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TEMPERATURE DEPENDENCE OF THE QUARTIC CONTRIBUTION TO THE DAMPING COEFFICIENT ASSOCIATED WITH LATTICE RESONANCE IN IONIC CRYSTALS: I *

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ABSTRACT

A comprehensive analytical analysis of the damping coefficient and its temperature dependence associated with lattice resonance in pure ionic crystals is presented. Contributions from cubic and quartic terms in the Hamiltonian and first- and second-order terms in the electric moment are taken into account. A first-order self-consistent method for obtaining a closed system of dependent Green-function equations is used to derive the appropriate expressions for the damping coefficients.

It is shown that the quartic contribution to the damping coefficient is different from the previously obtained T^2 (classical limit) expression. However, our result for the quartic contribution is in qualitative agreement with experimental results that take into account the implicit temperature dependence of the thermal expansion coefficient.

I. INTRODUCTION

Theories of lattice dynamics attempt to explain the nature of interatomic forces in crystals, and the variation of the associated damping coefficient with temperature is of fundamental importance in these theories. The problem of determining the proper temperature dependence of the damping coefficient has motivated considerable experimental and theoretical research for many years.[1] During the last ten years, significant progress (as a result of intensive research) has been made in identifying the essential features of the problem.[2-13] However, the basic problem remains unresolved. A comprehensive review of recent theoretical and experimental developments has been given by Mooij.[14]

The present paper is devoted to a complete and systematic analytical analysis of the damping coefficient and its temperature variation associated with lattice resonance in pure ionic crystals. Contributions from the second-order electric moment and quartic terms in the Hamiltonian are treated by use of the same method as that employed in the trivial harmonic approximation. The second step in this investigation is the numerical evaluation of the derived expressions for the damping coefficient in each case. This evaluation and a detailed comparison with available experimental results will be presented in a separate paper. In the present paper, we show that our results are in qualitative agreement with experimental results.

The equation-of-motion technique for two-time Green's functions is the main mathematical tool used in this analysis. It is well known that an infinite set of dependent (coupled) Green-function equations is implied in this technique. We use a symmetric decoupling method to terminate (form a closed system) this system of equations at the first-

order self-consistent stage. That is to say, our prescription is to (a) write the equations for the original Green's functions (these Green's functions will be referred to as the first kind, and their equations will be classified as first order), (b) examine the generated higher-order Green's functions (second kind) in the first-order equations and reduce (when possible), by means of symmetric decoupling, Green's functions of the second kind to those of the first kind, (c) write equations for the irreducible second kind (second-order equations), and (d) reduce the higher-order Green's functions in the second-order equations to Green's functions of the first and/or second kind. In general, higher-order equations may be needed. However, our problem does not require equations of orders higher than the second before self-consistence is achieved. The crucial point is the reduction of generated Green's functions to those previously defined at the earliest possible stage (the first-order self-consistent method).

By use of the above prescription for obtaining a closed system of equations, we have developed a solution for the well studied harmonic case and extended the analysis (same technique) to obtain results for higher-order contributions to the damping coefficient. The usual lower-order results are obtained, but the damping coefficient which includes the quartic contribution differs from the usual explicit T^2 (classical limit) expression. In our higher-order results, the arguments of the accompanying Dirac delta functions contain functions with an explicit temperature dependence.

The essentials of two-time Green-function theory needed in our analysis along with details related to the crystal and field interactions are presented in Sec. II. An analysis of the system in the

harmonic approximation is given in Sec. III, and the lowest-order damping coefficients are developed in Sec. IV. Section V is devoted to the calculation of quartic contributions to the damping coefficient. For continuity of presentation, we have placed the details of lengthy decoupling analyses in the Appendices.

II. MATHEMATICAL PRELIMINARIES

In this section, we (a) present a brief introduction to the theory of two-time temperature-dependent retarded Green's functions (two-time Green's functions), (b) express the linear electric susceptibility tensor in terms of the Fourier transform of a two-time Green's function, and (c) represent the anharmonic Hamiltonian (to quartic terms) and the electric moment (to second-order terms) of an ionic crystal in terms of phonon creation and annihilation operators.

A. Two-Time Green's Functions

Kubo,[15] in a now classical paper, developed a general statistical-mechanical scheme for calculating transport coefficients (response functions) without the traditional use of Boltzmann's equation. However, it is extremely difficult to evaluate Kubo's response formulas directly. By establishing a connection between Kubo's formulation and field-theoretical Green's functions,[16] Bogolyubov and Tyablikov[17] developed the method of two-time Green's functions. Hence the method of two-time Green's functions is a powerful theoretical tool which can be used to ascertain important physical information in response related problems. The two-time Green's function is defined by[18]

$$\begin{aligned} G(t - t') &= -i\theta(t - t')\langle[A(t), B(t')]\rangle \\ &= \langle\langle A(t); B(t')\rangle\rangle = G(A; B) \end{aligned} \tag{2.1}$$

where

$$\theta(t - t') = \begin{cases} 1; & t - t' > 0 \\ 0; & t - t' < 0 \end{cases} . \tag{2.2}$$

The causality condition, $G(t - t') = 0$ for $t - t' < 0$, clearly holds because of the conditions imposed on Θ. The canonical ensemble (thermal) average is indicated by $\langle \cdots \rangle$ and is defined by

$$\langle \cdots \rangle = \frac{\mathrm{Tr}(e^{-\beta H} \cdots)}{\mathrm{Tr}\ e^{-\beta H}} ; \quad \beta = (K_B T)^{-1} . \tag{2.3}$$

The operators A and B are second quantized operators in the Heisenberg representation. The trace is denoted by Tr; K_B and T are Boltzmann's constant and absolute temperature, respectively. The Hamiltonian of the isolated system under investigation is represented by H. Throughout this paper, $\hbar$ is taken to be unity.

Differentiation of $G(t - t')$ with respect to t leads to the following equation of motion for the two-time Green's function:

$$\frac{i dG(t - t')}{dt} = \delta(t - t')\langle [A(t),B(t')] \rangle + \langle\langle [A(t),H];\ B(t') \rangle\rangle . \tag{2.4}$$

The Fourier transform of $G(t - t')$ is given by

$$G(E) = G(A;\ B)_E = \frac{1}{2\pi} \int_{-\infty}^{\infty} G(\tau)\ e^{iE\tau}\ d\tau \tag{2.5}$$

where $\tau = t - t'$. On taking the Fourier transform of Eq. (2.4), we obtain (in the energy representation)

$$EG(A;\ B)_E = \frac{1}{2\pi} \langle [A,B] \rangle_E + G([A,H];\ B)_E \tag{2.6}$$

where $E = \omega + i\epsilon$. The frequency of the external perturbing field is ω, and the adiabatic switching-on of the perturbation is controlled by ϵ.

B. The Susceptibility Tensor

Zubarev[18] has shown that the $\mu\nu$ component of the linear complex susceptibility tensor per unit volume can be written in the following form:

$$\chi_{\mu\nu}(\omega) = -2\pi\, G(M_\mu;\, M_\nu)_E \tag{2.7}$$

where M is the electric moment of the system. The directions of the applied field and the response are ν and μ respectively.

One can readily establish the connection between $\chi_{\mu\nu}(\omega)$ and the differential (absorption) cross section for scattering by the system in question.[19] It can also be shown that the absorption coefficient corresponding to $\chi_{\mu\nu}(\omega)$ is given by[20]

$$\alpha_{\mu\nu}(\omega) = 4\pi \frac{\omega}{c} \operatorname{Im} \chi_{\mu\nu}(\omega) . \tag{2.8}$$

C. The Hamiltonian

The system to be placed in a perturbing electric field is a crystal with N unit cells each containing "rigid" ions. If periodic boundary conditions are imposed on the system, the lattice Hamiltonian (to quartic anharmonic terms) in terms of phonon creation and annihilation operators is[21]

$$H = H^+ = H_0 + H_A = H_0 + H_3 + H_4 \tag{2.9}$$

where

$$H_0 = \sum_{\alpha} \omega_\alpha a_\alpha^+ a_\alpha \tag{2.10}$$

$$H_3 = \sum_{\alpha,\beta,\gamma} V^{(3)}_{\alpha,\beta,\gamma} A_\alpha A_\beta A_\gamma \tag{2.11}$$

and

$$H_4 = \sum_{\alpha,\beta,\gamma,\rho} V^{(4)}_{\alpha,\beta,\gamma,\rho} A_\alpha A_\beta A_\gamma A_\rho \ . \tag{2.12}$$

The indices α, β, γ, and ρ are used to represent both the wave vectors $\underset{\sim}{k}$ and polarization (branch) index j. For example, $\alpha = \underset{\sim}{k}j$ and $\beta = \underset{\sim}{k}'j'$. Capital A is defined as follows: $A_\alpha = a_{-\alpha}^+ + a_\alpha$. The quantities $a_{-\alpha}^+$ and a_α are phonon creation and annihilation operators respectively. Note that $[a_\alpha, a_\beta^+] = \delta_{\alpha,\beta}$ and $[a_\alpha, a_\beta] = [a_\alpha^+, a_\beta^+] = 0$. The frequency of the normal mode of vibration is denoted by ω_α. The $V^{(3)}$ and $V^{(4)}$ coupling coefficients are Fourier transforms of the third- and fourth-order atomic force constants respectively. They are given by[1]

$$V^{(3)}_{\alpha,\beta,\gamma} = \frac{1}{6(2^{3/2})\, N^{1/2}} \frac{\Delta^{(3)}(\underset{\sim}{K})\ \Phi(\alpha,\beta,\gamma)}{(\omega_\alpha\, \omega_\beta\, \omega_\gamma)^{1/2}} \tag{2.13}$$

and

$$V^{(4)}_{\alpha,\beta,\gamma,\rho} = \frac{1}{4(24N)} \frac{\Delta^{(4)}(\underset{\sim}{K})\ \Phi(\alpha,\beta,\gamma,\rho)}{(\omega_\alpha\, \omega_\beta\, \omega_\gamma\, \omega_\rho)^{1/2}} \ . \tag{2.14}$$

The Φ's are force constants, and the V's are completely symmetric in the indices. For example (diatomic crystals),[1,7]

$$V^{(3)}_{\alpha,\beta,\gamma} = V^{(3)}_{\beta,\alpha,\gamma} = V^{(3)*}_{-\alpha,-\beta,-\gamma} = -V^{(3)}_{-\alpha,-\beta,-\gamma} \tag{2.15}$$

and

$$V^{(4)}_{\alpha,\beta,\gamma,\rho} = V^{(4)}_{\beta,\alpha,\gamma,\rho} = V^{(4)*}_{-\alpha,-\beta,-\gamma,-\rho} = V^{(4)}_{-\alpha,-\beta,-\gamma,-\rho} \, . \tag{2.16}$$

In addition to Eq. (2.15), we also require that $V^{(3)}_{-\alpha,\alpha,\ldots} = 0$.

Translational invariance restricts $\Delta^{(3)}(\underset{\sim}{K})$ by the following relation:[1,7]

$$\Delta^{(3)}(\underset{\sim}{K}) = \begin{cases} 1; & \underset{\sim}{K} = \underset{\sim}{k} + \underset{\sim}{k}' + \underset{\sim}{k}'' \\ 0; & \underset{\sim}{K} \neq \underset{\sim}{k} + \underset{\sim}{k}' + \underset{\sim}{k}'' \end{cases} \tag{2.17}$$

where $\underset{\sim}{K}$ is the primitive translational vector of the reciprocal lattice. A similar condition is valid for $\Delta^{(4)}(\underset{\sim}{K})$. The problem associated with mixed modes (polaritons) will not be considered, and the Born-Oppenheimer approximation is assumed to be valid.

D. The Electric Moment

The electric moment of our crystal can be represented as a general Taylor expansion in the nuclear displacements. The linear term in this expansion is the usual dipole moment. Higher-order terms result from charge deformation produced during lattice vibration. The μth component of the electric moment, to second-order, in terms of phonon creation and annihilation operators is[1]

$$M_\mu = M_\mu^{\,1} + M_\mu^{\,2} \tag{2.18}$$

where

$$M_\mu^{\,1} = N^{1/2} \sum_0 \frac{M_\mu(0)}{(2\omega_0)^{1/2}} A_0 \tag{2.19}$$

and

$$M_\mu^{\,2} = \frac{1}{4} \sum_{\alpha,\delta} \frac{M_\mu(\alpha,-\delta)}{(\omega_\alpha\ \omega_\delta)^{1/2}} A_\alpha A_{-\delta} \tag{2.20}$$

where $M_\mu(0)$ and $M_\mu(\alpha,-\delta)$ are the usual expansion coefficients,[1] $\delta = \underset{\sim}{k}j'$, and the index 0 means 0j. The following symmetry relations hold for the second-order expansion coefficient:

$$M_\mu(\alpha,-\delta) = M_\mu(-\delta,\alpha) = M_\mu^{\,*}(-\alpha,\delta) \ . \tag{2.21}$$

We neglect contributions related to the interactions of multipole moments of the ions with the applied electromagnetic radiation.

III. LINEAR SUSCEPTIBILITY AND ABSORPTION: $H_A = 0$

The physical fact that higher-order effects are generated from lower-order processes is an intrinsic feature of the first-order self-consistent method of forming a closed set of Green-function equations. This feature is revealed by use of a direct analysis of the basic equations for our main results in Sec. V. Our plan of attack, however, is to proceed from the simpler lower-order problem to the higher-order results. Using this plan, we are able to (a) clearly explain the basic mathematical method by solving a well-known problem and (b) show that the first-order self-consistent scheme is a viable method before obtaining results for M^2 and H_4 contributions to the damping coefficients. The harmonic approximation results in Eqs. (3.7) and (3.19) are well known.[1]

The complete expression for the $\mu\nu$ component of the linear complex susceptibility tensor is

$$\chi_{\mu\nu}^{1-2}(\omega) = \chi_{\mu\nu}^{11}(\omega) + \chi_{\mu\nu}^{12}(\omega) + \chi_{\mu\nu}^{21}(\omega) + \chi_{\mu\nu}^{22}(\omega) \tag{3.1}$$

where

$$\left.\begin{aligned} \chi_{\mu\nu}^{11}(\omega) &= -2\pi\, G(M_\mu{}^1;\, M_\nu{}^1)_E, \qquad \chi_{\mu\nu}^{12}(\omega) = -2\pi\, G(M_\mu{}^1;\, M_\nu{}^2)_E \\ \chi_{\mu\nu}^{21}(\omega) &= -2\pi\, G(M_\mu{}^2;\, M_\nu{}^1)_E, \text{ and } \quad \chi_{\mu\nu}^{22}(\omega) = -2\pi\, G(M_\mu{}^2;\, M_\nu{}^2)_E. \end{aligned}\right\} \tag{3.2}$$

The related problem of nonlinear susceptibility and damping (for $M^2 = 0$) has been treated.[13]

A. The One-Phonon Process

In this trivial case, we must evaluate ${}^{0}\chi^{11}_{\mu\nu}(\omega)$. Here the superscript on the left of χ refers to the order of approximation in the Hamiltonian, and the superscripts on the right refer to the orders of the moments considered. For example,

$$ {}^{0}\chi^{11}_{\mu\nu}(\omega) = -2\pi\, G(M_\mu{}^1;\, M_\nu{}^1)_E $$

$$ = -\frac{2\pi N}{2} \sum_{0,0'} \frac{M_\mu(0)\, M_\nu(0')}{(\omega_0\, \omega_{0'})^{1/2}} (G^1 + G^2) \qquad (3.3) $$

where $G^1 = G(a_0^+;\ A_{0'})_E$ and $G^2 = G(a_0;\ A_{0'})_E$.

The equations for G^1 and G^2 in the energy representation are

$$ EG^1 = \frac{1}{2\pi} \langle [a_0^+,\ A_{0'}] \rangle_E + G([a_0^+,\ H_0];\ A_{0'})_E \qquad (3.4) $$

and

$$ EG^2 = \frac{1}{2\pi} \langle [a_0, A_{0'}] \rangle_E + G([a_0, H_0];\ A_{0'})_E\ . \qquad (3.5) $$

On solving Eqs. (3.4) and (3.5) for G^1 and G^2 respectively and substituting the result into Eq. (3.3), we obtain

$$
{}^{0}\chi^{11}_{\mu\nu} = \frac{N}{2} \sum_{0} \frac{M_\mu(0)\, M_\nu(0)}{\omega_0} \left(\frac{1}{E + \omega_0} - \frac{1}{E - \omega_0} \right)
$$

$$
= \frac{N}{2} \sum_{0} \frac{M_\mu(0)\, M_\nu(0)}{\omega_0} \left\{ p\left(\frac{1}{\omega + \omega_0}\right) - p\left(\frac{1}{\omega - \omega_0}\right) \right.
$$

$$
\left. + i\pi[\delta(\omega - \omega_0) - \delta(\omega + \omega_0)] \right\} \quad . \tag{3.6}
$$

We have used the Dirac identity,

$$
\lim_{\epsilon \to 0} \left(\frac{1}{X \pm i\epsilon} \right) = p\left(\frac{1}{X}\right) \mp i\pi\, \delta(X),
$$

to obtain the final result in Eq. (3.6). The corresponding expression for the absorption coefficient is

$$
{}^{0}\alpha^{11}_{\mu\nu}(\omega) = \left(\frac{4\pi^2}{c}\right) \frac{N}{2} \sum_{0} \frac{M_\mu(0)\, M_\nu(0)}{\omega_0} [\delta(\omega - \omega_0) - \delta(\omega + \omega_0)] \; . \tag{3.7}
$$

The absorption, $\delta(\omega - \omega_0)$, consists of δ-function-type absorption lines at $\omega = \omega_0$ (no damping). This is the well-known result for the absorption by a system composed of N independent harmonic oscillators all of frequency ω_0. The $\delta(\omega + \omega_0)$ term represents a pole in the complex frequency (energy) plane for negative ω_0. This latter condition is needed to satisfy the requirements of causality and reality of the response function (Krammer-Kronig relations). Note that ${}^{0}\alpha^{11}_{\mu\nu}(\omega)$ has no explicit temperature dependence.

B. Two-Phonon Processes and ${}^{0}\chi^{12}_{\mu\nu}(\omega)$

Here we are interested in determining the effect of the second-order electric moment on the absorption. We must evaluate ${}^{0}\chi^{12}_{\mu\nu}(\omega)$, ${}^{0}\chi^{21}_{\mu\nu}(\omega)$, and ${}^{0}\chi^{22}_{\mu\nu}(\omega)$. The required expression for ${}^{0}\chi^{12}_{\mu\nu}(\omega)$ (one-phonon process) is

$$ {}^{0}\chi^{12}_{\mu\nu}(\omega) = \frac{-2\pi N^{1/2}}{4} \sum_{0,\alpha',\delta'} \frac{M_\mu(0)\, M_\nu(\alpha',-\delta')}{(2\omega_0\, \omega_{\alpha'}\, \omega_{\delta'})^{1/2}} (S^1 + S^2) \quad (3.8) $$

where $S^1 = G(a_0^+; A_{\alpha'}, A_{-\delta'})_E$ and $S^2 = G(a_0; A_{\alpha'}, A_{-\delta'})_E$. The required equations for S^1 and S^2 are

$$ ES^1 = \frac{1}{2\pi} \langle [a_0^+, A_{\alpha'}, A_{-\delta'}] \rangle_E + G([a_0^+, H_0]; A_{\alpha'}, A_{-\delta'})_E \quad (3.9) $$

and

$$ ES^2 = \frac{1}{2\pi} \langle [a_0, A_{\alpha'}, A_{-\delta'}] \rangle_E + G([a_0, H_0]; A_{\alpha'}, A_{-\delta'})_E \quad . \quad (3.10) $$

On solvings Eqs. (3.9) and (3.10) for S^1 and S^2 respectively, we obtain $S^1 = S^2 = 0$. Hence ${}^{0}\chi^{12}_{\mu\nu}(\omega) = 0$; similarly, ${}^{0}\chi^{21}_{\mu\nu}(\omega) = 0$. In obtaining this result, we have assumed that the thermal average of an odd number of creation or annihilation operators is equal to zero. We thus conclude that the interaction between the first- and second-order electric moments introduces no contribution to the absorption coefficient in the harmonic approximation.

We now proceed to evaluate ${}^{0}\chi^{22}_{\mu\nu}(\omega)$; its basic equation is

$$ {}^{0}\chi^{22}_{\mu\nu}(\omega) = -\frac{2\pi}{16} \sum_{\alpha',\delta'} \sum_{\alpha'',\delta''} \frac{M_\mu(\alpha',-\delta')\, M_\nu(\alpha'',-\delta'')}{(\omega_{\alpha'}\, \omega_{\delta'}\, \omega_{\alpha''}\, \omega_{\delta''})^{1/2}} $$

$$ \times\ (H^1 + 2H^2 + H^3) \tag{3.11} $$

where

$$ H^1 = G(a^+_{-\alpha'}, a^+_{\delta'}; A_{\alpha''} A_{-\delta''})_E, \quad H^2 = G(a^+_{-\alpha'}, a_{-\delta'}; A_{\alpha''} A_{-\delta''})_E , $$

$$ \text{and} \quad H^3 = G(a_{\alpha'}, a_{-\delta'}; A_{\alpha''} A_{-\delta''})_E . $$

The appropriate equations for the H-type Green's functions are

$$ H^1 = \frac{\frac{1}{2\pi} \langle [H'] \rangle_E}{E + \omega_{\alpha'} + \omega_{\delta'}} \tag{3.12} $$

$$ H^2 = \frac{\frac{1}{2\pi} \langle [H^2] \rangle_E}{E + \omega_{\alpha'} - \omega_{\delta'}} \tag{3.13} $$

and

$$ H^3 = \frac{\frac{1}{2\pi} \langle [H^3] \rangle_E}{E - \omega_{\alpha'} - \omega_{\delta'}} \tag{3.14} $$

where

$$ \langle [H^1] \rangle_E \approx -[n_{\delta'}\, \delta_{\delta',-\delta''}\, \delta_{-\alpha',\alpha''} + (n_{\delta'} + 1)\, \delta_{\alpha'',\delta'}\, \delta_{\alpha',\delta''} $$

$$ + n_{\alpha'}\, \delta_{\alpha',\delta''}\, \delta_{\delta',\alpha''} + (n_{\alpha'} + 1)\, \delta_{\alpha'',-\alpha'}\, \delta_{\delta',-\delta''}] \tag{3.15} $$

$$\langle [H^2] \rangle_E \approx n_{\alpha'} \delta_{\alpha',\delta''} \delta_{\delta',\alpha''} + (n_{\alpha'} + 1) \delta_{\alpha'',-\alpha'} \delta_{-\delta',\delta''}$$

$$- (n_{\delta''} + 1) \delta_{-\delta',\delta''} \delta_{-\alpha',\alpha''} - n_{\alpha''} \delta_{\alpha'',\delta'} \delta_{\alpha',\delta''} \tag{3.16}$$

$$\langle [H^3] \rangle_E \approx (n_{\delta''} + 1) \delta_{-\delta',\delta''} \delta_{\alpha',-\alpha''} + n_{\alpha''} \delta_{\alpha'',\delta'} \delta_{\alpha',\delta''}$$

$$+ (n_{\delta''} + 1) \delta_{\alpha',\delta''} \delta_{\delta',\alpha''} + n_{\alpha''} \delta_{-\alpha'',\alpha'} \delta_{-\delta',\delta''} \quad . \tag{3.17}$$

The susceptibility is

$$^0\chi^{22}_{\mu\nu}(\omega) = \frac{1}{8} \sum_{\alpha,\delta} \frac{M_\mu(\alpha,-\delta)\, M_\nu^*(\alpha,-\delta)}{\omega_\alpha\, \omega_\delta}$$

$$\times \left(\frac{n_\alpha + n_\delta + 1}{E + \omega_\alpha + \omega_\delta} + \frac{2(n_\delta - n_\alpha)}{E + \omega_\alpha - \omega_\delta} - \frac{n_\alpha + n_\delta + 1}{E - \omega_\alpha - \omega_\delta} \right) \quad . \tag{3.18}$$

The corresponding absorption coefficient is

$$^0\alpha^{22}_{\mu\nu}(\omega) = \frac{4\pi^2\omega}{8c} \sum_{\alpha,\delta} \frac{M_\mu(\alpha,-\delta)\, M_\nu^*(\alpha,-\delta)}{\omega_\alpha\, \omega_\delta} \Big\{ (n_\alpha + n_\delta + 1)$$

$$\times [\delta(\omega - \omega_\alpha - \omega_\delta) - \delta(\omega + \omega_\alpha + \omega_\delta)] + 2(n_\alpha - n_\delta)\, \delta(\omega + \omega_\alpha - \omega_\delta) \Big\} \quad . \tag{3.19}$$

We have restricted the calculation to the case of nonpiezoelectric crystals, $\omega_\alpha = \omega_{-\alpha}$. The $n_{\ldots}$ quantities are the usual average boson occupation numbers, $n_\alpha = (\exp \beta\, \omega_\alpha - 1)^{-1}$. Here we have absorption away from $\omega = \omega_0$.

The summation over $\underset{\sim}{k}$ in Eq. (3.19) indicates continuous absorption. This continuous absorption provides a mechanism for explaining the presence of absorption peaks on the short-wavelength side of the main peak in the spectra of alkali halide crystals. That is to say, short-wavelength modes produce charge deformations that result in absorption by a pair of short-wavelength phonons. The explicit temperature dependence of ${}^{0}\alpha^{22}_{\mu\nu}(\omega)$ is contained in the definition of the n's.

In homopolar materials (such as diamond, silicon, and germanium), $M^1 = 0$. Hence the intrinsic lattice absorption is explained by use of M^2. This process is clearly demonstrated in Eq. (3.19).

The results in this harmonic approximation are quite well known. However, it is important to note the straightforward manner in which they were obtained. Note particularly the ease at which the absorption coefficient for the second-order moment, Eq. (3.19), was obtained without solving a complicated integral equation.[22] Equation (3.19) has also been derived by use of the thermodynamic Green's function method which makes use of diagrams.[5] The results for higher-order contributions will be obtained using the same procedure.

IV. LINEAR SUSCEPTIBILITY AND DAMPING: $H_3 \neq 0$

A. One-Phonon Process: ${}^3\chi^{11}_{\mu\nu}(\omega)$

The right-hand side of Eq. (3.3) is the required expression for the susceptibility in this case. The equations for G^1 and G^2 now become

$$(E + \omega_0)G^1 = -\frac{\delta_{0,0'}}{2\pi} - 3\sum_{\alpha,\beta} V^{(3)}_{-\alpha,\beta,0}(g^1 + 2g^2 + g^3) \tag{4.1}$$

and

$$(E - \omega_0)G^2 = \frac{\delta_{0,0'}}{2\pi} + 3\sum_{\alpha,\beta} V^{(3)}_{-\alpha,\beta,0}(g^1 + 2g^2 + g^3) \ . \tag{4.2}$$

The higher-order Green's functions are given by $g^1 = G(a^+_\alpha a^+_{-\beta}; A_{0'})_E$ $g^2 = G(a^+_\alpha a_\beta; A_{0'})_E$, and $g^3 = G(a_{-\alpha} a_\beta; A_{0'})_E$. To solve the equations for G^1 and G^2, we must first find the appropriate expressions for the g Green's functions. The decoupled equations for the g Green's functions are

$$(E + \omega_\alpha + \omega_\beta)g^1 = -6[V^{(3)}_{\alpha,0,-\beta}(n_\alpha + n_\beta + 1)G^1 + V^{(3)}_{\alpha,-\beta,0}(n_\alpha + n_\beta + 1)G^2] \tag{4.3}$$

$$(E + \omega_\alpha - \omega_\beta)g^2 = 6V^{(3)}_{\alpha,0,-\beta}(n_\alpha - n_\beta)G^1 + 6V^{(3)}_{\alpha,0,-\beta}(n_\alpha - n_\beta)G^2 \tag{4.4}$$

and

$$(E - \omega_\alpha - \omega_\beta)g^3 = 6V^{(3)}_{\alpha,0,-\beta}(n_\alpha + n_\beta + 1)G^1$$

$$+ 6V^{(3)}_{\alpha,0,-\beta}(n_\alpha + n_\beta + 1)G^2 \ . \qquad (4.5)$$

A symmetric decoupling scheme which consists of retaining only diagonal terms was used to terminate the equations for the g Green's functions. This scheme is made self-consistent by terminating the system of dependent equations in such a way that the original Green's functions appear in the decoupled equations for the higher-order Green's functions. For example,

$$\langle\langle a^+_{-\alpha} a_\beta a^+_{-\gamma}; A_{0'}\rangle\rangle \approx [n_\beta \, \delta_{-\alpha,\beta} \, \delta_{-\gamma,0} + (n_\beta + 1)\delta_{\beta,-\gamma} \, \delta_{-\alpha,0}]G^1 \ . \qquad (4.6)$$

On solving Eqs. (4.3), (4.4), and (4.5) for g^1, g^2, and g^3 respectively, substituting the results into Eqs. (4.1) and (4.2), and neglecting terms of order $1/N^2$, the susceptibility becomes

$$^3\chi^{11}_{\mu\nu}(\omega) = N \sum_0 \frac{M_\mu(0) \, M_\nu(0)}{\omega_0^{\,2} - \omega^2 + 2\omega_0 \, {}^3\gamma^{11}(n,E)} \qquad (4.7)$$

where $^3\gamma^{11}(n,E) = {}^3\Delta^{11}(n,\omega) - i \, {}^3\Gamma^{11}(n,\omega)$. The frequency shifts, $^3\Delta^{11}$, and damping coefficient, $^3\Gamma^{11}$, are given respectively by

$$ {}^3\Delta^{11}(n,\omega) = 18 \sum_{\alpha,\beta} \left| V^{(3)}_{-\alpha,0,\beta} \right|^2 \left\{ (n_\alpha + n_\beta + 1) \left[p\left(\frac{1}{\omega - \omega_\alpha - \omega_\beta}\right) \right.\right. $$

$$ \left.\left. - \; p\left(\frac{1}{\omega + \omega_\alpha + \omega_\beta}\right) \right] + 2(n_\alpha - n_\beta)\; p\left(\frac{1}{\omega + \omega_\alpha - \omega_\beta}\right) \right\} \qquad (4.8) $$

and

$$ {}^3\Gamma^{11}(n,\omega) = 18\pi \sum_{\alpha,\beta} \left| V^{(3)}_{-\alpha,0,\beta} \right|^2 \{ (n_\alpha + n_\beta + 1)[\delta(\omega - \omega_\alpha - \omega_\beta) $$

$$ - \; \delta(\omega + \omega_\alpha + \omega_\beta)] + 2(n_\alpha - n_\beta)\; \delta(\omega + \omega_\alpha - \omega_\beta)\} \; . \qquad (4.9) $$

The corresponding absorption coefficient is

$$ {}^3\alpha^{11}_{\mu\nu}(\omega) = \frac{8\pi\omega N}{c} \sum_0 \frac{M_\mu(0)\; M_\nu(0)\; \omega_0\; {}^3\Gamma^{11}(n,E)}{(\omega_0{}^2 - \omega^2)^2 + 4\omega_0(\omega_0{}^2 - \omega^2)\; {}^3\Delta^{11}(n,E)} \; . \qquad (4.10) $$

Here we obtain the usual result of a damping coefficient which has an explicit T dependence in the classical limit. However, a complete analysis requires a detailed assessment of the implicit temperature dependence of the $V^{(3)}$ coefficient. This result for ${}^3\chi^{11}_{\mu\nu}(\omega)$ was first obtained by Vinogradov[23] and Wallis and Maradudin.[24]

In summary, we find that M^1 $(H_A = 0)$ leads to δ-function-type absorption lines (no damping), and H_3 removes the infinite sharpness (introduces frequency shifts and damping) of the absorption lines. The second-order electric moment, M^2, gives rise to

continuous absorption which leads to secondary absorption away from the main absorption peak.

B. One-Phonon Process: ${}^{3}\chi^{12}_{\mu\nu}(\omega)$

The required expression for the susceptibility is given in Eq. (3.8). The equations for the S Green's functions in this case are

$$(E + \omega_0)S^1 = -3 \sum_{\alpha,\beta} V^{(3)}_{-\alpha,\beta,0}(s^1 + 2s^2 + s^3) \tag{4.11}$$

and

$$(E - \omega_0)S^2 = 3 \sum_{\alpha,\beta} V^{(3)}_{-\alpha,\beta,0}(s^1 + 2s^2 + s^3) \ . \tag{4.12}$$

The higher-order Green's functions are defined as follows: $s^1 = G(a^+_{-\alpha}\, a^+_{-\beta};\ A_{\alpha'}, A_{-\delta'})_E$, $s^2 = G(a^+_{\alpha}\, a_{\beta};\ A_{\alpha'}, A_{-\delta'})_E$, and $s^3 = G(a_{-\alpha} a_{\beta};\ A_{\alpha'}, A_{-\delta'})_E$.

The standard procedure of solving the appropriate equations for s^1, s^2, and s^3 and substituting the result into Eqs. (4.11) and (4.12) is used to resolve this system of dependent equations. The equations for the higher-order Green's functions are

$$(E + \omega_\alpha + \omega_\beta)s^1 = \frac{1}{2\pi} \langle [s^1] \rangle_E - 6V^{(3)}_{\alpha,0,-\beta}(n_\alpha + n_\beta + 1)S^1$$

$$- 6V^{(3)}_{\alpha,0,-\beta}(n_\alpha + n_\beta + 1)S^2 \tag{4.13}$$

$$(E + \omega_\alpha - \omega_\beta)s^2 = \frac{1}{2\pi} \langle [s^2] \rangle_E + 6V^{(3)}_{\alpha,0,-\beta}(n_\alpha - n_\beta)s^1$$

$$+ 6V^{(3)}_{\alpha,0,-\beta}(n_\alpha - n_\beta)s^2 \qquad (4.14)$$

and

$$(E - \omega_\alpha - \omega_\beta)s^3 = \frac{1}{2\pi} \langle [s^3] \rangle_E + 6V^{(3)}_{\alpha,0,-\beta}(n_\alpha + n_B + 1)s^1$$

$$+ 6V^{(3)}_{\alpha,0,-\beta}(n_\alpha + n_\beta + 1)s^2 \qquad (4.15)$$

where

$$\langle [s^1] \rangle_E = -[n_\beta \, \delta_{\beta,\delta'} \, \delta_{-\alpha,\alpha'} + (n_\beta + 1)\delta_{\alpha',-\beta} \, \delta_{\alpha,\delta'}$$

$$+ n_\alpha \, \delta_{\alpha,\delta'} \, \delta_{-\beta,\alpha'} + (n_\alpha + 1)\delta_{-\alpha,\alpha'} \, \delta_{\beta,\delta'}] \qquad (4.16)$$

$$\langle [s^2] \rangle_E = n_\alpha \, \delta_{\alpha,-\delta'} \, \delta_{\beta,-\alpha'} + (n_\alpha + 1)\delta_{\alpha',\alpha} \, \delta_{\beta,\delta'}$$

$$- (n_{\delta'} + 1)\delta_{\beta,\delta'} \, \delta_{\alpha,\alpha'} - n_{\alpha'} \, \delta_{-\alpha',\beta} \, \delta_{\alpha,-\delta'} \qquad (4.17)$$

and

$$\langle [s^3] \rangle_E = (n_{\delta'} + 1)\delta_{\beta,\delta'} \, \delta_{\alpha,\alpha'} + n_{\alpha'} \, \delta_{-\alpha',\beta} \, \delta_{-\alpha,\delta'}$$

$$+ (n_{\delta'} + 1)\delta_{-\alpha,\delta'} \, \delta_{\beta,-\alpha'} + n_{\alpha'} \, \delta_{\alpha',\alpha} \, \delta_{\beta,\delta'} \, . \qquad (4.18)$$

On solving Eqs. (4.13), (4.14), and (4.15) for the higher-order Green's functions, substituting the results into Eqs. (4.11) and (4.12), and neglecting terms of order $1/N^2$, we find that the susceptibility becomes

$$ {}^{3}\chi^{12}_{\mu\nu}(\omega) = \frac{3}{2} N^{1/2} \sum_{0,\alpha',\delta'} \frac{M_\mu(0)\, M_\nu(\alpha',-\delta')}{(2\omega_0\, \omega_{\alpha'}\, \omega_{\delta'})^{1/2}} $$

$$ \times \left(\frac{\omega_0\, f^{12}(n,E)}{\omega_0^{\,2} - \omega^2 + 2\omega_0\, {}^{3}\gamma^{11}(n,E)} \right) \tag{4.19} $$

where

$$ f^{12}(n,E) = \frac{V^{(3)}_{-\alpha',\delta',0}\, 2(n_{\alpha'} + n_{\delta'} + 1)}{E - \omega_{\alpha'} - \omega_{\delta'}} $$

$$ - \frac{\left(V^{(3)}_{\alpha',\delta',0} + V^{(3)*}_{\alpha',\delta',0}\right)(n_{\alpha'} + n_{\delta'} + 1)}{E + \omega_{\alpha'} + \omega_{\delta'}} + \frac{V^{(3)}_{-\alpha',\delta',0}\, 2(n_{\delta'} - n_{\alpha'})}{E + \omega_{\delta'} - \omega_{\alpha'}} $$

$$ + \frac{V^{(3)}_{-\alpha',\delta',0}\, 2(n_{\alpha'} - n_{\delta'})}{E + \omega_{\alpha'} - \omega_{\delta'}} \quad . \tag{4.20} $$

This result was previously obtained by Cowley[5] using thermodynamic Green's functions. The corresponding absorption coefficient is given by

$$ {}^{3}\alpha^{12}_{\mu\nu}(\omega) = \frac{3}{2} N^{1/2} \sum_{0,\alpha',\delta'} \frac{M_\mu(0)\, M_\nu(\alpha',-\delta')}{(2\omega_0\, \omega_{\alpha'}\, \omega_{\delta'})^{1/2}} $$

$$ \times \frac{\omega_0[2\omega_0\, {}^{3}\Gamma^{11}(n,\omega) f_p + (\omega_0^{\,2} - \omega^2) + 2\omega_0\, {}^{3}\Delta^{11}(n,\omega) f_\delta]}{(\omega_0^{\,2} - \omega^2)^2 + 4\omega_0(\omega_0^{\,2} - \omega^2)\, {}^{3}\Delta^{11}(n,\omega)} \tag{4.21} $$

where $f^{12}(n,E) = f_p + if_\delta$. Here the form of the damping coefficient is the same as in the case for ${}^3\chi^{11}_{\mu\nu}(\omega)$. However, the absorption coefficient is modified by $f^{12}(n,E)$.

C. Two-Phonon Processes: ${}^3\chi^{21}_{\mu\nu}(\omega)$ AND ${}^3\chi^{22}_{\mu\nu}(\omega)$

The expression for ${}^3\chi^{21}_{\mu\nu}(\omega)$ is

$$ {}^3\chi^{21}_{\mu\nu}(\omega) = -\frac{2\pi N^{1/2}}{4} \sum_{0',\alpha',\delta'} \frac{M_\mu(\alpha',-\delta')\, M_\nu(0')}{(2\omega_{0'}\, \omega_{\alpha'}\, \omega_{\delta'})^{1/2}} (L^1 + 2L^2 + L^3) \tag{4.22} $$

where the L Green's functions are defined as follows:

$L^1 = G(a^+_{-\alpha'}, a^+_{\delta'}; A_{0'})_E$, $L^2 = G(a^+_{-\alpha'}, a_{-\delta'}; A_{0'})_E$, and $L^3 = G(a_{\alpha'}, a_{-\delta'}; A_{0'})_E$. The equations for the L Green's functions are

$$ (E + \omega_{\alpha'} + \omega_{\delta'})L^1 = G([a^+_{-\alpha'}, a^+_{\delta'}, H_3]; A_{0'})_E \tag{4.23} $$

$$ (E + \omega_{\alpha'} - \omega_{\delta'})L^2 = G([a^+_{-\alpha'}, a_{-\delta'}, H_3]; A_{0'})_E \tag{4.24} $$

and

$$ (E - \omega_{\alpha'} - \omega_{\delta'})L^3 = G([a_{\alpha'}, a_{-\delta'}, H_3]; A_{0'})_E \ . \tag{4.25} $$

The complete expressions for $L^1 + 2L^2 + L^3$ and $H^1 + 2H^2 + H^3$ for $H_3 \neq 0$ are derived in Appendix A. Hence we can obtain expressions for ${}^3\chi^{21}_{\mu\nu}(\omega)$ and ${}^3\chi^{22}_{\mu\nu}(\omega)$. The form of the contribution to the damping coefficient is the same in both cases.

For simplicity, we consider the following approximate expression for ${}^3\chi^{22}_{\mu\nu}(\omega)$:

$$
{}^3\chi^{22}_{\mu\nu}(\omega) \approx \frac{1}{8} \sum_{\alpha',\delta'} \frac{M_\mu(\alpha',-\delta')\, M_\nu^*(\alpha',-\delta')}{\omega_{\alpha'}\, \omega_{\delta'}}
$$

$$
\times \left(\frac{n_{\alpha'} + n_{\delta'} + 1}{\omega - \omega_{\alpha'} - \omega_{\delta'} - {}^3\gamma_3^{21}(n,E)} + \frac{2(n_{\alpha'} - n_{\delta'})}{\omega + \omega_{\alpha'} - \omega_{\delta'} + {}^3\gamma_2^{21}(n,E)} \right.
$$

$$
\left. \times \frac{n_{\alpha'} + n_{\delta'} + 1}{\omega + \omega_{\alpha'} + \omega_{\delta'} + {}^3\gamma_1^{21}(n,E)} \right) . \qquad (4.26)
$$

To obtain Eq. (4.26), we have neglected terms of order $1/N^{3/2}$. The corresponding expressions for the damping coefficients are

$$
{}^3\Gamma_1^{21}(n,\omega) = -6(18\pi) \sum_{\alpha,\beta} \left| V^{(3)}_{\alpha,\beta,\delta'} \right|^2 (n_\alpha + n_\beta + 1)
$$

$$
\times\ \delta(\omega + \omega_\alpha + \omega_{\alpha'} + \omega_\beta) - 18\pi \sum_{\alpha,\beta} \left| V^{(3)}_{\alpha,\beta,\delta'} \right|^2 (5n_\beta - 3n_\alpha + 1)
$$

$$
\times\ \delta(\omega + \omega_{\alpha'} + \omega_\alpha - \omega_\beta)
$$

$$
- 18\pi \sum_{\alpha,\beta} \left| V^{(3)}_{\alpha,\beta,\delta'} \right|^2 (5n_\alpha - 3n_\beta + 1)\ \delta(\omega + \omega_\beta + \omega_{\alpha'} - \omega_\alpha)
$$

$$
+ 18\pi \sum_{\alpha,\beta} \left| V^{(3)}_{\alpha,\beta,\delta'} \right|^2 (3n_\alpha + 2n_\beta + n_{\delta'} + \tfrac{1}{3})\ \delta(\omega + \omega_{\alpha'} - \omega_\alpha - \omega_\beta)
$$

(4.27)

$$
\begin{aligned}
{}^{3}\Gamma_{2}^{21}(n,\omega) \;=\; & -3(12\pi) \sum_{\alpha,\beta} \left|V^{(3)}_{\alpha,\beta,\delta'}\right|^{2} (n_\alpha + n_\beta + \tfrac{2}{3}) \\
& \times\; \delta(\omega + \omega_\alpha + \omega_\beta - \omega_{\alpha'}) \\
& + 18\pi \sum_{\alpha,\beta} \left|V^{(3)}_{\alpha,\beta,\delta'}\right|^{2} (5n_\alpha - n_\beta + \tfrac{4}{3})\; \delta(\omega + \omega_\alpha - \omega_\beta - \omega_{\alpha'}) \\
& + 18\pi \sum_{\alpha,\beta} \left|V^{(3)}_{\alpha,\beta,\delta'}\right|^{2} (5n_\beta - n_\alpha + \tfrac{4}{3})\; \delta(\omega + \omega_\beta - \omega_{\alpha'} - \omega_\alpha) \\
& + 3(18\pi) \sum_{\alpha,\beta} \left|V^{(3)}_{\alpha,\beta,\delta'}\right|^{2} (n_\alpha + n_\beta + 1)\; \delta(\omega - \omega_\alpha - \omega_{\alpha'} - \omega_\beta) \\
& + 3(18\pi) \sum_{\alpha,\beta} \left|V^{(3)}_{\alpha,\beta,\delta'}\right|^{2} (n_\alpha + n_\beta + 1)\; \delta(\omega + \omega_\alpha + \omega_{\alpha'} + \omega_\beta) \\
& + 3(24\pi) \sum_{\alpha,\beta} \left|V^{(3)}_{\alpha,\beta,\delta'}\right|^{2} (n_\beta + \tfrac{1}{3})\; \delta(\omega + \omega_{\alpha'} + \omega_\alpha - \omega_\beta) \\
& + 3(24\pi) \sum_{\alpha,\beta} \left|V^{(3)}_{\alpha,\beta,\delta'}\right|^{2} (n_\alpha + \tfrac{1}{3})\; \delta(\omega + \omega_\beta + \omega_{\alpha'} - \omega_\alpha) \\
& - 3(12\pi) \sum_{\alpha,\beta} \left|V^{(3)}_{\alpha,\beta,\delta'}\right|^{2} (n_\alpha + n_\beta + \tfrac{2}{3})\; \delta(\omega + \omega_{\alpha'} - \omega_\alpha - \omega_\beta) \qquad (4.28)
\end{aligned}
$$

and

$$
\begin{aligned}
{}^{3}\Gamma_{3}^{21}(n,\omega) = & -18\pi \sum_{\alpha,\beta} \left| V^{(3)}_{\alpha,\beta,\delta'} \right|^2 (3n_\alpha + 3n_\beta + \tfrac{7}{3}) \\
& \times \ \delta(\omega + \omega_\alpha + \omega_\beta - \omega_{\alpha'}) \\
& + 18\pi \sum_{\alpha,\beta} \left| V^{(3)}_{\alpha,\beta,\delta'} \right|^2 (6n_\alpha - 4n_\beta + \tfrac{5}{3})\ \delta(\omega + \omega_\alpha - \omega_\beta - \omega_{\alpha'}) \\
& + 18\pi \sum_{\alpha,\beta} \left| V^{(3)}_{\alpha,\beta,\delta'} \right|^2 (6n_\beta - 4n_\alpha + \tfrac{5}{3})\ \delta(\omega + \omega_\beta - \omega_{\alpha'} - \omega_\alpha) \\
& + 18\pi \sum_{\alpha,\beta} \left| V^{(3)}_{\alpha,\beta,\delta'} \right|^2 (n_\alpha + n_\beta + 1)\ \delta(\omega - \omega_\alpha - \omega_{\alpha'} - \omega_\beta) \ . \qquad (4.29)
\end{aligned}
$$

Again, we find that the explicit temperature dependence of the damping coefficient in the classical limit is T.

V. LINEAR SUSCEPTIBILITY AND DAMPING: $H_A \neq 0$

A. One-Phonon Processes: ${}^4\chi^{11}_{\mu\nu}(\omega)$ AND ${}^4\chi^{12}_{\mu\nu}(\omega)$

The equations for G^1 and G^2 to quartic terms in the Hamiltonian are

$$(E + \omega_0)G^1 = -\frac{\delta_{0,0}}{2\pi} + G([a_0^+, H_3]; A_{0'})_E + G([a_0^+, H_4]; A_{0'})_E \tag{5.1}$$

and

$$(E - \omega_0)G^2 = \frac{\delta_{0,0'}}{2\pi} + G([a_0, H_3]; A_{0'})_E + G([a_0, H_4]; A_{0'})_E \quad . \tag{5.2}$$

Our first-order self-consistent approach requires the reduction of terms involving H_4 in Eqs. (5.1) and (5.2) to the original G Green's functions. This procedure must be followed even if one is dealing with a model for H_3 only since the above H_4 terms are of order 1/N. The H_4 terms give rise to the usual Hartree-Fock approximation to the self-energy. This reduction introduces following contribution to the ${}^3\chi^{11}_{\mu\nu}(\omega)$:

$$ {}^3\chi^{11}_{\mu\nu}(\omega) = N \sum_0 M_\mu(0)\, M_\nu(0)$$

$$\times \left(\frac{1}{\omega_0^{\,2} - \omega^2 + 2\omega_0[\,\Sigma(0) + {}^3\gamma^{11}(n,E)]} \right) \tag{5.3}$$

where $\sum(0) = 12 \sum_{\alpha} V^{(4)}_{-\alpha,\alpha,0,-0}(2n_\alpha + 1)$. Here the damping coefficient for ${}^3\chi^{11}_{\mu\nu}(\omega)$ and ${}^3\chi^{12}_{\mu\nu}(\omega)$ remains the same, but the frequency shifts must be modified in the following manner:

${}^3\Delta^{11}_{new} = \sum(0) + {}^3\Delta^{11}_{old}(n,\omega)$.

The decoupled equations for the higher-order Green's functions (H_3 terms) in Eqs. (5.1) and (5.2) to quartic terms in the Hamiltonian are

$$[E + \omega_\alpha + \omega_\beta + R_1{}^1]g^1 = -[6V^{(3)}_{\alpha,0,-\beta}(n_\alpha + n_\beta + 1)G^1$$

$$+ 6V^{(3)}_{\alpha,-\beta,0}(n_\alpha + n_\beta + 1)G^2 + R_1{}^2\, g^2 + R_1{}^3\, g^3] \tag{5.4}$$

$$[E + \omega_\alpha - \omega_\beta - R_2{}^2]g^2 = 6V^{(3)}_{\alpha,0,-\beta}(n_\alpha - n_\beta)G^1$$

$$+ 6V^{(3)}_{\alpha,0,-\beta}(n_\alpha - n_\beta)G^2 + R_2{}^1\, g^1 + R_2{}^3\, g^3 \tag{5.5}$$

and

$$[E - \omega_\beta - \omega_\alpha - R_3{}^3]g^3 = 6V^{(3)}_{\alpha,0,-\beta}(n_\alpha + n_\beta + 1)G^1$$

$$+ 6V^{(3)}_{\alpha,0,-\beta}(n_\alpha + n_\beta + 1)G^2 + R_3{}^1\, g^1 + R_3{}^2\, g^2\ . \tag{5.6}$$

The details of the decoupling and expressions for the R functions are given in Appendix B. The susceptibility is

$$^{4}\chi_{\mu\nu}^{11}(\omega) = N \sum_{0} M_{\mu}(0)\, M_{\nu}(0) \left(\frac{1}{\omega_0^{\,2} - \omega^2 + 2\omega_0[\sum(0) + {}^{4}\gamma^{11}(n,E)]} \right) \tag{5.7}$$

where

$$^{4}\gamma^{11}(n,E) = 18 \sum_{\alpha,\beta} \left| V^{(3)}_{-\alpha,0,\beta} \right|^2 \left(\frac{n_\alpha + n_\beta + 1}{E - \omega_\alpha - \omega_\beta - R_3^{\,3}} \right.$$

$$\left. - \frac{n_\alpha + n_\beta + 1}{E + \omega_\alpha + \omega_\beta + R_1^{\,1}} + \frac{2(n_\alpha - n_\beta)}{E + \omega_\alpha - \omega_\beta - R_2^{\,2}} \right) \quad . \tag{5.8}$$

The frequency shifts, $^{4}\Delta^{11}(n,\omega)$, and damping coefficient, $^{4}\Gamma^{11}(n,\omega)$, can be obtained in the usual manner. The expression for the damping coefficient is

$$^{4}\Gamma^{11}(n,\omega) = 18\pi \sum_{\alpha,\beta} \left| V^{(3)}_{-\alpha,0,\beta} \right|^2 \{(n_\alpha + n_\beta + 1)[\delta(\omega - \omega_\alpha - \omega_\beta - R_3^{\,3})$$

$$- \delta(\omega + \omega_\alpha + \omega_\beta + R_1^{\,1})] + 2(n_\alpha - n_\beta)\, \delta(\omega + \omega_\alpha - \omega_\beta - R_2^{\,2})\} \ . \tag{5.9}$$

The damping coefficient for $^{4}\chi_{\mu\nu}^{12}(\omega)$ has the same form as that given in Eq.(5.9).

By use of this approach, the introduction of H_4 leads to an expression for the damping coefficient which is different from the previously obtained T^2 (classical limit) expression. The essential difference between $^{3}\Gamma^{11}$ and $^{4}\Gamma^{11}$ is the presence of the R

functions in the arguments of the δ-functions in Eq. (5.9). The R functions have explicit temperature dependence.

B. Two-Phonon Processes: ${}^4\chi^{21}_{\mu\nu}(\omega)$ AND ${}^4\chi^{22}_{\mu\nu}(\omega)$

The expression for the damping coefficient associated with ${}^4\chi^{21}_{\mu\nu}(\omega)$ is the same as that for ${}^4\chi^{22}_{\mu\nu}(\omega)$. However, it is important to note that ${}^4\chi^{21}_{\mu\nu}(\omega)$ is at least a factor of $N^{1/2}$ larger than ${}^4\chi^{22}_{\mu\nu}(\omega)$. The equations for L^1, L^2, and L^3 to quartic terms in the Hamiltonian are

$$(E + \omega_{\alpha'} + \omega_{\delta'})L^1 = G([a^+_{-\alpha'}, a^+_{\delta'}, H_3]; A_{0'})_E + G([a^+_{-\alpha'}, a^+_{\delta'}, H_4]; A_{0'})_E \tag{5.14}$$

$$(E + \omega_{\alpha'} - \omega_{\delta'})L^2 = G([a^+_{-\alpha'}, a_{-\delta'}, H_3]; A_{0'})_E + G([a^+_{-\alpha'}, a_{-\delta'}, H_4]; A_{0'})_E \tag{5.15}$$

and

$$(E - \omega_{\alpha'} - \omega_{\delta'})L^3 = G([a_{\alpha'}, a_{-\delta'}, H_3]; A_{0'})_E + G([a_{\alpha'}, a_{-\delta'}, H_4]; A_{0'})_E \quad . \tag{5.16}$$

We must reduce the H_4 terms to the original type Green's functions and write equations (to quartic terms in the Hamiltonian) for the higher-order Green's functions generated by the H_3 terms. The equations for the L Green's functions become

$$(E + \omega_{\alpha'} + \omega_{\delta'} + \tilde{R}_1^{\,1})L^1 = G([a^+_{-\alpha'}, a^+_{\delta'}, H_3]; A_{0'})_E - (\tilde{R}_1^{\,2} L^2 + \tilde{R}_1^{\,3} L^3) \tag{5.17}$$

$$(E + \omega_{\alpha'} - \omega_{\delta'} - \tilde{R}_2^{\,2})L^2 = G([a^+_{-\alpha'}, a_{-\delta'}, H_3]; A_{0'})_E \quad \tilde{R}_2^{\,1} L^1 + \tilde{R}_2^{\,3} L^3 \tag{5.18}$$

and

$$(E - \omega_{\alpha'} - \omega_{\delta'} - \tilde{R}_3^{\,3})L^3 = G([a_{\alpha'}, a_{-\delta'}, H_3]; A_{0'})_E \quad \tilde{R}_3^{\,1} L^1 + \tilde{R}_3^{\,2} L^2 \tag{5.19}$$

where

$$\tilde{R}_1^{\cdots} = R_1^{\cdots}\Big|_{\substack{\alpha \to \delta' \\ \beta \to \alpha'}}, \quad \tilde{R}_2^{\cdots} = R_2^{\cdots}\Big|_{\substack{\alpha \to -\alpha' \\ \beta \to -\delta'}}, \quad \text{and}$$

$$\tilde{R}_3^{\cdots} = R_3^{\cdots}\Big|_{\substack{\alpha \to \delta' \\ \beta \to \alpha'}} .$$

The resolution of the higher-order Green's functions in Eqs. (5.17), (5.18), and (5.19) is accomplished in a manner similar to that used in Appendix A. The required change is the addition of a term involving H_4 to the equations for the I generator Green's functions. The reduction of these H_4 terms in the equations for the I Green's functions gives rise to a damping coefficient of the same form as in Eq. (5.9). That is to say, the damping coefficient for ${}^4\chi^{21}_{\mu\nu}(\omega)$ and ${}^4\chi^{22}_{\mu\nu}(\omega)$ contains an explicit temperature dependent argument

for the accompanying δ-functions. It is therefore clear that the important contribution of H_4 to the damping coefficient is ${}^4\Gamma^{11}(n,\omega)$ since ${}^4\chi^{11}_{\mu\nu}(\omega)$ is at least a factor $N^{1/2}$ larger than ${}^4\chi^{21}_{\mu\nu}(\omega)$ and at least a factor N larger than ${}^4\chi^{22}_{\mu\nu}(\omega)$.

VI. DISCUSSION

In this paper, we have (a) presented the details of a first-order self-consistent method for developing a closed system of Green-function equations, (b) used this method to show that the contributions of M^2 and H_4 to the damping coefficient can be treated in a way that is consistent with lower-order calculations, (c) made unnecessary discussions concerning which diagrams to include or exclude, (d) circumvented the inherent difficulty associated with the use of arbitrary expansion parameters in the Hamiltonian, and (e) demonstrated the impact on the final results of each approximation made.

The main results are (a) the derivations of the expressions for the damping coefficients associated with ${}^3\chi_{\mu\nu}^{22}(\omega)$ and ${}^4\chi_{\mu\nu}^{11}(\omega)$ and (b) the comprehensive analysis of M^2 and H_4 contributions to the damping coefficient. It is shown that the damping coefficients associated with two-phonon processes are much smaller than those for one-phonon processes. The derived expressions for the various damping coefficients are valid for the whole temperature range. The expression for ${}^4\Gamma^{11}(n,\omega)$ contains δ-functions with explicit temperature dependent arguments.

In the classical limit, we find that ${}^3\Gamma^{11}(n,\omega)$ is proportional to $|V^{(3)}|^2\ T$ which is the same as previously derived results.[23,24] However, our expression for ${}^4\Gamma^{11}(n,\omega)$ is proportional to $(|V^{(3)}|^2/|V^{(4)}|^2)\frac{1}{T}$ which differs from the previously obtained T^2 dependent expression in the classical limit.[6,7] Our result for the quartic contribution is therefore in qualitative agreement with Mooij's experimental results for kBr.

If the Green-function equations are decoupled so that a T^2 (classical limit, quartic contribution) expression is forced, it has been shown that this contribution is inconsistent with the order of the approximation used in the overall calculation.[10] Moreover, this contribution has been shown to be negligible in comparison with the cubic contribution.[10]

APPENDIX A.

This Appendix is devoted to the details leading to the solution of the equations for the L Green's functions. The higher-order Green's functions in Eqs. (4.23), (4.24), and (4.25) reduce to

$$G([a^+_{-\alpha'}, a^+_{\delta'}, H_3]; A_{0'})_E = -6 \sum_{\alpha,\beta} V^{(3)}_{\alpha,\beta,\delta'}(\ell^1 + \ell^2 + \ell^3 + \ell^4) \tag{A.1}$$

$$G([a^+_{-\alpha'}, a_{-\delta'}, H_3]; A_{0'})_E = -3 \sum_{\alpha,\beta} V^{(3)}_{\alpha,\beta,\delta'}\{(\ell^5 + \ell^6 + \ell^7 + \ell^8) - (\ell^1 + \ell^2 + \ell^3 + \ell^4)\} \tag{A.2}$$

$$G([a_{\alpha'}, a_{-\delta'}, H_3]; A_{0'})_E = 6 \sum_{\alpha,\beta} V^{(3)}_{\alpha,\beta,\delta'}(\ell^5 + \ell^6 + \ell^7 + \ell^8) . \tag{A.3}$$

The ℓ higher order Green's functions are given by

$$\left.\begin{aligned} \ell^1 &= G(a^+_{-\alpha}\, a^+_{-\alpha'}, a^+_{-\beta}; A_{0'})_E; & \ell^2 &= G(a^+_{-\alpha'}, a_\beta\, a^+_{-\alpha}; A_{0'})_E \\ \ell^3 &= G(a^+_{-\beta}\, a_\alpha\, a^+_{-\alpha'}; A_{0'})_E; & \ell^4 &= G(a_\alpha\, a^+_{-\alpha'}, a_\beta; A_{0'})_E \end{aligned}\right\} \tag{A.4}$$

and

$$\left.\begin{aligned} \ell^5 &= G(a^+_{-\alpha}\, a_{\alpha'}, a^+_{-\beta}; A_{0'})_E; & \ell^6 &= G(a_\beta\, a^+_{-\alpha}\, a_{\alpha'}; A_{0'})_E \\ \ell^7 &= G(a_{\alpha'}, a^+_{-\beta}\, a_\alpha; A_{0'})_E; & \ell^8 &= G(a_\alpha\, a_{\alpha'}, a_\beta; A_{0'})_E \end{aligned}\right\} . \tag{A.5}$$

The solution of the equations for the ℓ Green's functions can be generated from the solution of the equations for four generator Green's functions. The generator Green's functions will be denoted by I; their equations are

$$(E + \omega_1 + \omega_2 + \omega_3)I^1 = G([a_1^+ a_2^+ a_3^+, H_3]; A_{0'})_E \tag{A.6}$$

$$(E + \omega_1 + \omega_3 - \omega_2)I^2 = \frac{\langle [I^2] \rangle_E}{2\pi} + G([a_1^+ a_2 a_3^+, H_3]; A_{0'})_E \tag{A.7}$$

$$(E + \omega_2 - \omega_1 - \omega_3)I^3 = \frac{\langle [I^3] \rangle_E}{2\pi} + G([a_1 a_2^+ a_3, H_3]; A_{0'})_E \tag{A.8}$$

and

$$(E - \omega_1 - \omega_2 - \omega_3)I^4 = G([a_1 a_2 a_3, H_3]; A_{0'})_E \tag{A.9}$$

where

$$\langle [I^2] \rangle_E = -[(n_3 + 1)\delta_{3,2}\, \delta_{0',1} + n_1\, \delta_{1,2}\, \delta_{0',3}] \tag{A.10}$$

and

$$\langle [I^3] \rangle_E = n_2\, \delta_{2,3}\, \delta_{0',1} + n_2\, \delta_{2,1}\, \delta_{0',3} \quad . \tag{A.11}$$

The decoupling of the higher-order Green's functions in the equations for the I Green's functions leads to the following results:

$$G([a_1^+ a_2^+ a_3^+, H_3]; A_{0'})_E \approx f^{31} L^1 + f^{32} L^2 \tag{A.12}$$

where

$$f^{31} = -3[V^{(3)}_{\alpha',1,2}(n_1 + n_2 + \tfrac{2}{3})\delta_{3,\delta'} + V^{(3)}_{\alpha',1,3}(n_1 + n_3 + \tfrac{2}{3})\delta_{2,\delta'}$$

$$+ V^{(3)}_{2,1,-\delta'}(n_1 + n_2 + \tfrac{4}{3})\delta_{3,-\alpha'} + V^{(3)}_{3,1,-\delta'}(n_1 + n_3 + \tfrac{4}{3})\delta_{2,-\alpha'}$$

$$+ V^{(3)}_{\alpha',2,3}(n_2 + n_3 + \tfrac{2}{3})\delta_{1,\delta'} + V^{(3)}_{3,2,-\delta'}(n_2 + n_3 + \tfrac{4}{3})\delta_{1,-\alpha'}] \quad \text{(A.13)}$$

and

$$f^{32} = -3[V^{(3)}_{1,2,-\delta'}(n_1 + n_2 + \tfrac{4}{3})\delta_{3,-\alpha'} + V^{(3)}_{3,1,-\delta'}(n_1 + n_3 + \tfrac{4}{3})\delta_{2,-\alpha'}$$

$$+ V^{(3)}_{3,2,-\delta'}(n_2 + n_3 + \tfrac{4}{3})\delta_{1,-\alpha'} + V^{(3)}_{\alpha',2,1}(n_1 + n_2 + \tfrac{2}{3})\delta_{3,\delta'}$$

$$+ V^{(3)}_{\alpha',3,1}(n_1 + n_3 + \tfrac{2}{3})\delta_{2,\delta'} + V^{(3)}_{\alpha',3,2}(n_2 + n_3 + \tfrac{2}{3})\delta_{1,\delta'}] \quad \text{(A.14)}$$

$$G([a_1^+ a_2 a_3^+, H_3]; A_{0'})_E \approx f^{21} L^1 + f^{22} L^2 + f^{23} L^3 \quad \text{(A.15)}$$

where

$$f^{21} = -3[V^{(3)}_{\alpha',1,-2}(n_2 - n_1 + \tfrac{1}{3})\delta_{3,\delta'} + V^{(3)}_{1,-2,-\delta'}(n_2 - n_1 - \tfrac{1}{3})\delta_{3,-\alpha'}$$

$$+ V^{(3)}_{\alpha',1,-\delta'}(n_3 + 1)\delta_{2,3} + V^{(3)}_{\alpha',-2,3}(n_2 - n_3 + \tfrac{1}{3})\delta_{1,\delta'}$$

$$+ V^{(3)}_{3,-2,-\delta'}(n_2 - n_3 - \tfrac{1}{3})\delta_{1,-\alpha'} + V^{(3)}_{\alpha',3,-\delta'}\, n_1\, \delta_{1,2}] \quad \text{(A.16)}$$

$$f^{22} = -3[V^{(3)}_{1,-2,-\delta'}(n_2 - n_1 - \tfrac{1}{3})\delta_{3,-\alpha'} + V^{(3)}_{\alpha',1,3}(n_1 + n_3 + \tfrac{2}{3})\delta_{2,-\delta'}$$

$$+ V^{(3)}_{\alpha',1,-\delta'}\, 2(n_3 + 1)\delta_{2,3} + V^{(3)}_{3,-2,-\delta'}(n_2 - n_3 - \tfrac{1}{3})\delta_{1,-\alpha'}$$

$$+ V^{(3)}_{\alpha',1,-2}(n_2 - n_1 - \tfrac{1}{3})\delta_{3,\delta'} + V^{(3)}_{\alpha',3,-2}(n_2 - n_3 - \tfrac{1}{3})\delta_{1,\delta'}$$

$$+ V^{(3)}_{1,3,-\delta'}(n_1 + n_3 + \tfrac{4}{3})\delta_{2,\alpha'} + 2V^{(3)}_{\alpha',3,-\delta'}\, n_1\, \delta_{1,2}] \qquad (A.17)$$

and

$$f^{23} = -3[V^{(3)}_{3,1,-\delta'}(n_1 + n_3 + \tfrac{4}{3})\delta_{2,\alpha'} + V^{(3)}_{\alpha',1,3}(n_1 + n_3)\delta_{2,-\delta'}$$

$$+ V^{(3)}_{\alpha',3,-\delta'}\, n_1\, \delta_{1,2} + V^{(3)}_{\alpha',1,-\delta'}(n_3 + 1)\delta_{2,3}] \qquad (A.18)$$

$$G([a_1\, a_2^+\, a_3,\, H_3];\, A_{0'})_E \approx f^{11}\, L^1 + f^{12}\, L^2 + f^{13}\, L^3 \qquad (A.19)$$

where

$$f^{11} = 3[V^{(3)*}_{1,3,\delta'}(n_1 + n_3)\delta_{2,-\alpha'} - 2V^{(3)*}_{\alpha',3,1}\, \delta_{2,\delta'}$$

$$+ V^{(3)}_{\alpha',-1,-3}(n_1 + n_2 + 2)\delta_{2,\delta'} + \left(V^{(3)}_{3,-1,-\delta'} + V^{(3)}_{1,-3,-\delta'}\right)\delta_{2,\alpha'}$$

$$+ V^{(3)}_{\alpha',-1,-\delta'}\, n_2\, \delta_{2,3} + V^{(3)}_{\alpha',-3,-\delta'}(n_2 + \tfrac{1}{3})\delta_{1,2}] \qquad (A.20)$$

$$f^{12} = 3[V^{(3)*}_{1,3,\delta'}(n_1 + n_3 + \tfrac{2}{3})\delta_{2,-\alpha'} + V^{(3)}_{\alpha',-1,2}(n_2 - n_1 - \tfrac{1}{3})\delta_{3,-\delta'}$$

$$+ 2V^{(3)}_{\alpha',-1,-\delta'}\, n_2\, \delta_{2,3} + V^{(3)}_{\alpha',2,-3}(n_2 - n_3 - \tfrac{1}{3})\delta_{1,-\delta'}$$

$$+ V^{(3)}_{\alpha',-3,-\delta'}\, 2(n_2 + \tfrac{1}{3})\delta_{1,2} + V^{(3)}_{2,-1,-\delta'}(n_2 - n_1 + \tfrac{1}{3})\delta_{3,\alpha'}$$

$$+ V^{(3)}_{2,-3,-\delta'}(n_2 - n_3 + \tfrac{1}{3})\delta_{1,\alpha'} + V^{(3)}_{\alpha',-3,-1}(n_1 + n_3 + \tfrac{4}{3})\delta_{2,\delta'}] \quad \text{(A.21)}$$

and

$$f^{13} = 3[V^{(3)}_{2,-1,-\delta'}(n_2 - n_1 + 1)\delta_{3,\alpha'} + V^{(3)}_{2,-3,-\delta'}(n_2 - n_3 + 1)\delta_{1,\alpha'}$$

$$+ V^{(3)}_{\alpha',-3,2}(n_2 - n_3 - \tfrac{1}{3})\delta_{1,-\delta'} + V^{(3)}_{\alpha',-1,2}(n_2 - n_1 - \tfrac{1}{3})\delta_{3,-\delta'}$$

$$+ V^{(3)}_{\alpha',-3,-\delta'}(n_2 + \tfrac{1}{3})\delta_{1,2} + V^{(3)}_{\alpha',-1,-\delta'}\, n_2\, \delta_{2,3}] \quad . \qquad \text{(A.22)}$$

The decoupling of the final generator Green's function is

$$G([a_1\, a_2\, a_3,\, H_3];\, A_{0'})_E \approx f^{02}\, L^2 + f^{03}\, L^3 \qquad \text{(A.23)}$$

where

$$f^{02} = 3[V^{(3)}_{\alpha',-1,-2}(n_1 + n_2 + \tfrac{4}{3})\delta_{3,-\delta'} + V^{(3)}_{\alpha',-2,-3}(n_2 + n_3 + \tfrac{4}{3})\delta_{1,-\delta'}$$

$$+ V^{(3)}_{\alpha',-1,-3}(n_1 + n_3 + \tfrac{4}{3})\delta_{2,-\delta'} + V^{(3)*}_{2,1,\delta'}(n_1 + n_2 + \tfrac{2}{3})\delta_{3,\alpha'}$$

$$+ V^{(3)*}_{3,1,\delta'}(n_1 + n_3 + \tfrac{2}{3})\delta_{2,\alpha'} + V^{(3)*}_{3,2,\delta'}(n_2 + n_3 + \tfrac{2}{3})\delta_{1,\alpha'}] \qquad \text{(A.24)}$$

and

$$f^{03} = 3[V^{(3)*}_{1,2,\delta'}(n_1 + n_2 + \tfrac{2}{3})\delta_{3,\alpha'} + V^{(3)*}_{3,1,\delta'}(n_1 + n_3 + \tfrac{2}{3})\delta_{2,\alpha'}$$

$$+ V^{(3)*}_{3,2,\delta'}(n_2 + n_3 + \tfrac{2}{3})\delta_{1,\alpha'} + V^{(3)}_{\alpha',-1,-2}(n_1 + n_2 + \tfrac{4}{3})\delta_{3,-\delta'}$$

$$+ V^{(3)}_{\alpha',-1,-3}(n_1 + n_3 + \tfrac{4}{3})\delta_{2,-\delta'} + V^{(3)}_{\alpha',-3,-2}(n_2 + n_3 + \tfrac{4}{3})\delta_{1,-\delta'}] \, . \tag{A.25}$$

We are now in a position to obtain the solution of the equations for the generator, I, Green's functions. Extreme care must be exercised when dealing with the coefficients of L^1 and L^3. In these cases, one may obtain an apparent zero because of the symmetry properties of the $V^{(3)}$ coefficients. This situation may be resolved by interchanging an index in $V^{(3)}$ with an index in the accompanying δ-function. The solutions of the equations for the I's are

$$I^1 = f^{31}(1,2,3)L^1 + f^{32} L^2 \tag{A.26}$$

$$I^2 = d^2(1,2,3) + f^{21}(1,2,3)L^1 + f^{22}(1,2,3)L^2 + f^{23}(1,2,3)L^3 \tag{A.27}$$

$$I^3 = d^3(1,2,3) + f^{11}(1,2,3)L^1 + f^{12}(1,2,3)L^2 + f^{13}(1,2,3)L^3 \tag{A.28}$$

$$I^4 = f^{02}(1,2,3)L^2 + f^{03}(1,2,3)L^3 \tag{A.29}$$

where

$$f^{31}(1,2,3) = \frac{f^{31}}{E + \omega_1 + \omega_2 + \omega_3} .$$

The other $f^{\cdots}(\cdots)$ are defined in a similar manner. The d's are defined by

$$d^2(1,2,3) = \frac{\frac{1}{2\pi}\langle[I^2]\rangle_E}{E + \omega_1 + \omega_3 - \omega_2} \quad \text{and} \quad d^3(1,2,3) = \frac{\frac{1}{2\pi}\langle[I^3]\rangle_E}{E + \omega_2 - \omega_1 - \omega_3}.$$

Equations (4.23), (4.24), and (4.25) now become

$$[E + \omega_{\alpha'} + \omega_{\delta'} + {}^3\gamma_1^{21}(n,E)]L^1 = -D^1 - F^{12} L^2 - F^{13} L^3 \qquad \text{(A.30)}$$

where

$$^3\gamma_1^{21}(n,E) = 6 \sum_{\alpha,\beta} V^{(3)}_{\alpha,\beta,\delta'}[f^{31}(-\alpha, -\alpha', -\beta) + f^{21}(-\alpha', \beta, -\alpha)$$

$$+ f^{21}(-\beta, \alpha, -\alpha') + f^{11}(\alpha, -\alpha', \beta)] \qquad \text{(A.31)}$$

$$F^{12} = 6 \sum_{\alpha,\beta} V^{(3)}_{\alpha,\beta,\delta'}[f^{32}(-\alpha, -\alpha', -\beta) + f^{22}(-\alpha', \beta, -\alpha)$$

$$+ f^{22}(-\beta,\alpha,-\alpha') + f^{12}(\alpha, -\alpha', \beta)] \qquad \text{(A.32)}$$

$$F^{13} = 6 \sum_{\alpha,\beta} V^{(3)}_{\alpha,\beta,\delta'}[f^{23}(-\alpha', \beta, -\alpha) + f^{23}(-\beta, \alpha, -\alpha')$$

$$+ f^{13}(\alpha, -\alpha', \beta)] \qquad (A.33)$$

and

$$D^{1} = 6 \sum_{\alpha,\beta} V^{(3)}_{\alpha,\beta,\delta'}[d^{2}(-\alpha', \beta, -\alpha) + d^{2}(-\beta, \alpha, -\alpha')$$

$$+ d^{3}(\alpha, -\alpha', \beta)] \quad . \qquad (A.34)$$

The equation for L^2 is

$$[E + \omega_{\alpha'} - \omega_{\delta'} + {}^{3}\gamma_{2}^{21}(n,E)]L^{2} = -D^{2} - F^{21} L^{1} - F^{23} L^{3} \qquad (A.35)$$

where

$${}^{3}\gamma_{2}^{21}(n,E) = 3 \sum_{\alpha,\beta} V^{(3)}_{\alpha,\beta,\delta'}[f^{22}(-\alpha, \alpha', -\beta) + f^{12}(\beta, -\alpha, \alpha')$$

$$+ f^{12}(\alpha', -\beta, \alpha) + f^{02}(\alpha, \alpha', \beta) - f^{32}(-\alpha, -\alpha', -\beta)$$

$$- f^{22}(-\alpha', \beta, -\alpha) - f^{22}(-\beta, \alpha, -\alpha') - f^{12}(\alpha, -\alpha', \beta)] \qquad (A.36)$$

$$F^{21} = 3 \sum_{\alpha,\beta} V^{(3)}_{\alpha,\beta,\delta'}[f^{21}(-\alpha, \alpha', -\beta) + f^{11}(\beta, -\alpha, \alpha')$$

$$+ f^{11}(\alpha', -\beta, \alpha) - f^{31}(-\alpha, -\alpha', -\beta) - f^{21}(-\alpha', \beta, -\alpha)$$

$$- f^{21}(-\beta, \alpha, -\alpha') - f^{11}(\alpha, -\alpha', \beta)] \qquad \text{(A.37)}$$

$$F^{23} = 3 \sum_{\alpha,\beta} V^{(3)}_{\alpha,\beta,\delta'}[f^{23}(-\alpha, \alpha', -\beta) + f^{13}(\beta, -\alpha, \alpha')$$

$$+ f^{13}(\alpha', -\beta, \alpha) + f^{03}(\alpha, \alpha', \beta) - f^{23}(-\alpha', \beta, -\alpha)$$

$$- f^{23}(-\beta, \alpha, -\alpha') - f^{13}(\alpha, -\alpha', \beta)] \qquad \text{(A.38)}$$

and

$$D^2 = 3 \sum_{\alpha,\beta} V^{(3)}_{\alpha,\beta,\delta'}[d^2(-\alpha, \alpha', -\beta) + d^3(\beta, -\alpha, \alpha')$$

$$+ d^3(\alpha', -\beta, \alpha) - d^2(-\alpha', \beta, -\alpha) - d^2(-\beta, \alpha, -\alpha') - d^3(\alpha, -\alpha', \beta)]. \qquad \text{(A.39)}$$

The L^3 equation reduces to

$$[E - \omega_{\alpha'} - \omega_{\delta'} - {}^3\gamma_3^{21}(n,E)]L^3 = -D^3 - F^{31} L^1 - F^{32} L^2 \qquad \text{(A.40)}$$

where

$$ {}^{3}\gamma_{3}^{21}(n,E) = 6 \sum_{\alpha,\beta} V^{(3)}_{\alpha,\beta,\delta'}[f^{23}(-\alpha, \alpha', -\beta) + f^{13}(\beta, -\alpha, \alpha') $$

$$ + f^{13}(\alpha', -\beta, \alpha) + f^{03}(\alpha, \alpha', \beta)] \qquad (A.41) $$

$$ F^{31} = -6 \sum_{\alpha,\beta} V^{(3)}_{\alpha,\beta,\delta'}[f^{21}(-\alpha, \alpha', -\beta) + f^{11}(\beta, -\alpha, \alpha') $$

$$ + f^{11}(\alpha', -\beta, \alpha)] \qquad (A.42) $$

$$ F^{32} = -6 \sum_{\alpha,\beta} V^{(3)}_{\alpha,\beta,\delta'}[f^{22}(-\alpha, \alpha', -\beta) + f^{12}(\beta, -\alpha, \alpha') $$

$$ + f^{12}(\alpha', -\beta, \alpha) + f^{02}(\alpha, \alpha', \beta)] \qquad (A.43) $$

and

$$ D^{3} = -6 \sum_{\alpha,\beta} V^{(3)}_{\alpha,\beta,\delta'}[d^{2}(-\alpha, \alpha', -\beta) + d^{3}(\beta, -\alpha, \alpha') $$

$$ + d^{3}(\alpha', -\beta, \alpha)] \, . \qquad (A.44) $$

The required expression for the combination of L's is

$$L^1 + 2L^2 + L^3 = -\left(\frac{D^1}{X_1} + \frac{2D^2}{X_2} + \frac{D^3}{X_3}\right) + \frac{2D^1 F^{21} + D^2 F^{21}}{X_1 X_2}$$

$$+ \frac{D^1 F^{31} + D^3 F^{13}}{X_1 X_3} + \frac{D^2 F^{32} + 2D^3 F^{23}}{X_2 X_3} \qquad (A.45)$$

where

$$X_1 = E + \omega_{\alpha'} + \omega_{\delta'} + {}^3\gamma_1^{21}(n,E)$$

$$X_2 = E + \omega_{\alpha'} - \omega_{\delta'} + {}^3\gamma_2^{21}(n,E)$$

and

$$X_3 = E - \omega_{\alpha'} - \omega_{\delta'} - {}^3\gamma_3^{21}(n,E) \quad .$$

The method for solving the equations for the H-type Green's functions is similar to that for the L-type Green's functions. The results are

$$[E + \omega_{\alpha'} + \omega_{\delta'} + {}^3\gamma_1^{21}(n,E)]H^1 = -C^1 - F^{12} H^2 - F^{13} H^3 \qquad (A.46)$$

$$[E + \omega_{\alpha'} - \omega_{\delta'} + {}^3\gamma_2^{21}(n,E)]H^2 = -C^2 - F^{21} H^1 - F^{23} H^3 \qquad (A.47)$$

$$[E - \omega_{\alpha'} - \omega_{\delta'} - {}^3\gamma^{21}(n,E)]H^3 = -C^3 - F^{31} H^1 - F^{32} H^2 \qquad (A.48)$$

where

$$C^1 = -\frac{1}{2\pi}\langle[H^1]\rangle_E , \qquad C^2 = -\frac{1}{2\pi}\langle[H^2]\rangle_E, \quad \text{and}$$

$$C^3 = -\frac{1}{2\pi}\langle[H^3]\rangle_E .$$

On solving the equations for the H Green's functions, we obtain

$$H^1 + 2H^2 + H^3 = -\left(\frac{C^1}{X_1} + \frac{2C^2}{X_2} + \frac{C^3}{X_3}\right) + \frac{2C^1 F^{21} + C^2 F^{12}}{X_1 X_2}$$

$$+ \frac{C^1 F^{31} + C^3 F^{13}}{X_1 X_3} + \frac{C^2 F^{32} + 2C^3 F^{23}}{X_2 X_3} \quad . \qquad \text{(A.49)}$$

APPENDIX B.

Here we present the details of the decoupling of higher-order Green's functions generated by H_4. For the one-phonon cases in Eqs. (5.1) and (5.2), we obtain

$$G([a_0^+, H_4]; A_{0'})_E \approx - \sum(0)(G^1 + G^2) \tag{B.1}$$

and

$$G([a_0, H_4]; A_{0'})_E \approx \sum(0)(G^1 + G^2) \tag{B.2}$$

where

$$\sum(0) = 12 \sum_{\alpha} V^{(4)}_{\alpha,-\alpha,0,0}(2n_\alpha + 1) . \tag{B.3}$$

The equations for the g-type Green's functions require the following three decouplings:

$$G([a_\alpha^+ a_{-\beta}^+, H_4]; A_{0'})_E \approx -[R_1^{\ 1} g^1 + R_1^{\ 2} g^2 + R_1^{\ 3} g^3] \tag{B.4}$$

$$G([a_\alpha^+ a_\beta, H_4]; A_{0'})_E \approx R_2^{\ 1} g^1 + R_2^{\ 2} g^2 + R_2^{\ 3} g^3 \tag{B.5}$$

and

$$G([a_{-\alpha} a_\beta, H_4]; A_{0'})_E \approx R_3^{\ 1} g^1 + R_3^{\ 2} g^2 + R_3^{\ 3} g^3 \tag{B.6}$$

where

$$R_1^{\ 1} = 24 \sum_{\alpha'} V^{(4)}_{\alpha',-\alpha',\alpha,-\alpha}(2n_{\alpha'} + 1) + 4V^{(4)}_{\alpha,-\alpha,\beta,-\beta}(2n_\alpha + 3n_\beta + 1) \tag{B.7}$$

$$R_1^{\ 2} = 24 \sum_{\alpha'} V^{(4)}_{\alpha',-\alpha',\beta,-\beta}(2n_{\alpha'} + 1) + V^{(4)}_{\alpha,-\alpha,\beta,-\beta}(n_\alpha + n_\beta + 1) \tag{B.8}$$

$$R_2^{\ 1} = 12 \sum_{\alpha'} V^{(4)}_{\alpha',-\alpha',\beta,-\beta}(2n_{\alpha'} + 1) + V^{(4)}_{\alpha,-\alpha,\beta,-\beta}(8n_\alpha - 12n_\beta - 2) \tag{B.9}$$

$$R_2^{\ 2} = 12 \sum_{\alpha'} V^{(4)}_{\alpha',-\alpha',\beta,-\beta}(2n_{\alpha'} + 1) - 12 \sum_{\alpha'} V^{(4)}_{\alpha',-\alpha',\alpha,-\alpha}(2n_{\alpha'} + 1)$$

$$+ V^{(4)}_{\alpha,-\alpha,\beta,-\beta}(18n_\alpha - 24n_\beta + 4) \tag{B.10}$$

$$R_3^{\ 3} = - \sum_{\alpha'} V^{(4)}_{\alpha',-\alpha',\alpha,-\alpha}(28n_{\alpha'} + 13) + V^{(4)}_{\alpha,-\alpha,\beta,-\beta}(12n_\alpha - 16n_\beta + 2) \tag{B.11}$$

$$R_3^{\ 1} = R_1^{\ 3} \tag{B.12}$$

$$R_3^{\ 2} = \sum_{\alpha'} V^{(4)}_{\alpha',-\alpha',\alpha,-\alpha}(46n_{\alpha'} + 24)$$

$$+ V^{(4)}_{\alpha,-\alpha,\beta,-\beta}(22n_\alpha + 24n_\beta + 13) \tag{B.13}$$

and

$$R_3^{\,3} = 25 \sum_{\alpha'} V^{(4)}_{\alpha',-\alpha',\alpha,-\alpha}(2n_{\alpha'} + 1)$$

$$+ V^{(4)}_{\alpha,-\alpha,\beta,-\beta}(12n_\alpha + 16n_\beta + 14) \; . \qquad \text{(B.14)}$$

FOOTNOTES AND REFERENCES

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