

Structure Factor for a Mass Disordered One-Dimensional Harmonic Crystal

by

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Abstract

The infrared divergence in the mean square atomic displacements of a one-dimensional pure crystal causes its Debye-Waller factor to vanish. A modified one-phonon expansion avoids this problem and gives a structure factor which is Lorentz peaked near the normal modes of the system. Also, because mass disordering is a harmonic perturbation, the displacement-displacement correlation function of a one-dimensional mass disordered crystal can be calculated to infinite order using many-body techniques. So here, the theoretical calculation of the static and dynamic structure factors of the one dimensional pure crystal is extended, as a first step, to the case of weak mass disordering. The static structure factor is approximately the same as in the pure case. The dynamic structure factor properly includes the effect of the impurity induced modes.

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1 Introduction

There exist real three-dimensional crystals which behave like one-dimensional harmonic crystals. These one-dimensional crystals are crystals in which some of the atoms are aligned along chains and are interconnected by springs [EA78, Tal80]. This is possible in a three-dimensional crystal if such chains can be identified and if at a sufficiently high temperature the interchain interaction is much less than the intrachain interaction. Hence, the properties of this real crystal can be related to the properties of a single chain.

Although the structure factor for three-dimensional crystals can be calculated using a one-phonon expansion, this can not be used for one-dimensional crystals. The mean square atomic displacement diverges at long wavelengths [Pei55, Frö54] because the wavevector space is also one-dimensional. But, for one-dimensional pure crystals, a modified one-phonon expansion [Tal80, WGC83] shows essentially that this infrared divergence produces Lorentz peaks near the reciprocal lattice vectors q_n and the normal modes in the static and dynamic structure factors respectively

$$S(q) = \frac{2\kappa_n/d}{\kappa_n^2 + \bar{q}^2}$$

$$S(q, \omega) = \frac{2\kappa_n^2 c}{\omega^2 d} \left(\frac{1}{\kappa_n^2 + (\bar{q} - \omega/c)^2} + \frac{1}{\kappa_n^2 + (\bar{q} + \omega/c)^2} \right). \quad (1)$$

The wavevector transfer q is expressed as

$$q = q_n + \bar{q}$$

where \bar{q} is the deviation from the reciprocal lattice vector.

Furthermore, the masses in the chain can be disordered by specifying the system configuration which is an assignment of an atom mass to each lattice site. Because the system configuration can not be experimentally controlled, the atom mass at each lattice site is described by a probability distribution which here is a weighted sum of two delta function peaks, peaked at the regular and impurity masses. Also, since an experiment would simultaneously probe many chains, what is needed is the average over all mass configurations of the phonon propagator as was calculated before by Langer [Lan61] and Maradudin [PGMA63].

The purpose of this paper is to study for methodological reasons how the combination of these two features, one-dimensionality and disorder, modifies the structure factors. This theoretical calculation uses finite temperature many-body techniques [Ric80]. The range of the parameters in this calculation is limited to small impurity concentrations, high temperatures but arbitrary impurity atom mass.

Because the disordered displacement-displacement correlation function, used to calculate the structure factors, has an energy gap related to the high impurity mode frequency but the anomalous structure factor properties are related to

the low frequency modes, only a small modification to the structure factor is expected. Also, the mathematical techniques used in this calculation such as the generalization of Wick's theorem and the generalization of the Debye-Waller factor, although they prove the standard results, are easier to use, more powerful and possibly new.

2 Phonon Propagator

The one-dimensional harmonic monatomic pure crystal is defined by the unperturbed Hamiltonian

$$H_o = \frac{1}{2} \sum_l \left(\frac{P_l^2}{M} + \gamma(u_{l+1} - u_l)^2 \right)$$

where M is the atomic mass, γ is the interatomic spring constant and the integer lattice site index l is

$$l \in [-N/2, N/2] \cap \mathbb{Z}$$

where N is the number of lattice sites in the crystal. The displacement and momentum operators, u_l and P_l respectively, are best expressed in terms of the phonon ladder operators

$$a(\eta, k) = \begin{cases} a(k), & \eta = +1, & \text{the annihilation operator} \\ a^+(k), & \eta = -1, & \text{the creation operator} \end{cases}$$

where $\eta = \pm 1$ gives the direction of the operator, by

$$u_l = \left(\frac{\hbar}{2NM} \right)^{1/2} \sum_{\eta, k} \left(\frac{1}{\omega_o(k)} \right)^{1/2} a(\eta, \eta k) e^{ikdl} \quad (2)$$

$$P_l = -i \left(\frac{\hbar M}{2N} \right)^{1/2} \sum_{\eta, k} (\omega_o(k))^{1/2} \eta a(\eta, \eta k) e^{ikdl} \quad (3)$$

where d is the interatomic spacing. By requiring periodic boundary conditions, the wavevectors k of the phonons are

$$k = n \left(\frac{2\pi}{Nd} \right)$$

where the integer n is

$$n \in [-N/2, N/2] \cap \mathbb{Z}.$$

By assuming traveling wave solutions for this system, the unperturbed dispersion relation is

$$\omega_o^2(k) = \omega_M^2 \sin^2 \left(\frac{1}{2} kd \right)$$

where the maximum frequency ω_M is defined by

$$\omega_M^2 = \frac{4\gamma}{M}.$$

In addition, for small k , the dispersion relation can be approximated as

$$\omega_o(k) = ck$$

where the speed c of sound in the crystal is

$$c = \left(\frac{\gamma}{M}\right)^{1/2} d.$$

The one-dimensional harmonic disordered crystal is defined by the perturbed Hamiltonian

$$H = \frac{1}{2} \sum_l \left(\frac{P_l^2}{M_l} + \gamma(u_{l+1} - u_l)^2 \right)$$

where the lattice site dependant mass which specifies the disorder, is

$$M_l = \begin{cases} M, & \text{the regular mass} \\ M', & \text{the impurity mass} \end{cases}.$$

The perturbed Hamiltonian can be expressed in the compact form

$$H = H_o + V$$

where the perturbation V is

$$V = \frac{1}{2} \frac{\lambda}{M} \sum_l \xi_l P_l^2$$

where

$$\lambda = \frac{M}{M'} - 1.$$

The disorder or choice of configuration is contained in unit less variable

$$\xi_l = \begin{cases} 0, & M_l = M \\ 1, & M_l = M' \end{cases}.$$

The key propagator in this problem is the phonon propagator or the displacement-displacement correlation function

$$C_{uu}(l, t) = \langle u_l(t) u_0(0) \rangle_{th, av}$$

where $\langle \rangle_{th}$ is the usual thermal ensemble average with respect to the perturbed Hamiltonian H . After this thermal average has been performed, then the configuration or impurity average $\langle \rangle_{av}$ must be performed. It is defined by the integral

$$\langle A \rangle_{av} = \int dm \rho(m) A(m)$$

where $A(m)$ is an arbitrary function of m , the atomic mass of one particular lattice site. The mass distribution function $\rho(m)$ here is

$$\rho(m) = (1 - n_I)\delta(m - M) + n_I\delta(m - M')$$

where the impurity concentration n_I is the relative number of impurity atoms N_I compared to the total number of atoms N in the system

$$n_I = \frac{N_I}{N}.$$

In addition, if $A(m)$ depends on several lattice sites, an integral must be performed for each lattice site with each integral using its own distribution function. Thus, in reality, the weighted sum over all configurations is just a sum over all configurations with the same impurity concentration n_I because the impurity concentration is an experimentally controllable parameter. The most important and fundamental impurity average calculated is

$$\langle \xi_I \rangle_{av} = n_I.$$

Therefore, it can be shown by using perturbation theory [Ric80] to infinite order and by calculating the self energy of the impurity average to only first order in n_I that [Lan61, PGMA63]

$$C_{uu}(l, \omega) = \frac{2\hbar}{NM} (f_-(\hbar\omega) + 1) \sum_k e^{ikdl} \Im(F(k, \omega + i0^+)) \quad (4)$$

$$C_{uu}(l, t = 0) = \frac{\hbar}{NM} \sum_k e^{ikdl} \frac{1}{\hbar\beta} \sum_{\omega_s} F(k, i\omega_s) \quad (5)$$

where $f_-(\hbar\omega)$ is the Bose distribution function and where the factor $F(k, \omega)$ can be put in the compact form (see Eq. (7.1.45) of Maradudin's article [PGMA63])

$$\begin{aligned} F(k, \omega) &= \frac{1}{\omega_\circ^2(k) - \omega^2} \left(1 - \frac{n_I \omega^2}{J(\omega^2)(\omega_\circ^2(k) - \omega^2) + n_I \omega_\circ^2(k)} \right) \\ &= \frac{J(\omega^2) + n_I}{J(\omega^2)(\omega_\circ^2(k) - \omega^2) + n_I \omega_\circ^2(k)} \\ &= \frac{1}{\omega_\circ^2 - \Omega^2(\omega^2)}. \end{aligned} \quad (6)$$

The renormalized frequency $\Omega^2(\omega^2)$ is

$$\Omega^2(\omega^2) = \frac{\omega^2}{1 + \frac{n_I}{J(\omega^2)}}$$

where the factor $J(z^2)$ is defined for the complex frequency z by

$$J(z^2) = \frac{1}{\epsilon} + \frac{z^2}{\pi} \int_{-\pi/2}^{\pi/2} \frac{dx}{\omega_M^2 \sin^2(x) - z^2}.$$

This form of $F(k, \omega)$ is justified because when $n_I = 0$, the renormalized frequency $\Omega^2(\omega^2)$ reduces to ω^2 and the unperturbed result for the displacement-displacement correlation function is recovered. For specific frequencies, J has the value

$$J(\omega^2 + i0^+) = \begin{cases} \epsilon^{-1} + i\beta(\omega^2), & \text{for } \omega^2 < \omega_M^2 \\ \epsilon^{-1} - \bar{\beta}(\omega^2), & \text{for } \omega^2 > \omega_M^2 \end{cases}$$

where

$$\begin{aligned} \epsilon &= \frac{\lambda}{1 + \lambda} = 1 - \frac{M'}{M} \\ \beta(\omega^2) &= \left(\left(\frac{\omega_M}{\omega} \right)^2 - 1 \right)^{-1/2} \\ \bar{\beta}(\omega^2) &= \left(1 - \left(\frac{\omega_M}{\omega} \right)^2 \right)^{-1/2}. \end{aligned}$$

An important feature of J is that it diverges at the maximum frequency ω_M . It is interesting that $\pi^{-1}\Im(J(\omega^2 + i0^+))$ is also the density of phonon states of the unperturbed lattice.

Also, the key frequencies of this problem are the impurity mode frequencies because they are the new feature introduced by the mass disorder. They are defined as the pole of $F(k, \omega^2)$ at $k = 0$ or equivalently as the zero of $J(\omega^2)$. The impurity mode frequencies can be of two different types depending on whether they fall inside or outside the band. The band is the region of normal mode frequencies of the unperturbed crystal or equivalently the region $0 < \omega^2 < \omega_M^2$.

The impurity mode outside the band is called the localized mode ω_L^2 because it is a normal mode of the perturbed crystal and because this excitation is centered around the impurity atoms. It is defined by

$$J(\omega_L^2) = 0$$

for $\omega_L^2 > \omega_M^2$ and is given by

$$\left(\frac{\omega_L}{\omega_M} \right)^2 = \frac{1}{1 - \epsilon^2}.$$

This solution for the localized mode is only possible when the impurity mass is lighter than the regular mass, i.e. $M' < M$. This impurity mode frequency is for $k = 0$ only. The poles of F for arbitrary k in the first Brillouin zone produce a band of frequencies called the impurity band.

Similarly, if the impurity mass is heavier than the regular mass, i.e. $M' > M$, the existence of an impurity mode inside the band is worth investigating. This mode would be called a resonant mode ω_R^2 because it is not a normal mode of the perturbed crystal but is a damped mode. It is defined by

$$\Re(J(\omega_R^2)) = 0$$

for $\omega_R^2 < \omega_M^2$. But, since this has no solution, a one-dimensional harmonic crystal has no resonant mode.

In order to calculate the perturbed dispersion relation, it is useful to have the derivative of $J(\omega^2)$

$$J'(\omega^2) = \frac{dJ(\omega^2)}{d(\omega^2)}.$$

For $\omega^2 > \omega_M^2$, this is

$$J'(\omega^2) = \frac{1}{2} \frac{1}{\omega_M^2} \left(\frac{\omega_M}{\omega} \right)^4 \left(1 - \left(\frac{\omega_M}{\omega} \right)^2 \right)^{-3/2}$$

and

$$J'(\omega_L^2) = \frac{1}{2} \frac{1}{\omega_M^2} \frac{(1 - \epsilon^2)^2}{\epsilon^3}.$$

By a power series expansion about the impurity mode frequency, $\omega^2 \approx \omega_L^2$, retaining terms up to first order, $J(\omega^2)$ can be approximated as

$$J(\omega^2) \approx (\omega^2 - \omega_L^2) J'(\omega_L^2)$$

since $J(\omega_L^2) = 0$.

The dispersion relation is defined as the pole of $F(k, \omega)$ in Eq.(6), namely

$$\omega_o^2(k) = \Omega^2(\omega^2).$$

When this is solved for the wavevector k as a function of frequency ω , it is a physically realistic single-valued function (see Figure 1.). To be useful in the calculation of the frequency sums, however, the dispersion relation must be solved for the frequency ω as a function of the wavevector k . Because this function is multiple-valued, $F(k, z)$ has two types of poles, a pole near the acoustic mode

$$z^2 = z_o^2(k) \approx \omega_o^2(k)$$

and a pole near the impurity mode

$$z^2 = z_L^2(k) \approx \omega_L^2.$$

For $z_o^2(k) \approx \omega_o^2(k)$, it can be shown that

$$z_o^2(k) \approx \omega_o^2(k) \left(1 + \frac{n_I}{J(\omega_o^2(k))} \right) \quad (7)$$

which is a shift from the unperturbed dispersion relation. A light impurity shifts the frequencies up. A heavy impurity shifts the frequencies down. For $z^2 \approx z_L^2(k)$, it can be shown that

$$F(k, z) \approx \frac{1}{z_o^2(k) - z^2} \left(1 + \frac{n_I}{J(\omega_o^2(k))} \right).$$

Similarly, for $z_L^2(k) \approx \omega_L^2$, it can be shown that

$$z_L^2(k) \approx \omega_L^2 + \frac{n_I}{J'(\omega_L^2)} \left(\left(\frac{\omega_L}{\omega_o(k)} \right)^2 - 1 \right)^{-1} \quad (8)$$

and for $z^2 \approx \omega_L^2$, that

$$F(k, z) \approx \frac{1}{z_L^2(k) - z^2} A(z^2),$$

where $A(z^2)$ is defined as

$$A(z^2) = \frac{1}{\omega_o^2(k)} \left(\left(\frac{\omega_L}{\omega_o(k)} \right)^2 - 1 \right)^{-1} \left(1 + \frac{n_I}{(z^2 - \omega_L^2) J'(\omega_L^2)} \right) (z^2 - \omega_L^2).$$

It is noteworthy that

$$A(z_L^2(k)) = \frac{1}{\omega_L^2} \frac{n_I}{J'(\omega_L^2)} \left(1 - \left(\frac{\omega_o(k)}{\omega_L} \right)^2 \right)^{-2}. \quad (9)$$

The dispersion relations, Eqs. (7) and (8), show that the impurity band is repelled by the acoustic band. (Figure. 1.)

3 Phonon Expansion

The ultimate quantity to be calculated is the dynamic structure factor $S(q, \omega)$ which is defined with respect to the differential neutron scattering cross section $\frac{\partial^2 \sigma}{\partial \Omega_f \partial \epsilon_f}$ by [AM76]

$$\frac{\partial^2 \sigma}{\partial \Omega_f \partial \epsilon_f} = N \frac{k_f}{k_i} \frac{|b|^2}{2\pi \hbar} S(q, \omega)$$

where σ is the cross section, Ω_f represents the final direction of the neutron, ϵ_f is the final energy of the neutron, k_f and k_i are the final and initial wavevectors of the neutron respectively and b is the neutron-nucleus scattering length. Additionally, the static structure factor which is related to the elastic scattering cross section, is defined to be

$$S(q) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} S(q, \omega).$$

By using the Fermi pseudo potential and Fermi's golden rule in this problem, the structure factors can be expressed as

$$S(q, \omega) = \frac{1}{N} \sum_{l, l'} \exp(iqd(l - l')) \int_{-\infty}^{\infty} dt e^{i\omega t} \langle \exp(iqu_l(t)) \exp(-iqu_{l'}(0)) \rangle_{th, av}$$

and

$$S(q) = \frac{1}{N} \sum_{l,l'} \exp(iqd(l-l')) \langle \exp(iqu_l(0)) \exp(-iqu_{l'}(0)) \rangle_{th,av}$$

By a non-trivial generalization of the Debye-Waller theorem for quadratically perturbed many-body systems in Appendix A, the thermal averages are raised into the exponential as

$$S(q, \omega) = \frac{1}{N} \sum_{l,l'} \exp(iqd(l-l')) \cdot \int_{-\infty}^{\infty} dt e^{i\omega t} \left\langle \exp \left(-\frac{1}{2} q^2 \langle u_l^2(t) - 2u_l(t)u_{l'}(0) + u_{l'}^2(0) \rangle_{th} \right) \right\rangle_{av}$$

and

$$S(q) = \frac{1}{N} \sum_{l,l'} \exp(iqd(l-l')) \cdot \left\langle \exp \left(-\frac{1}{2} q^2 \langle u_l^2(0) - 2u_l(0)u_{l'}(0) + u_{l'}^2(0) \rangle_{th} \right) \right\rangle_{av}.$$

These expressions involve a double lattice sum because an unaveraged disordered system does not have space translational symmetry although it does have time translational symmetry.

Because the remaining process of impurity averaging links all of the Feynman diagrams in this expression to scattering centers, the impurity average can not similarly be exactly raised into the exponential. However this can be done approximately by neglecting interference terms which are of higher order in the impurity concentration and which are represented by links inbetween the disconnected diagrams with external vertexes. Therefore, the structure factors can be approximated by

$$S(q, \omega) = \sum_l e^{iqdl} \int_{-\infty}^{\infty} dt e^{i\omega t} \exp(q^2 \langle (u_l(t) - u_0(0))u_0(0) \rangle_{th,av}) \quad (10)$$

and

$$S(q) = \sum_l e^{iqdl} \exp(q^2 A(l))$$

where $A(l)$ is defined by

$$A(l) = \langle u_l(0)u_0(0) \rangle_{th,av} - \langle u_0^2(0) \rangle_{th,av}. \quad (11)$$

The averaged disordered propagators have space translational symmetry.

Because the remaining Fourier transform of time is very difficult, a modified one-phonon expansion [Tal80, WGC83] may be used to expand the dynamic structure factor in powers of the time-dependent correlation function but keeping the static part to all orders. Thus, Eq. (10) is rearranged in the form

$$S(q, \omega) = \sum_l e^{iqdl} \exp(q^2 A(l)) \int_{-\infty}^{\infty} dt e^{i\omega t} \exp(q^2 \langle (u_l(t) - u_l(0))u_0(0) \rangle_{th,av}).$$

For $\omega \neq 0$, the dynamic structure factor can be approximated by its first order term

$$S(q, \omega) = \sum_l e^{iqdl} \exp(q^2 A(l)) q^2 C_{uu}(l, \omega). \quad (12)$$

This approximation is justified because in one-dimensional and two-dimensional pure crystals [Tal80, WGC83] the modified one-phonon expansion agrees with the exact results.

4 Static Structure Factor of the Disordered Crystal

Since the static structure factor depends only on the equal time correlation functions, using Eq. (5) in Eq. (11) gives

$$A(l) = \frac{\hbar}{NM} \sum_k (e^{ikdl} - l) \frac{1}{\hbar\beta} \sum_{\omega_s} F(k, i\omega_s). \quad (13)$$

The sum over imaginary frequencies $i\omega_s$ in this expression can be calculated by using the method of residues which gives (see for example Refs. [PGMA63] and [Ric80])

$$\frac{1}{\hbar\beta} \sum_{\omega_s} F(k, i\omega_s) = - \sum_j f_-(\hbar x_j) \text{Res}(x_j)$$

where $\text{Res}(x_j)$ is the residue of $F(k, \omega)$ at the pole denoted by x_j . In this problem, this can be calculated to be

$$\begin{aligned} & \frac{1}{\hbar\beta} \sum_{\omega_s} F(k, i\omega_s) \\ &= \frac{1 + 2f_-(\hbar z_o(k))}{2z_o(k)} \left(1 + \frac{n_I}{J(\omega_o^2(k))} \right) + \frac{1 + 2f_-(\hbar z_L(k))}{2z_L(k)} A(z_L^2(k)) \end{aligned} \quad (14)$$

where the second term is only present for the light impurity case. Because the acoustic mode frequencies $z_o(k)$ are much less than the ‘‘optical mode’’ frequencies $z_L(k)$, the high temperature approximation

$$f_-(\hbar z_o(k)) \approx \frac{k_B T}{\hbar z_o(k)}$$

can be used for $z_o(k)$ but not for $z_L(k)$. Therefore, the frequency sum in Eq. (14) reduces to

$$\frac{1}{\hbar\beta} \sum_{\omega_s} F(k, i\omega_s) = \frac{k_B T}{\hbar z_o^2(k)} \left(1 + \frac{n_I}{J(\omega_o^2(k))} \right) + \frac{1 + 2f_-(\hbar z_L(k))}{2z_L(k)} A(z_L^2(k)).$$

Using Eq. (7) for $z_o^2(k)$ and Eq. (9) for $A(z_L^2(k))$, the frequency sum reduces to

$$\frac{1}{\hbar\beta} \sum_{\omega_s} F(k, i\omega_s) = \frac{k_B T}{\hbar \omega_o^2(k)} + \frac{1 + 2f_-(\hbar z_L(k))}{2z_L(k)} \frac{1}{\omega_L^2} \frac{n_I}{J'(\omega_L^2)} \left(1 - \left(\frac{\omega_o(k)}{\omega_L} \right)^2 \right)^{-2}.$$

The optical term in this frequency sum can be neglected in comparison to the acoustic term because the Bose factor $f_-(\hbar z_L(k))$ is small, n_I is small and $z_L(k)$ is large. So, the frequency sum becomes the same as in the case of the pure crystal

$$\frac{1}{\hbar\beta} \sum_{\omega_s} F(k, i\omega_s) \approx \frac{k_B T}{\hbar\omega_o^2(k)}. \quad (15)$$

As discussed by Talbot[Tal80], using Eq. (15) in Eq. (13) gives

$$q^2 A(l) = -\kappa d |l|$$

where the width of the static structure factor is

$$\kappa = \frac{q^2 \sigma^2}{2d}.$$

The mean square deviation in the distance between adjacent atom is

$$\sigma^2 = \langle (u_1 - u_0)^2 \rangle_{th,o} = \frac{d^2 k_B T}{M c^2}.$$

Finally, the static structure factor in the disordered crystal

$$S(q) = \frac{\sinh(\kappa d)}{\cosh(\kappa d) - \cos(qd)}$$

is the same as that in the pure crystal because the new impurity induced modes do not have an important effect on it.

5 Dynamic Structure Factor of the Disordered Crystal

Since the first non-zero term (see Eq. (12)) in the modified one-phonon expansion of the dynamic structure factor depends linearly on the phonon propagator, given in Eq. (4), the key factor is the imaginary part of $F(k, \omega + i0^+)$. For $\omega^2 < \omega_M^2$, the term $J(\omega^2)$ is complex and the imaginary part is calculated using Eq. (6) in the same way as Maradudin [PGMA63]. The dynamic structure factor has the form

$$S(q, \omega) = \sum_l e^{iqdl} e^{-\kappa d |l|} \frac{2\hbar q^2}{NM} (f_-(\omega) + 1) \cdot \sum_k e^{ikdl} \frac{\beta(\omega^2) n_I \omega^2}{(\epsilon^{-1}(\omega_o^2(k) - \omega^2) + n_I \omega_o^2(k))^2 + \beta^2(\omega^2)(\omega_o^2(k) - \omega^2)^2}.$$

This shows that the dynamic structure factor has a peaked Lorentzian form. It is peaked at $\omega = z_o(k)$ the roots of the first term in the denominator, which is the perturbed dispersion relation

$$\left(\frac{z_o(k)}{\omega_o(k)} \right)^2 = 1 + n_I \epsilon.$$

The width of the peak is

$$\gamma^2 = \epsilon\beta(\omega^2)\omega_o^2(k)n_I|\epsilon|.$$

This width is proportional to how far the frequency has been perturbed and to the density of states.

But for $\omega^2 > \omega_M^2$, the term $J(\omega^2)$ is real. In this case the imaginary part can be calculated using Eq. (6), which gives a form which is analogous to the pure crystal discussed by Talbot [Tal80]. The reciprocal lattice vectors q_n are defined by

$$q_n d = 2\pi n$$

where n is an integer. When the wavevector transfer q is expressed by

$$q = q_n + \bar{q},$$

the deviation \bar{q} from the scattering peak at q_n is restricted by

$$\left(\bar{q} \pm \frac{\Omega(\omega)}{c}\right) d \ll 1$$

and

$$\kappa_n d \ll 1$$

where the width at the n^{th} peak is

$$\kappa_n = \frac{q_n^2 \sigma^2}{2d}$$

Therefore, the dynamic structure factor has the generalized form

$$S(q, \omega) = \frac{\hbar q_n^2 \kappa_n}{M\Omega(\omega)c} (f_-(\hbar\omega) + 1) \left(\frac{1}{\kappa_n^2 + (\bar{q} - \frac{\Omega(\omega)}{c})^2} + \frac{1}{\kappa_n^2 + (\bar{q} + \frac{\Omega(\omega)}{c})^2} \right).$$

This result is modified from the pure crystal case given by Eq. (1) most near the impurity mode frequencies.

6 Discussion

The work of Langer [Lan61] and Maradudin [PGMA63] was reviewed to calculate the displacement-displacement correlation function and the phonon dispersion relation. The rigorous generalization of Wick's theorem and the Debye-Waller theorem shows that quadratic perturbations allow terms to be calculated to infinite order. The modified one-phonon expansion was applied to the case of a mass disordered one-dimensional crystal. This work shows that the static and dynamic structure factors can be calculated and that the results are not unexpected in view of the fact that the new impurity modes have a finite energy gap at $k = 0$.

Improvements and further work have yet to be done on this problem. Because the operation of mass disordering would also disorder the scattering lengths b , this aspect should also be included in the calculation. Methodologically, a key simplification has been in the dropping of the interference terms in the calculation of the impurity average. Interesting features might appear if they were included.

A Appendix

Wick's theorem expresses the thermal average of a product of ladder operators as a sum of products of thermal averages of pairs of ladder operators. It is

$$\left\langle \prod_{i=1}^{2m} (a_i) \right\rangle_{th,\circ} = \sum_P' \prod_{j=1}^m (\langle a_{P(2j-1)} a_{P(2j)} \rangle_{th,\circ})$$

where the thermal average is taken with respect to an arbitrary harmonic Hamiltonian

$$H_\circ = \sum_k \epsilon(k) a^\dagger(k) a(k).$$

The ladder operators

$$a_i = a(\eta, k) = \begin{cases} a(k), & \eta = +1, & \text{the annihilation operator} \\ a^\dagger(k), & \eta = -1, & \text{the creation operator} \end{cases}$$

have no restriction on their direction η or their wavevector k . The subscript i is a shorthand for the variables (η, k) . A simple consequence of this notation is

$$a^\dagger(\eta, k) = a(-\eta, k)$$

The permutations P are maps from the set of integers $\mathbb{Z}(n)$ onto itself. They are described by

$$P : \mathbb{Z}(n) \rightarrow \mathbb{Z}(n)$$

where the set of integers is

$$\mathbb{Z}(n) = [1, n] \cap \mathbb{Z}.$$

So, $P(i)$ is an integer valued function. The sum is over a restricted set of all permutations P such that the indices of the operators are ordered inside each pair

$$P(2i-1) < P(2i)$$

and all the pairs are ordered

$$P(2i-1) < P(2j-1)$$

for all $i < j$. In a product of $2m$ ladder operators, where $m \in \mathbb{Z}$, the number of permutations is $\frac{(2m)!}{2^m m!}$. Wick's theorem may also be expressed more compactly in terms of the pairing operator Γ which is defined by

$$\Gamma \left(\prod_{i=1}^{2m} (a_i) \right) = \prod_{j=1}^m (\langle a_{2j-1} a_{2j} \rangle_{th,\circ}).$$

For bosons, the thermal average of a pair of ladder operators has the diagonal value

$$\langle a(\eta_1, k_1) a(\eta_2, k_2) \rangle_{th,\circ} = \delta_{\eta_1, \eta_2} \delta_{k_1, k_2} A(\eta_1, k_1)$$

where

$$A(\eta, k) = \begin{cases} f_-(\epsilon(k)) + 1, & \text{for } \eta = +1 \\ f_-(\epsilon(k)), & \text{for } \eta = -1 \end{cases}$$

Therefore, Wick's theorem is

$$\left\langle \prod_{i=1}^{2m} (a_i) \right\rangle_{th,\circ} = \sum_P \Gamma \left(\prod_{i=1}^{2m} (a_{P(i)}) \right).$$

Wick's theorem can be generalized to apply to any Hermitean operator D_i which is an arbitrary linear combination of the ladder operators

$$D_i = \sum_{x_i} d_i(x_i) a(x_i)$$

where the dummy variable is

$$x_i = (\eta_i, k_i).$$

The adjoint of D is

$$D_i^+ = \sum_{x_i} d_i^*(x_i) a^+(x_i)$$

so that

$$D_i = D_i^+.$$

There is no relation between the index of D and the index of x . Examples of D_i are the displacement and momentum operators, Eqs. (2) and (3). The thermal average of a pair of D operators is

$$\langle D_i D_j \rangle_{th,\circ} = \langle D_i D_j^+ \rangle_{th,\circ} = \sum_x d_i(x) d_j^*(x) A(x).$$

Therefore, the generalized Wick's theorem is

$$\left\langle \prod_{i=1}^{2m} (D_i) \right\rangle_{th,\circ} = \sum_P \Gamma \left(\prod_{i=1}^{2m} (D_{P(i)}) \right).$$

As an application of this theorem, it can be shown that

$$\langle D^2 \rangle_{th,\circ} = \sum_x d(x) d^+(x) A(x),$$

$$\langle D^{2m} \rangle_{th,\circ} = \frac{(2m)!}{2^m m!} (\langle D^2 \rangle_{th,\circ})^m,$$

and

$$\langle D^{2m+1} \rangle_{th,\circ} = 0.$$

Furthermore, any function F of the operator D must be expanded as a power series in D

$$F(D) = \sum_{n=0}^{\infty} f_n D^n.$$

For the exponential function, this gives the Debye-Waller theorem

$$\langle e^D \rangle_{th,\circ} = \exp\left(\frac{1}{2} \langle D^2 \rangle_{th,\circ}\right).$$

The binomial theorem for any two variables x and y is

$$(x+y)^N = \sum_{a+b=N} \frac{N!}{a!b!} x^a y^b.$$

Therefore, it can be shown that

$$\begin{aligned} & \langle D_1^{a_1} D_2^{a_2} \rangle_{th,\circ} \\ &= \frac{a_1! a_2!}{2^m} \sum'_{b_1, b_2, c_{1,2}} \frac{1}{b_1! b_2! c_{1,2}!} (\langle D_1^2 \rangle_{th,\circ})^{b_1} (2 \langle D_1 D_2 \rangle_{th,\circ})^{c_{1,2}} (\langle D_2^2 \rangle_{th,\circ})^{b_2} \end{aligned}$$

where a_1 and a_2 are fixed positive integers such that

$$a_1 + a_2 = 2m.$$

The primed summation sign indicates that the sum is done over all those positive integers b_1 , b_2 , and $c_{1,2}$ which conserve the number of D_1 and D_2 operators by

$$2b_1 + c_{1,2} = a_1$$

$$2b_2 + c_{1,2} = a_2$$

and so

$$b_1 + b_2 + c_{1,2} = m.$$

Applying this to the exponentials of the operators gives another form of the Debye-Waller theorem

$$\langle \exp(D_1) \exp(D_2) \rangle_{th,\circ} = \exp\left(\frac{1}{2} \langle D_1^2 + 2D_1 D_2 + D_2^2 \rangle_{th,\circ}\right).$$

Additionally, this result can be generalized to apply to an arbitrary number n of operators D_i . For any n variables x_i , it uses the multinomial theorem

$$\left(\sum_{i=1}^n x_i \right)^N = \sum_{\sum_{i=1}^n a_i = N} \frac{N!}{\prod_{i=1}^n (a_i!)} \prod_{i=1}^n (x_i^{a_i}).$$

Hence

$$\left\langle \prod_{i=1}^n (D_i^{a_i}) \right\rangle_{th, \circ} = \sum_{b_i, c_{i,j}} \frac{\prod_{i=1}^n (a_i!) 2^{\sum_{i<j} c_{i,j}}}{\prod_{i=1}^n (b_i!) \prod_{i<j} (c_{i,j}!) 2^m} \prod_{i=1}^n \langle (D_i^2)_{th, \circ}^{b_i} \rangle \prod_{i<j} \langle (D_i D_j)_{th, \circ}^{c_{i,j}} \rangle$$

where the positive integers a_i are fixed such that

$$\sum_{i=1}^n a_i = 2m.$$

The primed summation indicates that the positive integers b_i and $c_{i,j}$ are restricted for all i such that

$$a_i = 2b_i + \sum_{j<i} c_{j,i} + \sum_{j>i} c_{i,j}$$

and hence

$$\sum_{i=1}^n b_i + \sum_{i<j} c_{i,j} = m.$$

All together, these results give the final generalization of the Debye-Waller factor

$$\left\langle \prod_{i=1}^n (\exp(D_i)) \right\rangle_{th, \circ} = \exp \left(\frac{1}{2} \left\langle \sum_{i=1}^n D_i^2 + 2 \sum_{i<j} D_i D_j \right\rangle_{th, \circ} \right).$$

Now, for a Hamiltonian that has a harmonic perturbation

$$V = \sum_l V_l P_l^2$$

where the P_l are linear in the ladder operators, the interaction evolution operator is

$$\begin{aligned} U_I(\beta) &= T_\tau \exp \left(- \int_0^\beta d\sigma \tilde{V}(\sigma) \right) \\ &= \sum_{n=0}^{\infty} (-1)^n \frac{1}{n!} \int_0^\beta d\vec{\sigma} \sum_{\vec{l}} T_\tau \prod_{j=1}^n (V_{l_j} \tilde{P}_{l_j}^2(\sigma_j)) \end{aligned}$$

where for each n , the vectors are defined as

$$\vec{\sigma} = (\sigma_1, \dots, \sigma_n)$$

$$\vec{l} = (l_1, \dots, l_n).$$

The thermal average of a pair of linear operators under the perturbed Hamiltonian is

$$\begin{aligned} & \langle T_\tau D_i(\tau_i) D_j(\tau_j) \rangle_{th} \\ &= \langle T_\tau \tilde{D}_i(\tau_i) \tilde{D}_j(\tau_j) U_I(\beta) \rangle_{th, \circ, c} \\ &= \Gamma \left(T_\tau \tilde{D}_i(\tau_i) \sum_{n=0}^{\infty} (-1)^n \frac{1}{n!} \int_0^\beta d\vec{\sigma} \sum_{\vec{l}} \sum_R \prod_{k=1}^n (V_{l_{R(k)}} \tilde{P}_{l_{R(k)}}^2(\sigma_{R(k)})) \tilde{D}_j(\tau_j) \right) \\ &= \Gamma \left(T_\tau \tilde{D}_i(\tau_i) \sum_{n=0}^{\infty} (-1)^n \int_0^\beta d\vec{\sigma} \sum_{\vec{l}} \prod_{k=1}^n (V_{l_k} \tilde{P}_{l_k}^2(\sigma_k)) \tilde{D}_j(\tau_j) \right) \end{aligned}$$

The c indicates that only connected Feynmann diagrams are summed over. The R are all permutations of the n objects $k = 1, \dots, n$. Terms that differ only by a relabelling of the indices give the same contribution. Therefore, it can be shown that Wick's theorem applies to operators which are arbitrary linear combinations of the ladder operators, if the thermal average is evaluated for a Hamiltonian which is perturbed by a sum of squares of operators which are linear in the ladder operators. In this case, the generalized Wick's theorem is

$$\left\langle T_\tau \prod_{i=1}^{2m} (D_i(\tau_i)) \right\rangle_{th} = \sum_P \Gamma \left(\prod_{i=1}^{2m} (D_{P(i)}(\tau_{P(i)})) \right).$$

That the Hamiltonian is still quadratic and can be diagonalized is crucial here. Furthermore, all of the corollaries that apply in the unperturbed case also apply in the perturbed case and so

$$\langle F(\vec{A}) \rangle_{th, \circ} = G(\langle \vec{A} \otimes \vec{A} \rangle_{th, \circ}) \Rightarrow \langle F(\vec{A}) \rangle_{th} = G(\langle \vec{A} \otimes \vec{A} \rangle_{th})$$

where \otimes is the tensor product and \vec{A} is a vector of operators. That the average of a function F of operators can be expressed in terms of a function G of averages of pairs of operators, applies equally well to the unperturbed case and to the quadratically perturbed case.

References

- [AM76] Neil W. Ashcroft and N. David Mermin. *Solid State Physics*. Holt, Rinehart and Winston, 1976.
- [EA78] V. J. Emery and J. D. Axe. One-dimensional fluctuations and chain ordering transformations in $\text{Hg}_{3-\delta}\text{AsF}_6$. *Phys. Rev. Let.*, 40:1507, 1978.
- [Frö54] H. Frölich. On the theory of superconductivity: the one-dimensional case. *Proc. Roy. Soc.*, A223:296, 1954.

- [Lan61] J. S. Langer. Frequency spectrum of a disordered one-dimensional lattice. *J. Math. Phys.*, 2:584, 1961.
- [Pei55] R. F. Peierls. *Quantum Theory of Solids*. Clarendon Press, 1955.
- [PGMA63] E. N. Parker, J. S. Goldstein, A. A. Maradudin, and V. Ambegaokar. *Astrophysics and the Many-Body Problem*. Benjamin, 1963.
- [Ric80] G. Rickayzen. *Green's Functions and Condensed Matter*. Academic Press, 1980.
- [Tak63] S. Takeno. Resonance scattering of lattice waves by isotopes. *Prog. Theor. Phys.*, 29:191, 1963.
- [Tal80] E. F. Talbot. On the Emery-Axe theory of the dynamic structure factor of the mercury chain compound, $\text{Hg}_{3-\delta}\text{AsF}_6$. *University of Toronto*, 1980. unpublished M. Sc. report.
- [WGC83] F. Weling, A. Griffin, and M. Carrington. Phonon peaks in the dynamic structure factor of a two-dimensional harmonic crystal. *Phys. Rev.*, B28:5296, 1983.

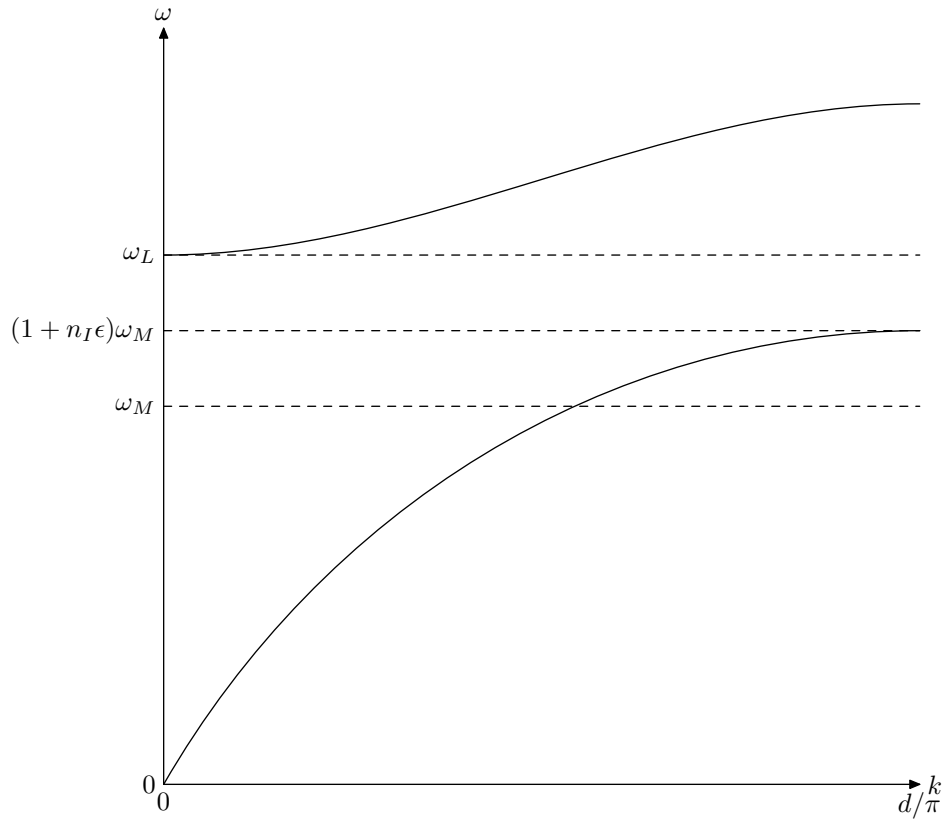


Figure 1: This is a plot of the dispersion relation $\omega^2(k) = \Omega^2(\omega^2)$ showing its two branches, the lower acoustic branch and the upper optical branch. (This graph uses splines to make a representation that is imprecise yet still suggestive.)