

Theory for the first-order vibrational spectra of disordered solids

F. L. Galeener and P. N. Sen*

Xerox Palo Alto Research Center, Palo Alto, California 94304

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Expressions are derived in the harmonic approximation which relate the first-order Raman and infrared spectra of a disordered solid to a common set of vibrational densities of states. The calculations assume no periodicity, and using standard approximations lead to expressions involving explicitly defined matrix elements. The results extend those of Shuker and Gammon from Raman to infrared response, and show that the matrix elements involved must be frequency dependent.

I. INTRODUCTION

In 1970 Shuker and Gammon¹ (SG) presented a calculation which related the Raman scattering intensity $I(\omega)$ of an amorphous material to the density of states $\rho_b(\omega)$ in various vibrational subbands b . Their expression for the Stokes intensity may be written in the form

$$I(\omega) = [n(\omega, T) + 1] \omega^{-1} \sum_b C_b \rho_b(\omega),$$

where $n(\omega, T) = [\exp(\hbar\omega/kT) - 1]^{-1}$ is the Bose-Einstein occupation number, ω is the magnitude of the Raman frequency shift, and C_b is a polarization-dependent coupling constant.² Experimentalists have found the SG result extremely useful for producing a reduced Raman spectrum³ $(\omega[n(\omega, T) + 1]^{-1}I(\omega))$ free of spurious structure due to thermal population effects. Nevertheless, some deficiencies have become evident. For example, Kobliska and Solin⁴ have noted the absence of an expected fourth-power dependence on ω_s , the frequency of the scattered photon. The SG method provides no procedure for theoretically investigating the magnitude of the C_b or their assumed lack of dependence on ω . This situation comes about because SG make an *ad hoc* assumption that the product $A(j)\Lambda_j^3$ is frequency independent over all the modes j of a subband, where $A(j)$ is the "optical-coupling tensor" and Λ_j^3 is the "mode volume." It automatically follows from this assumption that the C_b are frequency independent. Perhaps more importantly, from a conceptual point of view, SG proceed by analogy with the crystalline case and define the vibrational eigenmodes to be exponentially decaying *periodic* plane waves involving an unknown correlation length Λ_j which must be assumed to be quite small.⁵

In this paper we outline the steps in a first-principles calculation which we believe avoids these limitations and assumptions. Our intention is to improve upon the SG result while retaining

its partition of vibrational modes into "bands"; to set it on a firmer mathematical foundation; and to derive an analogous result for application to infrared (ir) data. We obtain explicit expressions of a similar form for both the first-order Raman scattering intensity I and for the imaginary part of the ir dielectric constant ϵ_2 . We find in both cases that the coupling coefficients may be strongly frequency dependent, and that their frequency dependence follows from that of the vibrational eigenvectors of the unperturbed system. The method also leads naturally to calculation of second-order spectra and treatment of anharmonic perturbations, as will be elaborated elsewhere.

II. VIBRATIONAL PROBLEM

From the beginning we assume the complete absence of long-range order in atomic positions. Since there is therefore no periodic reciprocal space, we do not attempt to label eigenstates with a "quantized" reciprocal-space vector of the sort encountered in studies of crystalline materials. In effect, the sample is treated as a giant molecule having no translational or rotational symmetry and consisting of N atoms, each vibrating harmonically about its own equilibrium position.⁶ At first sight it may seem foolhardy to treat such a large system; however, in this way we obtain results whose generality has not been masked by the imposition of unnecessary assumptions. These results allow for subsequent practical approximations and treatment of specific structures.

The Hamiltonian of this system is

$$H = \frac{1}{2} \sum_i \mu_i U_i^2 + \frac{1}{2} \sum_{i,m} A_{i,m} U_i U_m,$$

where the U_i are the $3N$ Cartesian displacements of the atoms from their equilibrium positions. The μ_i are the atomic masses and the $A_{i,m} \equiv \partial^2 \Phi / \partial U_i \partial U_m$, where Φ is the potential function accounting for all harmonic restoring forces. The deriva-

tives of Φ are to be evaluated at complete equilibrium ($U_i = 0$).

In order to solve the equations of motion associated with H , we assume

$$U_i(t) = u_i e^{-i\omega t} \equiv \mu_i^{-1/2} B_i e^{-i\omega t}, \quad (1)$$

and obtain a set of equations which in matrix notation takes the familiar form

$$[\mu_i^{-1/2} A_{im} \mu_m^{-1/2} - \omega^2 \delta_{im}] \{B_i\} = 0. \quad (2)$$

Solution of this eigenvalue problem involving the dynamical matrix $[\mu_i^{-1/2} A_{im} \mu_m^{-1/2}]$ yields $3N$ eigenfrequencies $\omega_i \geq 0$, each associated with a $3N$ component displacement eigenvector $\{u_i^j\} \equiv \{\mu_i^{-1/2} B_i^j\}$. The eigenvectors of the dynamical matrix $\{B_i^j\}$ are not assumed to be plane waves, so they are orthonormalized according to the relations

$$\sum_i^{3N} B_i^j B_m^i = \delta_{jm}$$

and

$$\sum_i^{3N} B_i^j B_i^k = \delta_{jk}.$$

The solutions of Eq. (2) completely define the harmonic vibrations of the unperturbed sample.

In order to parallel the approach of SG, we assume that there are groups of eigenvectors which are related to each other in some useful way, perhaps because they involve similar motions of atoms. Following SG, a particular group might be identified with a certain kind of vibration of a moleculelike unit whose frequency is spread over a band due to varying interactions with neighboring units of the structure at different sites. A simple example would be the breathinglike motions of tetrahedral units. More generally, a group of eigenvectors may have some more abstract features in common, analogous to those shared by the eigenvectors of a band in a crystalline solid. There may even be a wavevectorlike parameter relating the eigenvectors in a band, but the relationship will not be of the form of plane waves in Cartesian coordinates. In any case, we label each group of eigenvectors as to band index $b = 1, 2, \dots, B$, and let N_b be the number of eigenvectors (and corresponding eigenfrequencies) in each band. The frequencies in band b are listed in order of increasing value and are then labeled $(\omega_b)_j \equiv \omega_{bj}$, where $j = 1, 2, \dots, N_b$. Intuitively, one expects a disordered sample without long range order to exhibit no exact degeneracies. We can therefore expect a one-to-one relationship between eigenfrequencies and eigenvectors, and can label each eigenvector component u_i^{bj} by

its corresponding eigenfrequency: $u_i^{bj} = u_i^j(\omega_{bj})$. In this convenient and important step, each displacement eigenvector component is represented as a function of the vibrational eigenfrequencies in the band. Clearly, eigenfrequencies belonging to several different bands may coexist in the same small frequency range. Keeping this in mind, we define the density of states of vibrations in band b by the equation

$$g_b(\omega) \equiv \lim_{\Delta\omega \rightarrow 0} \frac{1}{\Delta\omega} \sum_{bj} (\omega - \omega_{bj}), \quad (3)$$

where the sum is over the range $(\omega) \leq \omega_{bj} \leq \omega + \Delta\omega$. The number of states per unit sample volume which are in band b and in frequency range $d\omega$ is given by $\rho_b(\omega)d\omega = g_b(\omega)d\omega/v$, where v is the sample volume.

III. INFRARED RESPONSE

Based on this formulation of the vibrational problem⁷ we can proceed to calculate the ir conductivity tensor associated with excitation of vibrations in the sample. We assume the disordered solid is constitutively isotropic, so that the conductivity tensor $\sigma_{\mu\nu}$ is diagonal, $\sigma_{\mu\nu}(\omega) = \sigma(\omega)\delta_{\mu\nu}$. Then, using linear response theory,⁸ and applying $\epsilon_2 = 4\pi\sigma/\omega$, we write

$$\epsilon_2(\omega) = \frac{2\pi}{\hbar v} n^{-1}(\omega, T) \int_{-\infty}^{+\infty} dt e^{-i\omega t} \langle M(t)M(0) \rangle, \quad (4)$$

where $M(t)$ is the Heisenberg representation operator for the sample dipole moment induced in the direction of the electric vector of the light wave. Procedures for calculating $M(t)$ from first principles are outlined in the literature.⁹ Evaluation of $M(t)$ depends on treatment of the electrons in the disordered solid, which we will not attempt in this work. The angular brackets in Eq. (4) indicate an average in the canonical ensemble described by the Hamiltonian of the unperturbed system.¹⁰ Adopting Placzek's procedure we expand the induced dipole moment in powers of the nuclear displacements

$$M(t) = \sum_{i=1}^{3N} M_i U_i(t) + \dots \quad (5)$$

and keep only the terms linear in $U_i(t)$. Substituting Eq. (5) into Eq. (4), and using an expression given by Maradudin¹¹ for the displacement correlation $\langle U_i(t)U_m(0) \rangle$, one obtains

$$\begin{aligned} \epsilon_2(\omega) = 2\pi^2 v n^{-1}(\omega, T) \sum_{i,m} M_i M_m \\ \times \sum_i u_i^i u_m^i \omega_i^{-1} [n_i \delta(\omega - \omega_i) \\ + (n_i + 1) \delta(\omega + \omega_i)], \quad (6) \end{aligned}$$

where $n_i \equiv n(\omega_i, T)$ and u_i^l is the l th-component of the i th displacement eigenvector determined by Eq. (2). The sum over i can be written in a form involving sums over the bands defined earlier

$$\sum_{b=1}^B \sum_{bj} u_i^b(\omega_{bj}) u_m^b(\omega_{bj}) \omega_{bj}^{-1} n_{bj} \delta(\omega - \omega_{bj}), \quad (7)$$

where we have used the fact that terms involving $\delta(\omega + \omega_i)$ vanish because ω and ω_i are positive. For any reasonable function it can be shown that

$$\sum_j f(\omega_j) \delta(\omega \mp \omega_j) = f(\pm\omega) g(\pm\omega);$$

hence, Eq. (7) becomes

$$\sum_{b=1}^B u_i^b(\omega) u_m^b(\omega) \omega^{-1} n(\omega, T) g_b(\omega). \quad (8)$$

Using this in Eq. (6), one obtains the final result that

$$\epsilon_2(\omega) = \frac{2\pi^2}{\omega} \sum_b D_b(\omega) \rho_b(\omega), \quad (9)$$

where

$$D_b(\omega) = \left| \sum_i M_i u_i^b(\omega) \right|^2. \quad (10)$$

The quantity M_i plays the role of an effective charge and is essentially independent of ω , as indicated; the frequency dependence of the $u_i^b(\omega)$ will be discussed later. Although M_i and $u_i^b(\omega)$ are both real, we have used the absolute value sign in Eq. (10) in order to emphasize that D_b is positive-definite. The sum in Eq. (10) does not diverge as $3N$ increases because of the normalization conditions previously imposed on the eigenvectors of Eq. (2). Accordingly, the sum does not have to be carried out over the entire sample, merely over a sufficiently large region to obtain satisfactory convergence.

IV. RAMAN RESPONSE

Having calculated the first-order contributions to the ir dielectric constant, we shall next use similar methods to calculate the first-order Raman spectra. The energy which is scattered from an active volume v , per unit of time and solid angle, can be written in the form¹²

$$I(\omega_0, \Omega) = \frac{(\omega_0 + \Omega)^4}{2\pi c^3} \sum_{k=1,2} \sum_{\alpha\beta,\gamma\lambda} n_\alpha^k n_\beta^k \times i_{\alpha\gamma,\beta\lambda}(\omega_0, \Omega) E_\gamma^- E_\lambda^+, \quad (11)$$

where ω_0 is the frequency of the incident light and $\omega = \omega_0 + \Omega$ is the frequency of the scattered light. Thus, Ω is the Raman shift, and $\Omega < 0$ corresponds to the Stokes spectrum while $\Omega > 0$ gives

the anti-Stokes spectrum. The quantities n_α^k ($k=1, 2$) are Cartesian components of two mutually perpendicular unit vectors, both perpendicular to the scattering direction, and E_γ^- and E_λ^+ are Cartesian components of the incident electric field.¹³ The Raman tensor is given by

$$i_{\alpha\gamma,\beta\lambda}(\omega_0, \Omega) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} dt e^{-i\Omega t} \langle P_{\beta\lambda}(t) P_{\alpha\gamma}^*(0) \rangle, \quad (12)$$

where $P_{\beta\lambda}$ is the operator for the electronic polarizability tensor.¹⁴ As in the case of $M(t)$, procedures for calculating $P_{\beta\lambda}$ from first principles are outlined in the literature.⁹ In order to obtain the Raman scattering, we again use Placzek's procedure and expand $P_{\beta\lambda}$ in $U_i(t)$ keeping only first-order terms¹⁵:

$$P_{\beta\lambda}(t) = \sum_i P_{\beta\lambda}^i U_i(t), \quad (13)$$

where l denotes the $3N$ Cartesian coordinates of the atoms in the excited volume v . Proceeding as in the case of ϵ_2 , the final result is

$$i_{\alpha\gamma,\beta\lambda}(\omega_0, \Omega) = \hbar v \frac{n(\Omega)}{\Omega} \sum_{b=1}^B C_b^{\alpha\gamma\beta\lambda}(\omega_0, \omega) \rho_b(\omega), \quad (14)$$

where

$$C_b^{\alpha\gamma\beta\lambda}(\omega_0, \omega) = \sum_{l,m} P_{\alpha\gamma}^l P_{\beta\lambda}^{*m} u_l^b(\omega) u_m^b(\omega) \quad (15)$$

and Ω has algebraic sign, as defined earlier, while $\omega = |\Omega|$ is the corresponding vibrational frequency. The $P_{\beta\lambda}^i$ are essentially independent of ω , but they may depend appreciably on ω_0 .

When one assumes the disordered solid to be constitutively isotropic, the electronic-transition polarizability tensor $|P_{\beta\lambda}^i|$ is found to contain only two independent quantities: the diagonal elements P_{VV}^i and the off-diagonal elements, denoted P_{VH}^i . It is these two quantities which are probed in the usual 90° scattering experiment: the polarization p of the experiment is VV when the incident (\vec{E}_0) and scattered electric fields are parallel, and is VH when they are perpendicular. Using this notation, one finds that

$$I^p(\omega_0, \omega) = \frac{(\omega_0 + \Omega)^4}{2\pi c^3} \hbar v E_0^2 \frac{n(\Omega)}{\Omega} \sum_{b=1}^B C_b^p(\omega_0, \omega) \rho_b(\omega), \quad (16)$$

where

$$C_b^p(\omega_0, \omega) = \left| \sum_i P_p^i u_i^b(\omega) \right|^2. \quad (17)$$

Here again, we note that $\Omega < 0$ gives the Stokes

spectrum, involving $\omega^{-1}[n(\omega)+1]$, while $\Omega > 0$ gives the anti-Stokes, involving $\omega^{-1}n(\omega)$.

V. DISCUSSION

Equations (9) and (16) allow for a meaningful comparison of the *intensities* of Raman and ir response. Each of these equations contains an expression of the type $\sum_b C_b(\omega)\rho_b(\omega)$ where the coupling coefficients $C_b(\omega)$ are derived from a frequency-independent linear combination of the eigenvector amplitudes $u_b^i(\omega)$. That is, the M_i and the P_b^i are both independent of vibrational frequency ω . Therefore, *Raman and ir spectra of disordered solids can be compared on an equivalent basis by contrasting the reduced Raman spectrum*

$$I_{\text{red}}^p \equiv \frac{I^p(\omega_0, \Omega)}{(\omega_0 + \Omega)^4 [n(\Omega)/\Omega]} \sim \sum_b C_b^p(\omega)\rho_b(\omega), \quad (18)$$

with the infrared derived quantity

$$\omega\epsilon_2(\omega) \sim \sum_b D_b(\omega)\rho_b(\omega). \quad (19)$$

Figure 1 illustrates such a comparison for vitreous SiO_2 . The curves were computed from data reported elsewhere.¹⁸ We will not attempt to analyze these spectra in detail here, as additional information is needed to proceed very far. For example, if one knows the *relative* number of states in each band b , or their actual density $\rho_b(\omega)$, one can extract the relative strength of the coupling coefficients D_b and C_b^p from plots like those in Fig. 1.

From Eqs. (10) and (17) it is clear that the first-order coupling coefficients $D_b(\omega)$ and $C_b^p(\omega_0, \omega)$ derive their vibrational frequency dependence from that of the displacement eigenvector components $u_b^i(\omega)$ of the band involved. It is important to note that the $u_b^i(\omega)$ cannot be assumed to be *independent* of ω since the prior assertion of nondegeneracy would be violated. In fact, the ω dependence will be strong if the localization of eigenmodes changes rapidly with eigenfrequency, as has been reported to be the case by Dean and Bell in computer calculations on linear chains¹⁶ and vitreous silica.¹⁷ Thus, for the full span of cases covered by Eqs. (9) and (16) we cannot specify *a priori* the frequency dependence of $u_b^i(\omega)$. Answers for this dependence will require theoretical analysis of individual structures or classes of structures. We note that the $\rho_b(\omega)$ can be obtained from a purely vibrational calculation, while the motion of the electrons must be considered in order to compute the M_i and the coupling coefficients.

The ir and Raman-coupling coefficients given by Eqs. (10), (15), and (17) involve sums over

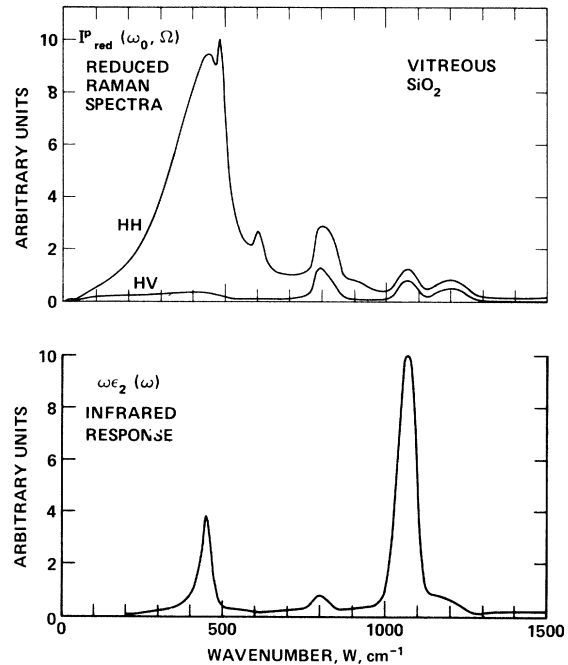


FIG. 1. Comparison of the reduced Raman intensity with the infrared values of $\omega\epsilon_2$ for fused silica. It shows that the band of frequencies centered at 1065 cm^{-1} is strongly infrared active, but weakly Raman active. It appears to show that the strong Raman modes at 450 cm^{-1} are also moderately infrared active; however, it is more likely that the 450-cm^{-1} Raman-active modes are even weaker in the infrared spectrum than appears to be the case, by analogy with the results for vitreous germania shown in Fig. 2.

displacement eigenvectors associated with the equilibrium positions of the atoms in a specific sample. Since these sums should not differ from sample to sample, they may ultimately be calculated as average values over an ensemble of statistically identical samples. Similarly, the densities of states $\rho_b(\omega)$ should not vary among sufficiently large samples and may be replaced by quantities averaged over a suitable ensemble.

The general form of the results presented in Eqs. (9), (10), (16), and (17) holds whenever the system can be described from the point of view of a harmonic-oscillator Hamiltonian. Therefore, the disordered-sample counterpart of local-field effects and of transverse and longitudinal modes¹⁸ are covered by appropriate explicit forms of the dynamical matrix¹⁹ to be used in Eq. (2). Contributions from highly anharmonic systems such as the two-level tunneling model discussed by Anderson, Halperin, and Varma²⁰ are not covered and must be calculated separately.

We remark that while $\omega\epsilon_2$ is the appropriate

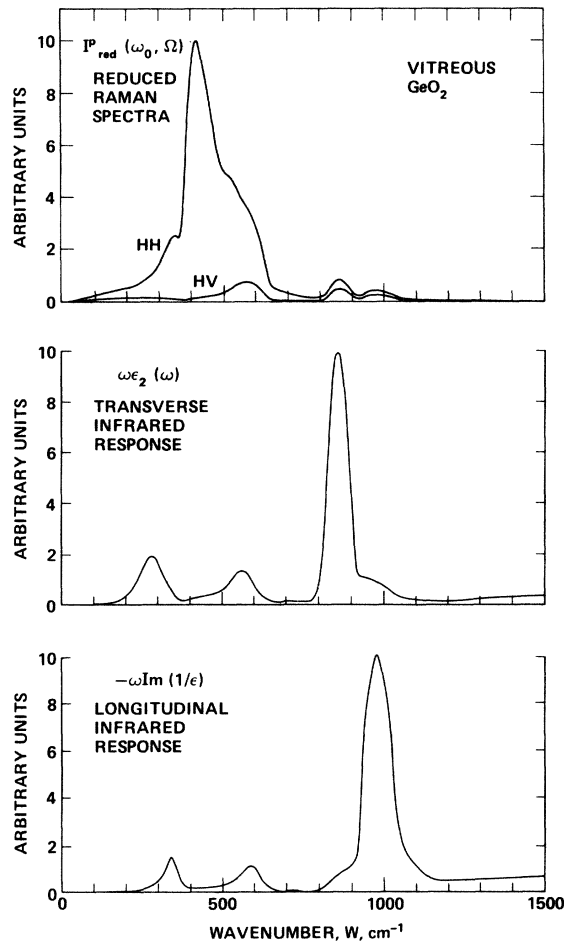


FIG. 2. Comparison of the reduced Raman intensity with infrared derived values of $\omega\epsilon_2$ and $-\omega\text{Im}(1/\epsilon)$ for vitreous germania. It is clear that the infrared active modes centered at 857 cm^{-1} in $\omega\epsilon_2$ are weakly Raman active and have little longitudinal character, while those in the shoulder at about 980 cm^{-1} are also weakly Raman active but are strongly longitudinal. The strongly Raman active modes seen in the reduced Raman spectrum at 420 cm^{-1} appear weakly (or not at all) in the data for $\omega\epsilon_2$, while the moderately infrared active modes seen at 280 cm^{-1} in $\omega\epsilon_2$ are seen to be very weak in Raman activity. It seems that these two different kinds of modes occur at about the same frequency (450 cm^{-1}) in vitreous SiO_2 , according to Fig. 1.

measure of the response of the system to transverse electromagnetic waves, the quantity $-\omega\text{Im}(1/\epsilon)$ is the analogous measure of response to longitudinal waves. Figure 2 illustrates the comparison of Raman, transverse, and longitudinal ir response for vitreous GeO_2 . The curves were computed from data reported by Galeener and Lucovsky,¹⁸ who discuss the nature of longitudinal-optical vibrations in glasses. The strong

peak at 980 cm^{-1} in the lowest part of Fig. 2 indicates the existence of a band of longitudinal-optical phonons having this energy. These are also seen in Raman scattering as shown in the uppermost part of the figure, and are apparently weakly excited in the near-normal incidence infrared reflectivity experiment, as indicated by the shoulder at $\sim 980\text{ cm}^{-1}$ in $\omega\epsilon_2$.

We are not certain how to write the Hamiltonian that includes the Coulomb interactions accounting for LO-TO splittings, so we have not attempted to include such terms formally in the present treatment.²¹ It is clear that in first order the TO and LO vibrations will be harmonic and that the problem can be put in the form of Eq. (2) so that relations of the form of Eqs. (8) and (19) will still hold.

VI. CONCLUDING REMARKS

In summary, we have derived simple expressions for the dielectric constant and Raman scattering intensity due to harmonic vibrations in a macroscopically isotropic disordered sample which has no long-range order, and have shown how both quantities are related to a single set of densities of vibrational subband states. We have pointed out that in general the coefficients measuring coupling to these bands cannot be independent of vibrational frequency; they may vary enormously over a given subband. In some cases, the shape of a peak in the Raman or ir spectrum of a glass may actually reflect the frequency dependence of the coupling coefficients $C_b(\omega)$ much more than that of the subband density of states $\rho_b(\omega)$. Nevertheless, partition of the problem into subbands appears to be advantageous because some coefficients will be only weakly dependent on frequency, and because the behavior of the separate coupling coefficients $C_b(\omega)$ will be more easily understood than that of the coefficient $C(\omega)$ defined by

$$C(\omega)\rho(\omega) \equiv \sum_b C_b(\omega)\rho_b(\omega),$$

where $\rho(\omega) = \sum_b \rho_b(\omega)$ is the total density of vibrational states.

Our method of derivation avoids any quasi-crystalline assumptions and, except for the assertion of complete nondegeneracy within bands, involves no approximations other than those normally encountered in treating coupled vibrational systems in the harmonic approximation. The ir dielectric constant result is entirely new. The Raman scattering results extend those of Shuker and Gammon but *require no restrictions on the correlation length* other than the absence of long-range order. The same approach is being used to

calculate multiphonon effects, and it should be applicable to calculation of other phenomena in disordered solids, both vibrational and electronic in origin.

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*Present address: Xonics, Inc., Santa Monica, Calif., 90401.

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¹⁰Reference 7, p. 288.

¹¹Reference 7, p. 292.

¹²Reference 9, pp. 203 and 368.

¹³Reference 9, p. 199.

¹⁴Reference 8, p. 39.

¹⁵Terms independent of $U_1(t)$ give Rayleigh scattering, and quadratic terms give two-band combination Raman spectra; expressions for $I(\omega_0, \Omega)$ and for $\epsilon_2(\omega)$ have been obtained using these terms and will be published elsewhere.

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