + 3 S_{-2} \omega_{-\beta} (\omega^2 - \omega_{-\beta}^2)^{-1}] \tag{22e}$$

$$\Gamma_k^D(\omega) = 8\pi\varepsilon(\omega) \sum_{k_1} R(-\mathbf{k}, \mathbf{k}_1) R^*(-\mathbf{k}, \mathbf{k}_1) \omega_{k_1} \delta(\omega^2 - \tilde{\omega}_{k_1}^2) \tag{23a}$$

$$\Gamma_k^A(\omega) = \Gamma_k^{3A}(\omega) + \Gamma_k^{4A}(\omega) \tag{23b}$$

$$\Gamma_k^{AD}(\omega) = 16 \sum_{k_1} |C(-\mathbf{k}, \mathbf{k}_1)|^2 \omega_k^{-2} \Gamma_{k_1}^A(\omega) \tag{23c}$$

$$\Gamma_k^{3A}(\omega) = 18\pi\varepsilon(\omega) \sum_{k_1, k_2} |V_3(\mathbf{k}_1, \mathbf{k}_2, -\mathbf{k})|^2 \eta_1 \times [S_{+1} \omega_{+\alpha} \delta(\omega^2 - \omega_{+\alpha}^2) + S_{-1} \omega_{-\alpha} \delta(\omega^2 - \omega_{-\alpha}^2)] \tag{23d}$$

$$\Gamma_k^{4A}(\omega) = 48\pi\varepsilon(\omega) \sum_{k_1, k_2, k_3} |V_4(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3, -\mathbf{k})|^2 \eta_2 \times [S_{+2} \omega_{+\beta} \delta(\omega^2 - \omega_{+\beta}^2) + 3 S_{-2} \omega_{-\beta} \delta(\omega^2 - \omega_{-\beta}^2)] \tag{24}$$

with

$$R(\mathbf{k}_1, \mathbf{k}_2) = (\omega_{k_1}/\omega_{k_2}) C(\mathbf{k}_1, \mathbf{k}_2) + D(\mathbf{k}_1, \mathbf{k}_2) + 4 \sum_{k_2'} C(-\mathbf{k}_1, \mathbf{k}_2') D(-\mathbf{k}_2', \mathbf{k}_2)/\omega_{k_1} \tag{25}$$

$$\eta_1 = \omega_{k_1}\omega_{k_2}/\tilde{\omega}_{k_1}\tilde{\omega}_{k_2}; \qquad \eta_2 = \omega_{k_1}\omega_{k_2}\omega_{k_3}/\tilde{\omega}_{k_1}\tilde{\omega}_{k_2}\tilde{\omega}_{k_3} \tag{26a}$$

$$\omega_{\pm\alpha} = \tilde{\omega}_{k_1} \pm \tilde{\omega}_{k_2}; \qquad \omega_{\pm\beta} = \tilde{\omega}_{k_1} \pm \tilde{\omega}_{k_2} \pm \tilde{\omega}_{k_3} \tag{26b}$$

$$S_{\pm 1} = n_{k_2} \pm n_{k_1}; \qquad S_{\pm 2} = 1 \pm n_{k_1} n_{k_2} + n_{k_2} n_{k_3} \pm n_{k_3} n_{k_1} \tag{27}$$

$$\begin{aligned} \varepsilon(\omega) &= 1 \qquad \text{for} \quad \omega > 0 \\ &= -1 \qquad \text{for} \quad \omega < 0 \end{aligned} \tag{28}$$

and

$$n_k = \coth(\beta\hbar\omega_k/2)$$

The correlation function $\langle A_k(t) A_k^*(0)\rangle$ is related to the Green's function in the form

$$\langle A_k(t) A_k^*(0)\rangle = \int_{-\infty}^{\infty} \exp(\beta\hbar\omega)\, J_{kk'}(\omega) \exp(-i\omega t)\, d\omega \tag{29}$$

where the spectral function $J_{kk'}(\omega)$ is given by

$$J_{kk'}(\omega) = \lim_{\varepsilon \to 0} n(\omega)[G_{kk'}(\omega + i\varepsilon) - G_{kk'}(\omega - i\varepsilon)] \tag{30}$$

With this evaluation the basic requirements to investigate the inelastic neutron scattering are in hand.

4. THE DEBYE–WALLER FACTOR

Making use of equation (18) in equation (3), we can obtain the exponent of the Debye–Waller factor in the form

$$2W = (MN)^{-1} \sum_{k,k'} [\mathbf{K}\cdot\mathbf{e}(\mathbf{k})][\mathbf{K}\cdot\mathbf{e}(\mathbf{k}')] \int_{-\infty}^{\infty} 2n(\omega)\eta_{kk'}\omega_k\Gamma_k(\omega) \times [(\omega^2 - \tilde{\tilde{\omega}}_k^2)^2 + 4\omega_k^2\Gamma_k^2(\omega)]^{-1}\, d\omega \tag{31}$$

If the conditions $\partial\Gamma_k(\omega)/\partial\omega \ll 1$ and $\partial\Delta_k(\omega)/\partial\omega \ll 1$ are satisfied under a reasonable limiting procedure, the integrand of equation (31) has a steep

maximum at the frequencies $\omega = \tilde{\tilde{\omega}}_k$. If we further assume that in the neighborhood of $\omega = \tilde{\tilde{\omega}}_k$, $\Gamma_k(\omega)$ varies slowly with ω, i.e., $\Gamma_k(\omega) \simeq \Gamma_k(\tilde{\tilde{\omega}}_k)$, then the integrand shows a Lorentzian line-shape distribution with a maximum at $\omega \simeq \tilde{\tilde{\omega}}_k$. If the variation of widths and shifts with frequency is taken into account, then, apart from the nature of combination bands arising as a result of anharmonic interactions, there will appear new sharp delta-function peaks and the band shape will be altered (Thomson, 1963; Mavroyannis and Pathak, 1969). For values of ω in the immediate vicinity of $\tilde{\tilde{\omega}}_k$, the $\Gamma_k^2(\omega)$ will have a vanishingly small contribution in the denominator of the integrand of equation (31) and hence it takes the form

$$2W = (MN)^{-1} \sum_{k,k'} [\mathbf{K} \cdot e(\mathbf{k})][\mathbf{K} \cdot e(\mathbf{k}')] \int_{-\infty}^{\infty} 2\eta_{kk'} \omega_k n(\omega) \times \Gamma_k(\omega)(\omega^2 - \tilde{\tilde{\omega}}_k^2)^{-2} \, d\omega \tag{32}$$

Using the orthogonality relation $\sum_j e_\alpha(kj) e_\beta(kj) = \delta_{\alpha\beta}$ and the nature of $\eta_{kk'}$, we can separate the exponant of the Debye-Waller factor into diagonal $[2W]_d$ and nondiagonal $[2W]_{nd}$ parts,

$$2W = [2W]_d + [2W]_{nd} \tag{33a}$$

where

$$[2W]_d = (MN)^{-1} \sum_k K^2 \int_{-\infty}^{\infty} 2\omega_k n(\omega) \Gamma_k(\omega)(\omega^2 - \tilde{\tilde{\omega}}_k^2)^{-2} \, d\omega \tag{33b}$$

$$[2W]_{nd} = 8(MN)^{-1} \sum_{k \neq k'} [\mathbf{K} \cdot e(\mathbf{k})][\mathbf{K} \cdot e(\mathbf{k}')] \int_{-\infty}^{\infty} C(\mathbf{k}, -\mathbf{k}) n(\omega) \times \Gamma_k(\omega)(\omega^2 - \tilde{\tilde{\omega}}_k^2)^{-2} \, d\omega \tag{33c}$$

The diagonal part can be further separated into three terms arising due to the defects, anharmonicities, and simultaneous involvement of defects and anharmonicities. This can be easily obtained as the direct consequence of $\Gamma_k(\omega)$ in the light of equation (21). The nondiagonal contribution chiefly depends on the mass change parameter and drops to zero in the absence of an impurity. This can also be understood in the form of a phonon field interaction arising due to the localized fields developed as a direct consequence of mass changes at the impurity sites with anharmonic and local phonons. After some complicated algebra the diagonal part of Debye-Waller factor can be obtained in the form

$$[2W]_d = [2W]_d^D + [2W]_d^{3A} + [2W]_d^{4A} + [2W]_d^{AD} \tag{34}$$

where

$$[2W]_{\mathrm{d}}^{D} = 16\pi K^2 \varepsilon(\tilde{\omega}_{k_1}^2)(MN)^{-1} \sum_{k_1} \omega_k \omega_{k_1} R(-\mathbf{k}, \mathbf{k}_1) R^*(-\mathbf{k}, \mathbf{k}_1) \times n(\tilde{\omega}_{k_1})(\tilde{\omega}_{k_1}^2 - \tilde{\tilde{\omega}}_k^2)^{-2} \tag{35a}$$

$$[2W]_{\mathrm{d}}^{3A} = 36\pi\omega_k K^2 (MN)^{-1} \sum_{k_1 k_2} |V_3(\mathbf{k}_1, \mathbf{k}_2, -\mathbf{k})|^2 \eta_1 \times [\varepsilon(\omega_{+\alpha}^2) S_{+1} \omega_{+\alpha} n(\omega_{+\alpha})(\omega_{+\alpha}^2 - \tilde{\tilde{\omega}}_k^2)^{-2} + \varepsilon(\omega_{-\alpha}^2) S_{-1} \omega_{-\alpha} n(\omega_{-\alpha})(\omega_{-\alpha}^2 - \tilde{\tilde{\omega}}_k^2)^{-2}] \tag{35b}$$

$$[2W]_{\mathrm{d}}^{4A} = 96\pi\omega_k K^2 (MN)^{-1} \sum_{k_1 k_2 k_3} |V_4(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3, -\mathbf{k})|^2 \eta_2 \times [\varepsilon(\omega_{+\beta}^2) S_{+2} \omega_{+\beta} n(\omega_{+\beta})(\omega_{+\beta}^2 - \tilde{\tilde{\omega}}_k^2)^{-2} + 3\varepsilon(\omega_{-\beta}^2) S_{-2} \omega_{-\beta} n(\omega_{-\beta})(\omega_{-\beta}^2 - \tilde{\tilde{\omega}}_k^2)^{-2}] \tag{35c}$$

and

$$[2W]_{\mathrm{d}}^{AD} = 16 \sum_{k_1} |C(-\mathbf{k}, \mathbf{k}_1)|^2 \omega_k^{-2} [2W]_{k-k_1}^{A} \tag{35d}$$

Here $[2W]_{k-k_1}^{A}$ stands for the value of $[2W]_{\mathrm{d}}^{A}$ when all the k values are replaced by k_1. It is obvious from the above expressions that the inclusion of defects makes it dependent on impurity concentrations and force constant changes, and on the other hand it becomes temperature dependent due to the inclusion of anharmonicities.

5. DIFFERENTIAL SCATTERING CROSS SECTION

The scattering function $S_0(\mathbf{k}, \omega)$ for all processes in which no energy is exchanged between the neutrons and the crystal is given by all terms in the expansion of the terms appearing in the angular bracket of equation (2) which are independent of $\exp(\pm i\omega t)$. The scattering term $S_1(\mathbf{k}, \omega)$ reflects the emission or absorption of one phonon, while $S_2(\mathbf{k}, \omega)$ arises due to the creation or destruction of two phonons. The term $S_1(\mathbf{k}, \omega)$ gives the dominant contribution to the differential cross section for temperature not much higher than the Debye temperature, and $S_2(\mathbf{k}, \omega)$ gives the higher-order correction to the differential scattering cross section.

5.1. Scattering of Neutrons by One-Phonon Processes

The evaluation of $S_1(\mathbf{k}, \omega)$ is straightforward from equation (6). For $n = 1$, the normal coordinate substitution in second-quantized notation

yields

$$S_1(\mathbf{k}, \omega) = (\hbar/2M) \exp(-2W) \sum_{kk'} \Delta(\mathbf{K}-\mathbf{k})\Delta(\mathbf{K}-\mathbf{k}')(\omega_{k_1}\omega_{k_2})^{-1/2}$$
$$\times [\mathbf{K}\cdot e(\mathbf{k})][\mathbf{K}\cdot e(\mathbf{k}')] \int_{-\infty}^{\infty} \langle A_k(t) A_{k'}^*(0)\rangle$$
$$\times \exp(-i\omega t)\, dt \tag{36}$$

Substituting the value of the correlation function from equations (29), (30), and (18) in equation (36), we obtain

$$S_1(\mathbf{k}, \omega) = \exp(-2W)[S_{1,\mathrm{d}}(\mathbf{K}, \omega) + S_{1,\mathrm{nd}}(\mathbf{K}, \omega)] \tag{37}$$

where the diagonal and nondiagonal parts, respectively, are obtained as

$$S_{1,\mathrm{d}}(\mathbf{K}, \omega) = \frac{2\hbar}{M} \sum_k |\Delta(\mathbf{K}-\mathbf{k})|^2 K^2 \omega_k$$
$$\times \int_{-\infty}^{\infty} \frac{e^{\beta\hbar\omega}\Gamma_k(\omega)\, d\omega}{(e^{\beta\hbar\omega}-1)[(\omega^2-\tilde{\omega}_k^2)^2+4\omega_k^2\Gamma_k^2(\omega)]} \tag{38a}$$

and

$$S_{1,\mathrm{nd}}(\mathbf{K}, \omega) = \frac{2\hbar}{M} \sum_{k\neq k'} \Delta(\mathbf{K}-\mathbf{k})\Delta(\mathbf{K}-\mathbf{k}')[\mathbf{K}\cdot e(\mathbf{k})][\mathbf{K}\cdot e(\mathbf{k}')]$$
$$\times \frac{\omega_k}{(\omega_{k_1}\omega_{k_2})^{1/2}} \int_{-\infty}^{\infty} \frac{e^{\beta\hbar\omega} C(\mathbf{k}, -\mathbf{k})\Gamma_k(\omega)\, d\omega}{(e^{\beta\hbar\omega}-1)[(\omega^2-\tilde{\omega}_k^2)^2+4\omega_k^2\Gamma_k^2(\omega)]} \tag{38b}$$

The diagonal part $S_{1,\mathrm{d}}(\mathbf{K}, \omega)$ gives the principal contribution to $S(\mathbf{K}, \omega)$ and the nondiagonal part $S_{1,\mathrm{nd}}(\mathbf{K}, \omega)$ gives small corrections (however, not negligible) due to the interference of anharmonic force constants and defect parameters and the mass change parameter $C(\mathbf{k}_1, \mathbf{k}_2)$. The diagonal part can be studied in more detail. In light of equation (21), it can be obtained in terms of individual contributions coming from defects, anharmonic effects, and from simultaneous involvement of anharmonic force and localized fields. The appreciable contribution of defects and anharmonicities distorts the Lorentzian line-shape distribution of $S(\mathbf{K}, \omega)$ and for a small value of phonon frequency widths we can reexamine $S_{1,\mathrm{d}}(\mathbf{K}, \omega)$ in the immediate vicinity of $\omega = \tilde{\omega}_k$, which yields

$$S_{1,\mathrm{d}}(\mathbf{K}, \omega) = S_{1,\mathrm{d}}^{D}(\mathbf{K}, \omega) + S_{1,\mathrm{d}}^{A}(\mathbf{K}, \omega) + S_{1,\mathrm{d}}^{AD}(\mathbf{K}, \omega) \tag{39}$$

where

$$S_{1,\mathrm{d}}^{\alpha}(\mathbf{K}, \omega) = (2\hbar/M) \sum_k |\Delta(\mathbf{K}-\mathbf{k})|^2 K^2 \omega_k \int_{-\infty}^{\infty} [n(\omega)+1]\Gamma_k^{\alpha}(\omega)$$
$$\times (\omega^2-\tilde{\omega}_k^2)^{-2}\, d\omega \qquad (\alpha = D, A, AD) \tag{40}$$

Equation (38) shows that the scattering function is not only a two-delta-function distribution like the harmonic distribution, but has a Lorentzian line-shape distribution. Equation (40) shows that a large number of impurity-concentration- and temperature-dependent delta-function peaks appear in the band mode distribution at the frequencies $\pm\omega_{\pm\alpha}$, $\pm\omega_{\pm\beta}$. The one-phonon differential scattering cross section can now be given in the form

$$\frac{d^2\sigma_{\text{coh}}}{d\Omega\, d\varepsilon} = \int_{-\infty}^{\infty} \sum_{k} A_k(\mathbf{K}, \omega)\Gamma_k^{\alpha}(\omega)(\omega^2 - \tilde{\omega}_k^2)^{-2}\, d\omega + \int_{-\infty}^{\infty} \sum_{k \neq k'} A_{kk'}(\mathbf{K}, \omega)\Gamma_k^{\alpha}(\omega)(\omega^2 - \tilde{\omega}_k^2)^{-2}\, d\omega \tag{41}$$

where

$$A_k(\mathbf{K}, \omega) = (2a^2/M)|K_f/K_i|\,|\Delta(\mathbf{K}-\mathbf{k})|^2 K^2 \omega_k \times [n(\omega)+1]\exp(-2W) \tag{42a}$$

$$A_{kk'}(\mathbf{K}, \omega) = (2a^2/M)|K_f/K_i|\Delta(\mathbf{K}-\mathbf{k})\Delta(\mathbf{K}-\mathbf{k}') \times [\mathbf{K}\cdot e(\mathbf{k})][\mathbf{K}\cdot e(\mathbf{k}')] \times \omega_k(\omega_{k_1}\omega_{k_2})^{-1/2}[n(\omega)+1]\exp(-2W) \tag{42b}$$

The impurity concentration, frequency, and temperature dependences of the phonon linewidths have been obtained in the form (Indu, 1990)

$$\Gamma_k^{D}(\omega) = A_1 c(1-c)\omega^4 + A_2 c\omega^2 \tag{43a}$$

$$\Gamma_k^{3A}(\omega) = B_1\omega^2 T; \qquad \Gamma_k^{4A}(\omega) = B_2\omega^2 T^2 \tag{43b}$$

$$\Gamma_k^{3AD}(\omega) = b_1 c(1-c)\omega^4 T; \qquad \Gamma_k^{4AD}(\omega) = b_2 c(1-c)\omega^4 T^2 \tag{43c}$$

where A_1, A_2, B_1, B_2, b_1, and b_2 are some constants and c is the impurity concentration. Use of equation (43) in equations (38), (39), and (41) clearly shows that the one-phonon differential cross section depends on the temperature linearly and nonlinearly and varies similarly with impurity concentration.

5.2. Two-Phonon Differential Cross Section

The two-phonon scattering factor can be obtained from equation (6) for $n=2$ by the usual simplifications in the form

$$S_2(\mathbf{K}, \omega) = (N/4\pi)(\hbar/2NM)^2\exp(-2W) \sum_{k_1,k_2,k_3,k_4} \Delta(-\mathbf{k}+\mathbf{k}_1+\mathbf{k}_3) \times \Delta(\mathbf{K}-\mathbf{k}_2-\mathbf{k}_4)[\mathbf{K}\cdot e(\mathbf{k}_1)][\mathbf{K}\cdot e(\mathbf{k}_2)] \times [\mathbf{K}\cdot e(\mathbf{k}_3)][\mathbf{k}\cdot e(\mathbf{k}_4)] \times (\omega_{k_1}\omega_{k_2}\omega_{k_3}\omega_{k_4})^{-1/2} \int_{-\infty}^{\infty} dt\exp(i\omega t) \times \langle A_{k_1}(t)A_{k_2}(t)A_{k_3}(0)A_{k_4}(0)\rangle \tag{44}$$

Thus, we notice that the problem of evaluating the two-phonon differential scattering cross section of an impure anharmonic solid is reduced to that of the calculation of a two-time correlation function on a suitable model Hamiltonian. The correlation function can be evaluated by factorizing it according to a suitable decoupling scheme (Gorkov, 1958; Zubarev, 1960). Thus, for the average of a four-operator product, we write

$$\begin{aligned}&\langle A_{k_1}(t)A_{k_2}(t)A_{k_3}(0)A_{k_4}(0)\rangle\\&\quad=\langle A_{k_1}(t)A_{k_3}(0)\rangle\langle A_{k_2}(t)A_{k_4}(0)\rangle\\&\qquad+\langle A_{k_1}(t)A_{k_4}(0)\rangle\langle A_{k_2}(t)A_{k_3}(0)\rangle\end{aligned}\tag{45}$$

In writing equation (45), we have neglected the correlation of operators with the same time argument, because they do not contribute in the energy transfer relating the present problem. After some complicated algebra and use of equation (45), we can obtain a considerably simplified form of equation (44) as

$$\begin{aligned}S_2(\mathbf{k},\omega)&=(N/2\pi)(\hbar/2NM)^2\exp(-2W)\sum_{k_1,k_2}|\Delta(-\mathbf{k}+\mathbf{k}_1+\mathbf{k}_2)|\\&\times\{[\mathbf{K}\cdot e(\mathbf{k}_1)][\mathbf{K}\cdot e(\mathbf{k}_2)]\}^2(\omega_{k_1}\omega_{k_2})^{-1}I(\omega)\end{aligned}\tag{46}$$

where

$$I(\omega)=\int_{-\infty}^{\infty}dt\,\exp(i\omega t)\,(\langle A_{k_1}A_{k_3}^*\rangle\langle A_{k_2}A_{k_4}^*\rangle)\tag{47}$$

Evaluation of the correlation functions present in $I(\omega)$ yields

$$\begin{aligned}I(\omega)&=(4\omega_{k_1}\omega_{k_2})^2\xi_{k_1k_2}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}d\omega_1\,d\omega_2\\&\times\frac{N(\omega_1)N(\omega_2)\Gamma_{k_1}(\omega_1)\Gamma_{k_2}(\omega_2)}{[(\omega_1^2-\tilde{\tilde{\omega}}_{k_1}^2)^2+4\omega_{k_1}^2\Gamma_{k_1}^2(\omega_1)][(\omega_2^2-\tilde{\tilde{\omega}}_{k_2}^2)^2+4\omega_{k_2}^2\Gamma_{k_2}^2(\omega_2)]}\end{aligned}\tag{48}$$

where $\Gamma_{k_i}(\omega_i)$ and $\tilde{\tilde{\omega}}_{k_i}$ are obtainable from equations (19) and (21), respectively, on appropriate replacement of indices and

$$\begin{aligned}\xi_{k_1k_2}&=[1+8C(\mathbf{k}_1,-\mathbf{k}_1)/\omega_{k_1}]\delta_{k_1k_2}+16C(\mathbf{k}_1,-\mathbf{k}_1)\\&\times C(\mathbf{k}_2,-\mathbf{k}_2)/\omega_{k_1}\omega_{k_2}\end{aligned}\tag{49a}$$

and

$$N(\omega_i)=\exp(\beta\hbar\omega_i)/[\exp(\beta\hbar\omega_i)-1];\qquad i=1,2\tag{49b}$$

The exact evaluation of the double integral (48) is not possible. We can evaluate this integral for the nonperturbative approach in which the

integrand is peaked around the immediate vicinity of frequencies $\tilde{\omega}_{k_1}$ and $\tilde{\omega}_{k_2}$ for small values of the phonon linewidth (Shukla and Muller, 1980; Pathak, 1965; Shukla, 1980). Hence,

$$I(\omega) = (4\omega_{k_1}\omega_{k_2})^2 \xi_{k_1 k_2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} d\omega_1 \, d\omega_2 \, N(\omega_1) N(\omega_2) \times \Gamma_{k_1}(\omega_1)\Gamma_{k_2}(\omega_2)[(\omega_1^2 - \tilde{\omega}_{k_1}^2)(\omega_2^2 - \tilde{\omega}_{k_2}^2)]^2 \tag{50}$$

However, a next permissible approximation can be used. The integrand may be peaked as a delta-function peak, but at the same time the contribution of widths at $\tilde{\omega}_{k_i}$ cannot be neglected in the denominator of the integrand. Thus,

$$I(\omega) = \xi_{k_1 k_2} N(\tilde{\omega}_{k_1}) N(\tilde{\omega}_{k_2}) \Gamma_{k_1}^{-1}(\omega_{k_1}) \Gamma_{k_2}^{-1}(\omega_{k_2}) \tag{51}$$

Substitution of equation (51) in equation (46) yields

$$S_2(\mathbf{K}, \omega) = \exp(-2W)\,[S_{2,d}(\mathbf{K}, \omega) + S_{2,\mathrm{nd}}(\mathbf{K}, \omega)] \tag{52}$$

where

$$S_{2,\mathrm{d}}(\mathbf{K}, \omega) = (N/2\pi)(\hbar K^2/2NM)^2 \sum_{k_1} |\Delta(-\mathbf{K} + \mathbf{K}_1)|[1 + 8C(\mathbf{k}_1, -\mathbf{k}_1)/\omega_{k_1}] \times [N(\tilde{\omega}_{k_1})/\omega_{k_1}\Gamma_{k_1}(\tilde{\omega}_{k_1})]^2 \tag{53a}$$

$$S_{2,\mathrm{nd}}(\mathbf{K}, \omega) = (2N/\pi)(\hbar/NM)^2 \sum_{k_1 \neq k_2} |\Delta(-\mathbf{k} + \mathbf{k}_1 + \mathbf{k}_2)|\{[\mathbf{K} \cdot e(\mathbf{k}_1)] \times [\mathbf{K} \cdot e(\mathbf{k}_2)]\}^2 (\omega_{k_1}\omega_{k_2})^{-2} C(\mathbf{k}_1, -\mathbf{k}_1) C(\mathbf{k}_2, -\mathbf{k}_2) \times N(\tilde{\omega}_{k_1}) N(\tilde{\omega}_{k_2}) / \Gamma_{k_1}(\tilde{\omega}_{k_1}) \Gamma_{k_2}(\tilde{\omega}_{k_2}) \tag{53b}$$

The nondiagonal contribution $S_{2,\mathrm{nd}}(\mathbf{K}, \omega)$ contributes only in the presence of impurities and arises due to the interaction of mass change parameters with various other terms. The diagonal term also shows a strong dependence on mass change parameter, but for a pure crystal this contributes as

$$S_{2,\mathrm{d}}(\mathbf{K}, \omega) = (N/2\pi)(\hbar k^2/2NM)^2 \times \sum_{k_1} |\Delta(-\mathbf{k} + \mathbf{k}_1)|[N(\tilde{\omega}_{k1})/\omega_{k_1}\Gamma_{k_1}^{A}(\tilde{\omega}_{k_1})]^2 \tag{54}$$

6. DISCUSSION

We have investigated the contribution of defects and anharmonicities in a crystal to the exponent of the Debye–Waller factor and the one- and two-phonon differential inelastic neutron scattering cross sections. For the first time it has been established that quite a significant contribution of the

impurity-anharmonicity interactions appears to describe these physical properties. As the defects alter the crystal properties drastically, the intensity of the interactions of localized fields with the anharmonic fields is quite significant in comparison to anharmonic effects. It is found that these quantities can be separated into diagonal and nondiagonal terms. The diagonal terms are found to give principal contributions, while the nondiagonal terms give a significant contribution but have a smaller magnitude and contribute only in the presence of impurities. The perturbed mode frequencies are found to depend on temperature, impurity concentration, and force constant changes. The renormalized mode frequency $\tilde{\omega}_k$ describes the perturbed mode frequency and can be evaluated from equation (12) as

$$\tilde{\omega}_k^2 = \omega_k^2 + 4\omega_k[\omega_k^A + \omega_k^D + (\omega_k^{AD})^2/\omega_k] \tag{55}$$

with

$$\omega_k^A \cong -18 \sum_{k_1} V_3(\mathbf{k}', \mathbf{k}', -\mathbf{k})\, V_3(-\mathbf{k}', \mathbf{k}_1, -\mathbf{k}_1) n_k \omega_k^{-1} \tag{56a}$$

$$\omega_k^D \cong C(\mathbf{k}_1, -\mathbf{k}) + D(\mathbf{k}_1, -\mathbf{k}) + 4 \sum_{k_1} C(\mathbf{k}_1, -\mathbf{k}) D(\mathbf{k}_1, -\mathbf{k}) \omega_k^{-1} \tag{56b}$$

$$\omega_k^{AD} = C(\mathbf{k}_1, -\mathbf{k}) \omega_k^A \tag{56c}$$

The impurity modes which significantly contribute to the Debye-Waller factor and neutron scattering in the present case can be characterized by the gap (G) or local (L) mode frequencies according to the nature of the impurities as

$$\omega_{G,L}^2 = [\tilde{\omega}_k^2 + 2\omega_k \Delta_k'(\tilde{\omega}_k) - 16\omega_k I_1]/(1 + 16\omega_k I_2) \tag{57}$$

where

$$\Delta_k'(\tilde{\omega}_k) = \Delta_k^A(\tilde{\omega}_k) + \Delta_k^{AD}(\tilde{\omega}_k) \tag{58}$$

$$I_1 = [V/(2\pi)^3] \int R(-\mathbf{k}, \mathbf{k}_1) R^*(-\mathbf{k}, \mathbf{k}_1) \times (\omega_{k_1}/\tilde{\omega}_{k_1}^2) k_1^3 \sin\theta \, dk_1 \, d\theta \, d\phi \tag{59a}$$

and

$$I_2 = [V/(2\pi)^3] \int R(-\mathbf{k}, \mathbf{k}_1) R^*(-\mathbf{k}, \mathbf{k}_1) \times (\omega_{k_1}/\tilde{\omega}_{k_1}^4) k_1^3 \sin\theta \, dk_1 \, d\theta \, d\phi \tag{59b}$$

Equations (31), (38), and (48) show that the distributions are asymmetric in character and tend to distort the Lorentzian line-shape distribution. These distributions are certainly not of similar nature to what has been found in the harmonic approximation to give the delta-function peaks. However, if the Breit-Wigner approximation is introduced, these distributions tend to give a delta shape distribution, but one that is temperature

and impurity concentration dependent. The temperature dependence of the distribution and frequency adds a new feature to the present approach.

The phonon linewidth $\Gamma_k(\omega)$ frequently appears in the results, which is a measure of phonon lifetime, thus making the problem dependent on phonon lifetime. The nature of a phonon lifetime showing temperature, frequency, and defect dependence is described by equation (43), which on substitution in equations (31), (38), and (48) gives obvious temperature and defect dependences. Apart from the methods of harmonic approximations (Shukla, 1980), we have obtained the two-phonon inelastic differential cross section completely on the anharmonic treatment. The contribution of phonon lifetimes to the two-phonon processes appears in the form

$$\Gamma^2_{k_1}(\tilde{\tilde{\omega}}_{k_1}) \cong [\Gamma^D_{k_1}(\tilde{\tilde{\omega}}_{k_1})]^2 + [\Gamma^A_{k_1}(\tilde{\tilde{\omega}}_{k_1})]^2 + 2\Gamma^A_{k_1}(\tilde{\tilde{\omega}}_{k_1})\Gamma^D_{k_1}(\tilde{\tilde{\omega}}_{k_1}) \tag{60}$$

which indicates that the magnitude of this quantum correction is not very large, but still is not negligible. A large number of phonon peaks appearing in the frequency spectrum exhibits the energy distribution at different frequencies, $\pm\tilde{\omega}_{k_1}$, $\pm\omega_{\pm\alpha}$, $\pm\omega_{\pm\beta}$, $\pm\tilde{\tilde{\omega}}_{k_i}$, etc., due to the neutron scattering.

It emerges from the present study that all the salient features of the Debye-Waller factor and the scattering of neutrons from single- and two-phonon band modes in an isotropically disordered crystal can be discussed by considering the defect and anharmonic terms in the crystal Hamiltonian. The appearance of an impurity-anharmonicity interference mode is the salient feature of the present work which cannot be ignored from the theory. We have not evaluated the values of various contributions to the neutron scattering, due to the great complexity of the computation, which chiefly arises from the large range of phonon frequencies involved and the presence of anharmonicity and defects in the crystal. The present results are general and can be applied to specific model crystals with the help of a computer.

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