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Inelastic neutron scattering from amorphous solids. I. Calculation of the scattering law for model structures*

J. M. Carpenter
Department of Nuclear Engineering, University of Michigan, Ann Arbor, Michigan 48105
and Argonne National Laboratory, Argonne, Illinois 60439

C. A. Pelizzari
Argonne National Laboratory, Argonne, Illinois 60439
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We present a method for calculating the one- and two-phonon law $S(Q,\omega)$ for models of amorphous solids. The calculation is considerably simplified by explicitly performing the necessary orientation averages. We illustrate use of the method by calculating the one-phonon scattering from long-wavelength acoustic waves in an amorphous solid. In the small-Q region a Brillouin scattering is predicted.

I. INTRODUCTION

Inelastic neutron scattering is a powerful technique for study of the structure and atomic motions of amorphous solids. Quantitative understanding of the measured scattering functions is necessary for any complete interpretation of the results of measurement of course, but in most cases in which neutron scattering cross sections have been computed, the incoherent or other approximations have been used. These are neither necessary nor appropriate in the case of coherently-scattering materials. The purpose of this paper is to provide means for more accurately computing both coherent and incoherent neutron-scattering cross sections, for one-phonon and two-phonon scattering. Results are based on earlier work on the coherent scattering from molecular gases, to which the present results are still applicable.

It is appropriate (until the contrary is established experimentally) to treat amorphous solids as harmonically vibrating systems of atoms with fixed equilibrium positions. Application of the present results requires the frequencies and the displacement vectors for the normal modes of harmonic vibration and the atomic coordinates for a model of an amorphous solid. The isotropic nature of the amorphous system is explicitly accounted for by performing orientation averages of the functions which appear. For calculations based on very large models, or on a large ensemble of smaller models, the explicit averaging is superfluous, and well-known expressions for the scattering from molecules or large crystals can be used. For calculations based on practical, smaller models, the explicit averaging should enhance the accuracy of the result. The one-phonon inelastic scattering for acoustic-wave-like modes of amorphous solids is computed using this analysis.

The basic theory of neutron scattering from harmonically oscillating systems was worked out by Zemach and Glauber.[1] Application of this analysis to crystalline solids is the basis for phonon-dispersion measurements in crystals—introductions to this application are found for example in Lomer and Low[2] or Marshall and Lovesey.[3] More recently, Leadbetter[4] has explored the inelastic scattering in the incoherent approximation, and drawn many useful analogies between the results for coherent scattering from glasses, and from liquids and polycrystals.

Amorphous solids differ from crystals at minimum because of the lack of long-range periodicity among atomic positions. Consequently, there is not the complete destructive and constructive interference which gives rise to Bragg peaks in the elastic scattering, and to momentum-conserving selection rules in the one-phonon inelastic scattering. Furthermore, the normal vibrational modes of amorphous solids cannot be described as propagating waves (except possibly for long-wavelength acoustic-wave modes).

II. SUMMARY OF FUNDAMENTALS

The differential cross section per scattering unit for slow neutron scattering is conveniently discussed in terms of the scattering law (the scattering unit may be one atom in the case of monatomic substances, one chemical formula unit in the case of polyatomic substances)

$$\frac{\partial^2\sigma}{\partial\Omega\partial\epsilon}=\frac{k_f}{\hbar k_0}\exp(-\hbar\omega/2k_BT)(\bar{\sigma}/4\pi)S(Q,\omega)\ . \quad (1)$$

$S(Q,\omega)$ is written here in symmetric form (incorporating the detailed balance factor)

$$S(Q,\omega)=S(Q,-\omega)\ . \quad (2)$$

Here, $\hbar k_0$ and $\hbar k_f$ are neutron initial and final momenta, $\hbar\vec{Q}=\hbar(\vec{k}_f-\vec{k}_0)$ and $\hbar\omega=(\hbar^2/2m)(k_f^2-k_0^2)$ are neutron momentum and energy gain. For isotropic

systems such as amorphous solids, the scattering law is a function of only the magnitude of the momentum transfer $\hbar Q = \hbar|\vec{Q}|$. $\bar{\sigma}$ is an arbitrarily chosen microscopic scattering cross section which enters for normalization purposes; $\bar{\sigma}$ has been taken in the following as the sum of the bound-atom-scattering cross sections for atoms in the scattering unit,

$$\bar{\sigma} = 4\pi \sum_t n_t \langle a_t^2 \rangle , \tag{3}$$

where n_t is the number of atoms of type t in the scattering unity, $\langle a_t^2 \rangle$ is the mean-squared bound-atom scattering length of atoms of type t (averaged over isotopes and scattering spin states).

For harmonically oscillating systems, the scattering law may be developed in a "phonon expansion"

$$S(Q, \omega) = S^{(0)}(Q, \omega) + S^{(1)}(Q, \omega) + S^{(2)}(Q, \omega) + \cdots , \tag{4}$$

in which $S^{(0)}(Q, \omega)$ is the elastic scattering contribution, $S^{(1)}(Q, \omega)$ is the contribution due to excitation or deexcitation of one quantum in a normal mode (phonon), $S^{(2)}(Q, \omega)$ the two-phonon contribution; higher-order terms describe 3, 4 $\cdots$ phonon processes.

When the scatterer is polyatomic, it is further convenient to express each of the terms of the phonon expansion as a weighted sum

$$S^{(i)} = \frac{1}{N\bar{\sigma}} \sum_{\nu\nu'} \sigma_{\nu\nu'} S^{(i)}_{\nu\nu'} , \tag{5}$$

in which the sum is taken twice over all atoms ν of the system and N is the number of scattering units in the system. The $\sigma_{\nu\nu'}$ are the direct or interference cross sections, respectively, for $\nu = \nu'$ or $\nu \neq \nu'$,

$$\sigma_{\nu\nu'} = 4\pi[\langle a_\nu \rangle \langle a_{\nu'} \rangle + (\langle a_\nu^2 \rangle - \langle a_\nu \rangle^2)\delta_{\nu\nu'}] , \tag{6}$$

where $\langle a_\nu \rangle$ and $(\langle a_\nu^2 \rangle - \langle a_\nu \rangle^2)^{1/2}$ are the coherent and incoherent, bound-atom-scattering lengths for the atom at site ν. The average is taken over isotopes and scattering spin states.

III. EXPRESSIONS FOR THE SCATTERING LAW FOR AMORPHOUS SOLIDS

A. Elastic scattering

The elastic scattering term is

$$S^{(0)}(Q, \omega) = S_{el}(Q)\delta(\omega);$$

$$S_{el}(Q) = \frac{1}{N\bar{\sigma}} \sum_{\nu\nu'} \sigma_{\nu\nu'} F^{(0)}_{\nu\nu'}(Q) , \tag{7}$$

where

$$F^{(0)}_{\nu\nu'}(Q) = \langle e^{i\vec{Q}\cdot\vec{b}_{\nu\nu'}} e^{-2W_{\nu\nu'}(\vec{Q})} \rangle_{\text{orientation av}} \approx \langle e^{i\vec{Q}\cdot\vec{b}_{\nu\nu'}} \rangle = j_0(Qb_{\nu\nu'}) . \tag{8}$$

$\vec{b}_\nu$ is the equilibrium position vector of the νth atom,

$$\vec{b}_{\nu\nu'} = \vec{b}_\nu - \vec{b}_{\nu'} ; \quad b_{\nu\nu'} = |\vec{b}_{\nu\nu'}| , \tag{9}$$

and $j_n(x)$ is the nth-order spherical Bessel function. $e^{-2W_{\nu\nu'}(\vec{Q})}$ is the Debye-Waller factor, with

$$2W_{\nu\nu'}(\vec{Q}) = \hbar^2 \sum_\lambda \left(\frac{(\vec{Q}\cdot\gamma^\lambda_\nu)^2}{m_\nu} + \frac{(\vec{Q}\cdot\gamma^\lambda_{\nu'})^2}{m_{\nu'}} \right) \frac{\coth(\hbar\omega^\lambda/2kT)}{4N\hbar\omega^\lambda} . \tag{10}$$

Here γ^λ_ν is the vector displacement of the νth atom in the λth normal mode, with normalization

$$\sum_\nu |\gamma^\lambda_\nu|^2 = N , \tag{11}$$

[with this normalization, the γ's are of order unity and the vibrational displacement is $|\vec{u}^\lambda_\nu| = (\hbar/N m_\nu \omega^\lambda)^{1/2} |\gamma^\lambda_\nu|$], and m_ν is the mass of the νth atom. An asterisk denotes complex conjugation.

It is useful to compare the elastic structure factor with the static structure factor,

$$S(Q) \equiv \int_{-\infty}^{\infty} \cosh\frac{\hbar\omega}{2kT} S(Q, \omega)\, d\omega = \frac{1}{N} \sum_{\nu\nu'} \frac{\sigma_{\nu\nu'}}{\bar{\sigma}} \langle e^{i\vec{Q}\cdot b_{\nu\nu}} e^{-2B_{\nu\nu'}(\vec{Q})} \rangle_{\text{orientation av}} \tag{12}$$

where, in the temperature factor $e^{-2B_{\nu\nu'}(\vec{Q})}$,

$$2B_{\nu\nu'}(\vec{Q}) = \hbar^2 \sum_\lambda \left[\vec{Q}\cdot\left(\frac{\gamma^\lambda_\nu}{\sqrt{m_\nu}} - \frac{\gamma^\lambda_{\nu'}}{\sqrt{m_{\nu'}}} \right) \right]^2 \frac{\coth(\hbar\omega^\lambda/2kT)}{4N\hbar\omega^\lambda} . \tag{13}$$

Both $\bar{e}^{2W}$ and $\bar{e}^{2B}$ are near unity when Q is small, and we ignore them in the following, taking $\bar{e}^{2W} \approx \bar{e}^{2B} \approx 1$.

The radial density function (of equilibrium positions, since we ignore $\bar{e}^{2W}$ and $\bar{e}^{2B}$) may be introduced,

$$4\pi r^2 \bar{\rho}(r) = \frac{1}{N} {\sum_{\nu,\nu'}}' \frac{\sigma_{\nu\nu'}}{\bar{\sigma}} \delta(r - b_{\nu\nu'}) \tag{14}$$

(the primed sum excludes $\nu = \nu'$), which has the property

$$\lim_{r\to\infty} \bar{\rho}(r) \equiv \bar{\rho}_\infty = n(4\pi\langle a \rangle^2/\bar{\sigma}) . \tag{15}$$

Here

$$\langle a \rangle \equiv \sum_t n_t \langle a_t \rangle \tag{16}$$

is the sum of scattering lengths for all atoms in one scattering unit, and n is the number density of scattering units. Then, noting that

$$\lim_{Q\to\infty} S(Q) \equiv S_\infty = \frac{1}{N} \sum_\nu \frac{\sigma_{\nu\nu}}{\bar{\sigma}} = 1 , \tag{17}$$

one has

$$4\pi r[\bar{\rho}(r)-\bar{\rho}_\infty]=\frac{2}{\pi}\int_0^\infty Q\sin Qr[S(Q)-S_\infty]\,dQ. \tag{18}$$

Thus the radial density function can be extracted from the measured scattered intensity; this is the well-known result of Zernike and Prins.[5]

B. One-phonon scattering

The one-phonon term is

$$S_{\nu\nu'}^{(1)}(Q,\omega)=\sum_\lambda \frac{\hbar^2\,\mathrm{csch}(\hbar\omega^\lambda/2k_BT)}{4(\hbar\omega^\lambda)}[\delta(\omega-\omega^\lambda)+\delta(\omega+\omega^\lambda)]F_{\nu\nu'}^{(\pm\lambda)}(Q)\,, \tag{19}$$

where ω^λ is the frequency of the λth normal mode, the summation extends over all normal modes of the system, and $F_{\nu\nu'}^{\lambda}(Q)$ is the partial inelastic structure factor for the λth normal mode

$$F_{\nu\nu'}^{(\lambda)}(Q)=\frac{1}{N}\frac{1}{\sqrt{m_\nu m_{\nu'}}}\langle(\vec{\gamma}_\nu^{\lambda}\cdot\vec{Q})(\vec{\gamma}_{\nu'}^{\lambda *}\cdot\vec{Q})e^{i\vec{Q}\cdot\vec{b}_{\nu\nu'}}e^{-2W_{\nu\nu'}(\vec{Q})}\rangle\,. \tag{20}$$

Again taking the Debye-Waller factor as unity, and using the results of earlier work[6] in which the orientation-averaged one-phonon inelastic structure factors were computed for molecular gases, one may express

$$F_{\nu\nu'}^{(\lambda)}(Q)=\frac{Q^2}{\sqrt{m_\nu m_{\nu'}}}\left[\frac{1}{3}(\vec{\gamma}_\nu^{\lambda}\circ\vec{\gamma}_{\nu'}^{\lambda *})j_0(Qb_{\nu\nu'})+\left(\frac{1}{3}(\vec{\gamma}_\nu^{\lambda}\cdot\vec{\gamma}_{\nu'}^{\lambda *})-\frac{1}{b_{\nu\nu'}^2}(\vec{b}_{\nu\nu'}\circ\vec{\gamma}_\nu^{\lambda})(\vec{b}_{\nu\nu'}\circ\vec{\gamma}_{\nu'}^{\lambda *})\right)j_2(Qb_{\nu\nu'})\right]. \tag{21}$$

The forms used here are actually first-order expansions of modified Bessel functions. Thus, we have expanded $I_n(x)\approx|\frac{1}{2}x|^n/n!$. Only when

$$x=\frac{\hbar(\vec{Q}\cdot\vec{\gamma}_\nu^{\lambda})(\vec{Q}\circ\vec{\gamma}_{\nu'}^{\lambda})}{2N\omega^\lambda}\,\mathrm{csch}\frac{\hbar\omega^\lambda}{2k_BT}\ll 1$$

are these approximations valid (this is usually the case, except for smallest ω, or when N is small, as in small molecules).

C. Two-phonon scattering

The two-phonon terms are of several kinds: those involving excitation or deexcitation of two quanta in a single normal mode, and those involving excitation or deexcitation of one quantum in each of two distinct normal modes. Thus

$$S_{\nu\nu'}^{(2)}(Q,\omega)=\sum_\lambda S_{\nu\nu'}^{(\pm 2\lambda)}(Q,\omega)+\sum_{\lambda<\lambda'} S_{\nu\nu'}^{(\pm\lambda)(\pm\lambda')}(Q,\omega). \tag{22}$$

The sum on modes in the distinct-mode term includes each pair only once—thus (λ,λ') and (λ',λ) are to be counted as only one distinct pair. Here, the single-mode terms are

$$S_{\nu\nu'}^{\pm 2\lambda}(Q,\omega)=\frac{\hbar^4\,\mathrm{csch}^2(\hbar\omega^\lambda/2k_BT)}{32(\hbar\omega^\lambda)^{1/2}}\delta(\omega\pm 2\omega^\lambda)F_{\nu\nu'}^{(2\lambda)}(Q), \tag{23}$$

where $F_{\nu\nu'}^{(2\lambda)}(Q)$ is the inelastic structure factor for exchange of two quanta with the λth normal mode,

$$F_{\nu\nu'}^{(2\lambda)}(Q)=(1/N^2)\langle(1/m_\nu m_{\nu'})(\vec{\gamma}_\nu^{\lambda}\cdot\vec{Q})^2\times(\vec{\gamma}_{\nu'}^{\lambda *}\cdot\vec{Q})^2\,e^{i\vec{Q}\cdot\vec{b}_{\nu\nu'}}e^{-2W_{\nu\nu'}(\vec{Q})}\rangle\,. \tag{24}$$

The distinct-mode terms are

$$S_{\nu\nu'}^{(\pm\lambda)(\pm\lambda')}(Q,\omega)=\frac{\hbar^4\,\mathrm{csch}(\hbar\omega^\lambda/2k_BT)\,\mathrm{csch}(\hbar\omega^{\lambda'}/2k_BT)}{16(\hbar\omega^\lambda)(\hbar\omega^{\lambda'})}\times\delta(\omega\pm\omega^\lambda\pm\omega^{\lambda'})F_{\nu\nu'}^{(\lambda,\lambda')}(Q)\,, \tag{25}$$

where

$$F_{\nu\nu'}^{(\lambda,\lambda')}(Q)=(1/N^2)\langle(1/m_\nu m_{\nu'})(\vec{\gamma}_\nu^{\lambda}\circ\vec{Q})(\vec{\gamma}_{\nu'}^{\lambda *}\circ\vec{Q})\times(\vec{\gamma}_\nu^{\lambda'}\circ\vec{Q})(\vec{\gamma}_{\nu'}^{\lambda' *}\circ\vec{Q})\,e^{i\vec{Q}\cdot\vec{b}_{\nu\nu'}}e^{-2W_{\nu\nu'}(\vec{Q})}\rangle. \tag{26}$$

Both $F_{\nu\nu'}^{(2\lambda)}(Q)$ and $F_{\nu\nu'}^{(\lambda,\lambda')}(Q)$ are of the same form:

$$F_{\nu\nu'}^{(2\lambda)}=F_{\nu\nu'}^{(\lambda,\lambda)}\,.$$

Taking the Debye-Waller factor as unity, the indicated orientation averages which appear are the same as those which were worked out earlier for two-quantum scattering by molecular gases[7]:

$$F_{\nu\nu'}^{(\lambda,\lambda')}(Q)=\frac{1}{N^2}\frac{Q^4}{m_\nu m_{\nu'}}\left[\frac{X}{3.5}j_0(Qb_{\nu\nu'})+\left(\frac{2X}{3.7}-\frac{B}{7}\right)\times j_2(Qb_{\nu\nu'})+\left(\frac{2X}{5.7}-\frac{B}{7}+P\right)j_4(Qb_{\nu\nu'})\right], \tag{27}$$

where

$$X=x_{12}x_{34}+x_{13}x_{24}+x_{14}x_{23}\,,$$

$$B=x_{12}b_3b_4+x_{13}b_2b_4+x_{14}b_2b_3+x_{23}b_1b_4+x_{24}b_1b_3+x_{34}b_1b_2\,,$$

$$P=b_1b_2b_3b_4\,,$$

in which

$$x_{12}=\vec{\gamma}_\nu^{\lambda 1}\cdot\vec{\gamma}_\nu^{\lambda 2}\,,$$
$$x_{13}=\vec{\gamma}_\nu^{\lambda 1}\cdot\vec{\gamma}_{\nu'}^{\lambda 1}\,,$$
$$x_{14}=\vec{\gamma}_\nu^{\lambda 1}\cdot\vec{\gamma}_{\nu'}^{\lambda 2}\,,$$
$$x_{23}=\vec{\gamma}_\nu^{\lambda 2}\cdot\vec{\gamma}_{\nu'}^{\lambda 1}\,,$$
$$x_{24}=\vec{\gamma}_\nu^{\lambda 2}\circ\vec{\gamma}_{\nu'}^{\lambda 2}\,,$$
$$x_{34}=\vec{\gamma}_{\nu'}^{\lambda 1}\cdot\vec{\gamma}_{\nu'}^{\lambda 2}\,,$$
$$b_1=(1/b_{\nu\nu'})(\vec{b}_{\nu\nu'}\cdot\vec{\gamma}_\nu^{\lambda 1})\,,$$
$$b_2=(1/b_{\nu\nu'})(\vec{b}_{\nu\nu'}\cdot\vec{\gamma}_\nu^{\lambda 2})\,,$$
$$b_3=(1/b_{\nu\nu'})(\vec{b}_{\nu\nu'}\cdot\vec{\gamma}_{\nu'}^{\lambda 1})\,,$$
$$b_4=(1/b_{\nu\nu'})(\vec{b}_{\nu\nu'}\cdot\vec{\gamma}_{\nu'}^{\lambda 2})\,.$$

(Note we have suppressed subscripts ν, ν' on X, B, P, x_{ij}, and b_i.)

The two-phonon terms may now be combined, taking advantage of the fact that the structure factors are of the same form for (2λ) and (λ,λ').

$$S^{(2)}_{\nu\nu'}(Q,\omega)=\sum_{\lambda} S^{(2\lambda)}_{\nu\nu'}(Q,\omega)+\sum_{\lambda<\lambda'} S^{(\lambda,\lambda')}_{\nu\nu'}(Q,\omega)$$

$$=\sum_{\lambda}\frac{\hbar^4\,\mathrm{csch}^2(\hbar\omega^\lambda/2k_BT)}{32(\hbar\omega^\lambda)^2}[\delta(\omega+2\omega^\lambda)+\delta(\omega-2\omega^\lambda)]F^{(\lambda,\lambda)}_{\nu\nu'}(Q)+\sum_{\lambda<\lambda'}\frac{\hbar^4\,\mathrm{csch}(\hbar\omega^\lambda/2k_BT)\,\mathrm{csch}(\hbar\omega^{\lambda'}/2k_BT)}{16(\hbar\omega^\lambda)(\hbar\omega^{\lambda'})}$$

$$\times[\delta(\omega+\omega^\lambda+\omega^{\lambda'})+\delta(\omega+\omega^\lambda-\omega^{\lambda'})+\delta(\omega-\omega^\lambda+\omega^{\lambda'})+\delta(\omega-\omega^\lambda-\omega^{\lambda'})]F^{(\lambda,\lambda')}_{\nu\nu'}(Q)\,. \tag{28}$$

D. Recombining results

Combining the results for all atom pairs as in Eq. (4), we obtain for the elastic scattering

$$S^{(0)}(Q,\omega)=\frac{1}{N\bar{\sigma}}\sum_{\nu,\nu'}\sigma_{\nu\nu'}S^{(0)}_{\nu\nu'}(Q,\omega)\,, \tag{29}$$

for one-phonon scattering

$$S^{(1)}(Q,\omega)=\frac{1}{N\bar{\sigma}}\sum_{\nu\nu'}\sigma_{\nu\nu'}S^{(1)}_{\nu\nu'}(Q,\omega)\,, \tag{30}$$

and for two-phonon scattering

$$S^{(2)}(Q,\omega)=\frac{1}{N\bar{\sigma}}\sum_{\nu\nu'}\sigma_{\nu\nu'}S^{(2)}_{\nu\nu'}(Q,\omega)\,. \tag{31}$$

The elastic scattering term is familiar,

$$S^{(0)}(Q,\omega)=\frac{1}{N\bar{\sigma}}\sum_{\nu\nu'}\sigma_{\nu\nu'}j_0(Qb_{\nu\nu'})\,\delta(\omega)\,. \tag{32}$$

The one-phonon term (for $\omega>0$) is

$$S^{(1)}(Q,\omega)=\frac{1}{N\bar{\sigma}}\sum_{\nu\nu'}\sigma_{\nu\nu'}\sum_{\lambda}\frac{\hbar^2\,\mathrm{csch}(\hbar\omega^\lambda/2kT)}{4\hbar\omega^\lambda}\times\delta(\omega-\omega^\lambda)F^{(\lambda)}_{\nu\nu'}(Q)\,. \tag{33}$$

Since the ω-dependent factor can be brought out of the summation after evaluation at $\omega^\lambda=\omega$ (by virtue of the δ-function) one can write

$$S^{(1)}(Q,\omega)=\frac{\hbar^2Q^2\,\mathrm{csch}(\hbar\omega/2kT)}{4\bar{m}\hbar\omega^\lambda}\sum_{\lambda}\delta(\omega-\omega^\lambda)\times\frac{1}{N\bar{\sigma}}\sum_{\nu\nu'}\sigma_{\nu\nu'}\frac{\bar{m}F^{(\lambda)}_{\nu\nu'}(Q)}{Q^2}\,. \tag{34}$$

Here, $\bar{m}$ is the mass of the scattering unit,

$$\bar{m}=\sum_t n_t m_t \tag{35}$$

and the factor Q^2 has been brought out, making the first factor dimensionless.

IV. USE OF RESULTS IN COMPUTING FROM MODELS

Use of the results developed above is straightforward. For finite models, with a total of N_T atoms, some number $N<N_T$ of these, which are remote from boundaries may be considered to constitute the model. The requirement on such a model is that it be large enough so that there is a significant interior volume in which atoms do not experience forces which depend on the presence of the boundaries. The interior must have a diameter at least as great as the ordering distance (distance between atoms beyond which order disappears). All the roughly $3N_T$ normal modes of the structure must be included in the sum on normal modes. In the double atom sum, the first atom index ν ranges over the N interior atoms, while the second atom index ν' ranges over neighbors of the first atom which are within the ordering distance. The second atom sum may range outside the interior N atoms. The model is large enough so that all those second atoms lie remote from boundaries. When the second atom sum is confined to the interior N atoms, corrections for finite model size may be required.

For periodic (thus infinite) models, with N atoms per cell, there are no boundaries (the cell boundaries can be drawn anywhere). All atoms are then "interior" atoms. The requirement on such a model is that it have a cell larger on each side than the ordering distance. The normal mode sum ranges over all $3N$ modes of the cell. In the double atom sum, the first atom index ν ranges over all N atoms in one cell, and the second atom index ν' ranges only among neighbors of the νth atom which lie within the ordering distance, in whatever cell they lie.

V. SCATTERING OF NEUTRONS FROM ACOUSTIC WAVES IN DISORDERED SOLIDS

As an illustrative example, we utilize the results of the preceding sections to calculate the one-quantum scattering from an analytical model for traveling plane-wave modes in an isotropic medium. We consider modes for which the polarization vectors have the simple form

$$\vec{\gamma}^{\,\lambda}_{\nu}=m_\nu^{1/2}\,\vec{\alpha}_j(\vec{q})e^{-i\vec{q}\cdot\vec{b}_\nu}\,, \tag{36}$$

in which $\vec{\alpha}_j(\vec{q})$ determines the polarization of mode $(j,\vec{q})$ with respect to propagation vector $\vec{q}$, and the mode λ is simply renumbered $(j,\vec{q})$. The propagation vectors $\vec{q}$ are in principle a discrete set determined by the external dimensions of the system. The normalization condition (11) applied to these modes implies $\vec{\alpha}_j^{\,2}=\bar{m}^{-1}$, $\bar{m}$ the total mass of a scattering unit. For these modes then, Eq. (10) has

the simple form

$$F^{j\vec{q}}_{\nu\nu'} = (Q^2/M)[\tfrac{1}{3} j_0(Qb_{\nu\nu'}) + (\tfrac{1}{3} - \mu_j^2) j_2(Qb_{\nu\nu'})]\, e^{i\vec{q}\cdot\vec{b}_{\nu\nu'}}, \quad (37)$$

with $M \equiv n\bar{m}$, and μ_j the cosine of the angle between $\vec{q}_j$ and $\vec{b}_{\nu\nu'}$.

The mode sum in Eq. (19) is now replaced by a sum over j and $\vec{q}$; we change the $\vec{q}$ sum to an integral in the usual manner:

$$\sum_{j,\vec{q}} \rightarrow \frac{V}{(2\pi)^3} \sum_j \int d^3q ,$$

where V is the system volume. We limit the j sum to a single longitudinal and two orthogonal transverse polarizations, characterized by distinct sound speeds

$$\omega_L(\vec{q}) = v_L q , \quad (38a)$$

$$\omega_{T_1}(\vec{q}) = \omega_{T_2}(\vec{q}) = v_T q . \quad (38b)$$

It is important to note that the dispersion relations (38) will only be valid for small propagation vectors (long wavelengths) in a microscopically nonuniform medium such as a glass.

Equation (19) can now be written as

$$S^{(1)}_{\nu\nu'}(Q,\omega) = \frac{\hbar Q^2}{2M} \frac{V}{(2\pi)^3} \sum_{j=L,T_1,T_2} \int (2v_j q)^{-1} \times \operatorname{csch} \frac{\hbar v_j q}{2k_B T} \delta(\omega - v_j q)[\tfrac{1}{3} j_0(Qb_{\nu\nu'}) + (\tfrac{1}{3} - \mu_j^2) j_2(Qb_{\nu\nu'})]\, e^{i\vec{q}\cdot\vec{b}_{\nu\nu'}}\, d^3q . \quad (39)$$

The integral in Eq. (39) can be evaluated for our simple modes by noting that

$$\mu_L = \hat{q} \cdot \hat{b}_{\nu\nu'} , \quad (40a)$$

while

$$\mu_{T_1}^2 + \mu_{T_2}^2 = [1 - (\hat{q} \cdot \hat{b}_{\nu\nu'})^2] , \quad (40b)$$

hats denoting unit vectors. On evaluation of the q integral, Eq. (39) can finally be written as

$$S^{(1)}_{\nu\nu'}(Q,\omega) = \frac{\hbar Q^2}{2M} \frac{V}{(2\pi)^2} (2\omega)^{-1} \operatorname{csch} \frac{\hbar\omega}{2k_B T} \{q_L^2/v_L^2 \times [\tfrac{2}{3} j_0(Qb) j_0(q_L b) + \tfrac{4}{3} j_2(Qb) j_2(q_L b)] + q_T^2/v_T [\tfrac{4}{3} j_0(Qb) j_0(q_T b) - \tfrac{4}{3} j_2(Qb) j_2(q_T b)]\}, \quad (41)$$

where $q_j = \omega/v_j$; $j = L, T$. (Note we have dropped the subscripts on $b_{\nu\nu'}$.) From a model of the system structure, from which to obtain the $b_{\nu\nu'}$, the one-phonon scattering law can now be computed.

The one-phonon scattering can also be computed from the structure factor determined by neutron diffraction (or from a model). We can transform the sum on ν, ν' into an integral by introducing the static pair density $\bar{\rho}(r)$ weighted for neutron scattering (this is of course the quantity obtained by Fourier transformation of the neutron-diffraction pattern), which was defined in Eq. (14). Any scattering length weighted sum over all the atomic separations can be conveniently expressed in terms of this function, by

$$\frac{1}{N\bar{\sigma}} \sum_{\nu,\nu'} \sigma_{\nu\nu'} f(b_{\nu\nu'}) = \int_{r=0}^{\infty} f(r)[4\pi r^2 \bar{\rho}(r) + \delta(r)]\, dr . \quad (42)$$

We can therefore express the one-quantum scattering law in terms of $\bar{\rho}(r)$ as

$$S^{(1)}(Q,\omega) = \frac{\hbar Q^2}{2M} \frac{V}{(2\pi)^2} (2\omega)^{-1} \operatorname{csch}\left(\frac{\hbar\omega}{2k_B T}\right)\left(\int_{r=0}^{\infty} 4\pi r^2 \bar{\rho}(r)\{q_L^2/v_L[\tfrac{2}{3} j_0(Qr) j_0(q_L r) + \tfrac{4}{3} j_2(Qr) j_2(q_L r)] + q_T^2/v_T[\tfrac{4}{3} j_0(Qr) j_0(q_T r) - \tfrac{4}{3} j_2(Qr) j_2(q_T r)]\}\, dr + \tfrac{2}{3} q_L^2/v_L + \tfrac{4}{3} q_T^2/v_T\right) . \quad (43)$$

Calculations based on (43) require as input only the pair correlation function $\bar{\rho}(r)$, e.g., from neutron diffraction or a model, and sound speeds v_L and v_T either measured or inferred from other data. For small systems such as the usual random-network physical models, the use of $\bar{\rho}(r)$ probably increases the amount of labor required, by comparison with a straightforward summation of terms such as (41) substituted into (5).

Equation (43) can be cast in a particularly compact and useful form by introducing the neutron structure factor via the Zernike-Prins relation, Eq. (18),

$$S^{(1)}(Q,\omega) = \frac{\hbar Q^2}{2M} \frac{V}{(2\pi)^2} (2\omega)^{-1} \operatorname{csch}\left(\frac{\hbar\omega}{2k_B T}\right)\left[\frac{2}{\pi} \int_{r=0}^{\infty} \int_{K=0}^{\infty} (Kr)^2\, i(K)\, j_0(Kr)\left(\frac{q_L^2}{v_L}[\tfrac{2}{3} j_0(Qr) j_0(q_L r) + \tfrac{4}{3} j_2(Qr) j_2(q_L r)] + \frac{q_T^2}{v_T}[\tfrac{4}{3} j_0(Qr) j_0(q_T r) - \tfrac{4}{3} j_2(Qr) j_2(q_T r)]\right) dK\, dr + \frac{2}{3}\frac{q_L^2}{v_L} + \frac{4}{3}\frac{q_T^2}{v_T} + \int_{r=0}^{\infty} 4\pi r^2 \bar{\rho}_\infty \times \left(\frac{q_L^2}{v_L}[\tfrac{2}{3} j_0(Qr) j_0(q_L r) + \tfrac{4}{3} j_2(Qr) j_2(q_L r)] + \frac{q_T^2}{v_T}[\tfrac{4}{3} j_0(Qr) j_0(q_T r) - \tfrac{4}{3} j_2(Qr) j_2(q_T r)]\right) dr\Bigg] , \quad (44)$$

where $i(Q) \equiv S(Q) - S_\infty$. The integration over r is fairly straightforward but quite tedious, yielding

$$S^{(1)}(Q,\omega) = \frac{\hbar Q^2}{2M}\frac{V}{(2\pi)^2}(2\omega)^{-1}\operatorname{csch}\left(\frac{\hbar\omega}{2k_BT}\right)\left(\int_{\Delta_L}\kappa i(\kappa)\right.$$

$$\times\frac{q_L}{v_L Q}\mu_L^2\,d\kappa + \int_{\Delta_T}\kappa i(\kappa)\frac{q_T}{v_T Q}(1-\mu_T^2$$

$$\left.+\frac{2}{3}\frac{q_L^2}{v_L}+\frac{4}{3}\frac{q_T^2}{v_T}+4\pi^2\frac{\rho_\infty}{v_L}\delta(Q-q_L)\right), \qquad (45)$$

where $\mu_j = (Q^2 + q_j^2 - \kappa^2)/2Qq_j$ and Δ_j is the interval of κ such that $-1 \le \mu_j \le 1$ (i.e., such that Q, q_j, and κ form a triangle). Finally, changing the integral over κ to an integral over μ_L and μ_T,

$$S^{(1)}(Q,\omega) = \frac{\hbar Q^2}{2M}\frac{V}{(2\pi)^2}(2\omega)^{-1}\operatorname{csch}\left(\frac{\hbar\omega}{2k_BT}\right)\left(\frac{q_L^2}{v_L}f_L(Q,\omega)\right.$$

$$\left.+\frac{q_T^2}{v_T}f_T(Q,\omega)+4\pi^2\rho_\infty\delta(\omega - v_L Q)\right) \qquad (46)$$

with

$$f_L(Q,\omega) \equiv \int_{-1}^{1}\mu^2 S((Q^2+q_L^2-2Qq_j\mu)^{1/2})\,d\mu, \qquad (47a)$$

$$f_T(Q,\omega) = \int_{-1}^{1}(1-\mu^2)\,S((Q^2+q_T^2-2Qq_T\mu)^{1/2})\,d\mu. \qquad (47b)$$

The last term in (46) is equivalent to the Brillouin scattering in fluids, while the first two terms correspond to phonon scattering in polycrystals. Calculations based on diffraction data using Eq. (46) do not require the intermediate step of Fourier transformation to $\bar{\rho}(r)$, and relate the inelastic scattering to the measured structure factor. Any possible problems with the ever present termination effects are thereby avoided.

The Brillouin scattering term represents the small-Q limit of the acoustic-wave-mode inelastic scattering. If the Brillouin scattering term in Eq. (46) is used in Eq. (12) to determine the inelastic contribution to the small-Q limit of the total structure factor,

$$\lim_{Q\to 0} S(Q) = 2\int_0^\infty \cosh\tfrac{1}{2}\beta\hbar\omega\,[S(Q,\omega)]_{\text{Brillouin}}\,d\omega, \qquad (48)$$

one obtains the well-known result

$$\lim_{Q\to 0} S(Q) = \rho_\infty k T \chi_s, \qquad (49)$$

where χ_s is the adiabatic compressibility, and

$$v_L^2 = \frac{M}{V}\frac{1}{\chi_s}. \qquad (50)$$

(Of course in a harmonic solid, the adiabatic and isothermal compressibilities are identical.)

VI. CONCLUSION

We have developed a computational method for determining the one- and two-phonon inelastic scattering from amorphous solids. This requires as input information atomic coordinates from a model of the structure, and the eigenvectors and frequencies of the normal modes of the structure. We have also derived means for computing the one-phonon inelastic scattering due to acoustic-wave modes in isotropic media, which require as input the (neutron) weighted radial density function, or the neutron-diffraction pattern and the sound speeds.

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