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Scattering Response of a Phonon Damped Harmonic Oscillator

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We calculate the response for scattering off a particle bound by an isotropic harmonic force and damped through hybridization with a phonon bath. An exact expression for the scattering response is studied numerically to provide a detailed picture of its dependence on wave vector, frequency, temperature and phonon hybridization strength.

1. Introduction

A damped harmonic oscillator is often used to model the response of a bound particle, or resonant system, to an external probe [1, 2]. In most cases, the damping mechanism is mimiced by a phenomenological force proportional to the particle's velocity, and the strength is termed a friction coefficient. This amounts to a separation of the medium force into components distinguished by their characteristic time scales as in the theory of Brownian motion [3]. The difference between the medium and frictional forces is attributed to a rapidly varying (random) force that averages to zero in time intervals of interest. In consequence, the random force does not enter response functions explicitly. A friction damped oscillator is unphysical in that it does not conserve energy, and a Hamiltonian description does not exist.

In two previous papers [4, 5] we calculated the scattering response function of more realistic models of bound particles. The models have a Hamiltonian description and recognize that the target particles inhabit a non-passive particulate medium. All particle forces are harmonic, so the scattering response is obtained exactly. We demonstrated, among other things, that the observed response incorporates the dynamic properties of the medium in a non-trivial manner, generating highly structured response spectra over extensive ranges of scattering vector and sample temperature. The models are based on mass defects in harmonic lattices; extensive descriptions have been given of a single mass defect (Rubin model) and an ordered binary system.

In this paper we report on the scattering response for a Hamiltonian model that is more akin to the friction damped harmonic oscillator. We consider an oscillator hybridized with a phonon bath, using a single parameter hybridization strength in place of a friction coefficient. The model is of interest both as an exactly solvable, non-trivial, many-particle system [6], and also as a plausible model for the interpretation of proton dynamics in macromolecules, revealed in neutron scattering, for example. We do not dwell on the basic theory of the model or the calculation of the scattering response function since this is similar to our previous work, as is the method of numerical analysis. We should remark that, at least in one dimension, it is possible to find a hybridization strength, which is frequency dependent, that maps the hybridized oscillator model onto the mass defect problem [6]. However, we do not admit a frequency dependence of the strength and, as already remarked, our model is akin to the friction damped oscillator except that we have a microscopic prescription for the damping mechanism.

2. Model Definition

Let Q and P denote the reduced displacement and momentum operators for an oscillator of natural frequency ω_0. The oscillator Hamiltonian is $\omega_0(Q^2$

$+P^2)/2$. The oscillator is hybridized with a phonon bath which contains N modes denumerated by the index ν. Our model is described in the Hamiltonian,

$$\begin{aligned}\mathscr{H} &= \omega_0(Q^2+P^2)/2 \\ &\quad +(1/2)\sum_\nu(\omega_\nu(q_\nu^2+p_\nu^2)+2g_\nu(Qq_\nu+Pp_\nu)) \\ &= (\omega_0-\sum_\nu g_\nu^2/\omega_\nu)(Q^2+P^2)/2 \\ &\quad +(1/2)\sum_\nu \omega_\nu((q_\nu+g_\nu Q/\omega_\nu)^2+(p_\nu+g_\nu P/\omega_\nu)^2). \quad (2.1)\end{aligned}$$

Here, g_ν is the strength of the hybridization, and q_ν and p_ν are the phonon displacement and momentum operators. The strengths are specified in the following section.

From the second form of the Hamiltonian it is evident that the system is stable provided the hybridization strength, for a given ω_0 and phonon frequency spectrum, satisfies the inequality

$$\omega_0 > \sum_\nu (g_\nu^2/\omega_\nu). \quad (2.2)$$

3. Displacement Response $S(k, \omega)$

The response function observed in scattering off a particle at the position defined by R is,

$$\begin{aligned}S(k,\omega) = \int_{-\infty}^{\infty} & (dt/2\pi)\exp(-i\omega t) \\ & *\langle \exp(-i\mathbf{k}\cdot\mathbf{R})\exp\{i\mathbf{k}\cdot\mathbf{R}(t)\}\rangle. \quad (3.1)\end{aligned}$$

Here, angular brackets denote a thermal average of the enclosed quantity, $\mathbf{R}(t)$ is the Heisenberg operator formed with the Hamiltonian (2.1) and $\mathbf{k}$ and ω are the changes in the wave vector and frequency of the scattered radiation. For an isotropic environment the response function is independent of the orientation of $\mathbf{k}$, and this fact is incorporated in the notation used in the Definition (3.1). Note that the displacement operators in the correlation function in (3.1) do not commute at different times. For $t=0$ the correlation function is unity, and thus

$$\int_{-\infty}^{\infty} d\omega S(k,\omega)=1. \quad (3.2)$$

Another useful frequency sum rule is the so-called f-sum rule

$$\int_{-\infty}^{\infty} d\omega\cdot\omega\cdot S(k,\omega)=k^2/2M, \quad (3.3)$$

where M is the mass of the oscillator.

For a harmonic system, such as (2.1), the correlation function in (3.1) reduces to

$$\exp\{k^2\langle xx(t)-x^2\rangle\}, \quad (3.4)$$

with the displacement operator

$$x=(1/M\omega_0)^{1/2}Q. \quad (3.5)$$

The displacement autocorrelation function in (3.4) is readily calculated. We find it convenient to express the result in the form

$$\langle xx(t)\rangle = (-1/2\pi M\omega_0)\int_0^{\infty} d\omega\, G''(\omega)J(\omega,t) \quad (3.6)$$

where

$$J(\omega,t)=\cosh\{\omega(it+1/2T)\}/\sinh(\omega/2T), \quad (3.7)$$

and T is the temperature. The function $G''(\omega)$ in (3.6) is the imaginary part of

$$G(\omega)=\{\omega-\omega_0-\sum_\nu g_\nu^2/(\omega-\omega_\nu)\}^{-1}, \quad (3.8)$$

evaluated with $\omega=\omega+i\varepsilon$ and $\varepsilon\to 0$.

In the limit $g_\nu=0$, $G''(\omega)=-\pi\delta(\omega-\omega_0)$ and (3.6) reduces to the standard expression for the displacement correlation function of a harmonic particle. Inserting the result in (3.1) and exploiting the identity

$$\exp\{y\cosh(x)\} = \sum_{n=-\infty}^{\infty} I_n(y)\exp(nx), \quad (3.9)$$

where I_n is a Bessel function of order n, shows that the response of an undamped oscillator is the sum of an infinite number of delta functions that result from events in which the radiation frequency is changed by a multiple of ω_0 [4, 5]. The amplitude of an event depends on k through the quantity

$$\exp\{-2W(k)\}I_n(y)$$

in which

$$2W(k)=k^2\langle x^2\rangle = y\cosh(\omega_0/2T), \quad (3.10)$$

and $y=k^2/\{2M\omega_0\sinh(\omega_0/2T)\}$.

Damping of the oscillator by the phonon bath arises from the imaginary part of the sum in $G(\omega)$. We adopt the following prescription,

$$\mathrm{Im}\sum_\nu g_\nu^2/(\omega+i\varepsilon-\omega_\nu) = -\pi g^2 Z(\omega) = -\Gamma(\omega), \quad (3.11)$$

where $Z(\omega)$ is a normalized phonon density of states,

$$Z(\omega)=(1/N)\sum_\nu \delta(\omega-\omega_\nu). \quad (3.12)$$

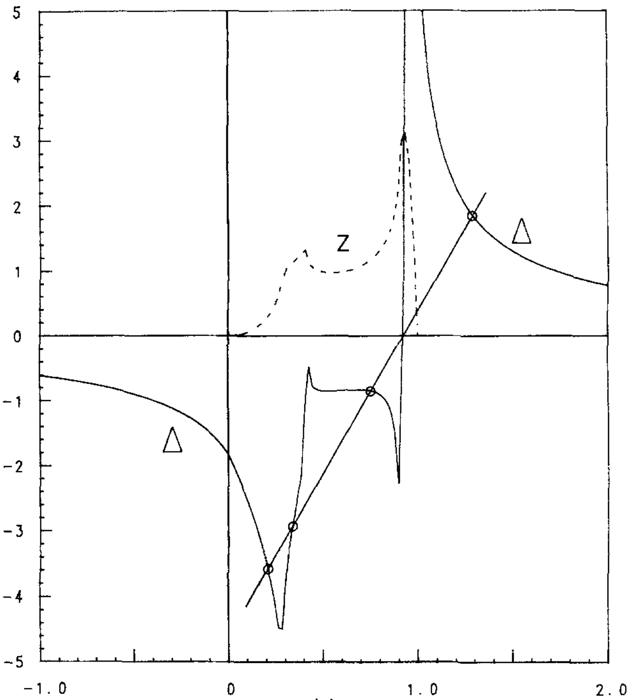

Fig. 1. The model phonon density of states $Z(\omega)$ adopted is displayed together with the corresponding $\Delta(\omega)$, defined in (3.13). Solutions of (4.1) are indicated for $\omega_0=0.921$, $\omega_r=1.3$ and $g^2=0.2$; the maximum $g^2=0.348$. Energies are measured in units of the maximum phonon energy

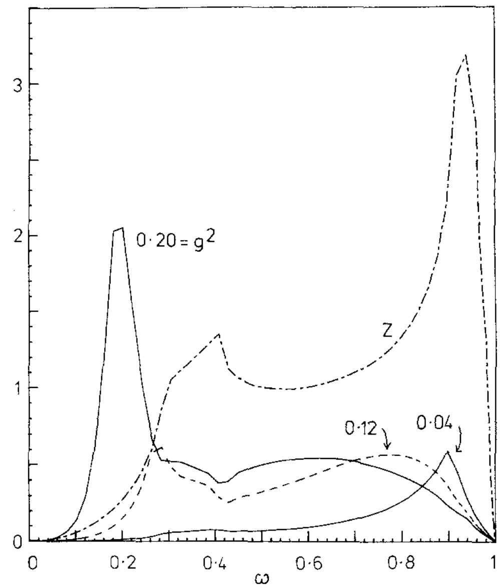

Fig. 2. The imaginary part of $G(\omega)$ in (3.8) which is a spectral weight or, alternatively, an effective density of states for displacements is shown for the parameters used in Fig. 1, and $g^2=0.04$ and 0.12

In (3.11) we have effectively replaced the strengths g_ν by an averaged strength g. The prescription (3.11) completes the definition of our model of a phonon damped oscillator.

The real part of the sum in (3.8) is denoted by $\Delta(\omega)$. Using the standard dispersion relation we arrive at the result

$$\Delta(\omega)=P\int_0^\infty \mathrm{d}u\, Z(u)/(\omega-u), \tag{3.13}$$

where P denotes the principal part of the integral. The stability condition (2.2) now reads

$$\omega_0+g^2\Delta(0)>0. \tag{3.14}$$

For a given phonon spectrum and oscillator frequency (3.14) provides an upper bound on g^2. It is interesting to observe that if g^2 is too large the f sum rule is violated.

4. Basic Features of $S(k,\omega)$

Let us begin by considering properties of the displacement correlation function (3.6) induced by the phonon bath. Solutions of the equation

$$\omega-\omega_0=g^2\Delta(\omega), \tag{4.1}$$

define mode frequencies that essentially determine the dynamic properties of the model. The function $\Delta(\omega)$ for a Debye spectrum $Z(\omega)=3\omega^2/\omega_d^3$, for example, is

$$\Delta(\omega)=(-3/2\omega_d)\{1+2x+x^2\ln([1-1/x]^2)\}, \tag{4.2}$$

where $x=\omega/\omega_d$.

For finite g, there is always a solution of (4.1) above the maximum phonon frequency. This frequency ω_r defines a true resonant mode since it is undamped by the phonon bath. Solutions of (4.1) in the range of ω in which $Z(\omega)$ is finite are physically significant when they occur in a region in which $Z(\omega)$ is very small, since they give pronounced structure in $G''(\omega)$.

This point is illustrated in Figs. 1 and 2 which shows $\Delta(\omega)$ for a realistic density of states. The parameters, in units of the maximum phonon frequency, are $\omega_r=1.3$ and $g^2=0.04$, 0.12 and 0.2, and the maximum $g^2=0.348$. The significant peak in $G''(\omega)$ at $\omega=0.2$ for $g^2=0.2$ arises from the lowest of the three solutions of (4.1). Above this frequency $G''(\omega)$ is, more or less, proportional to $1/Z(\omega)$, except at the band edge.

The case with $g^2=0.2$ is an example where the oscillator frequency $\omega_0=0.912$ lies within the phonon band. When ω_0 lies above the band edge the maximum value of g^2 is enhanced, e.g. with $\omega_0=2.438$, $\omega_r=3.0$ the maximum $g^2=1.325$. For such values of g^2, $G''(\omega)$ is much weaker in the band although it can be structured. The intensity is shifted from the band contribution to the resonance contribution at $\omega=\omega_r$, of course, as we discuss next.

The resonance contribution to $G''(\omega)$ is readily shown to be

$$G''(\omega)=-\pi\delta(\omega-\omega_r)/\{1-g^2\Delta'(\omega_r)\}, \tag{4.3}$$

where $\Delta'(\omega)$ is the frequency derivative of $\Delta(\omega)$. Using the f-sum rule (3.3) it follows that the quantity

$$(\omega/\omega_0)/\{1+(\omega_0-\omega)\Delta'/\Delta\}, \tag{4.4}$$

evaluated at $\omega=\omega_r$ is a measure of the depletion of the band contribution to the displacement correlation function; if the quantity (4.4) is unity the resonance contribution exhausts the sum rule, and there is no band contribution. In this latter, extreme, case the scattering response $S(k,\omega)$ is the same as that of an undamped oscillator of frequency ω_r, i.e. an infinite series of delta functions. For intermediate values of the parameters, the phonon band contributes to the response leading to significant effects in $S(k,\omega)$, as we demonstrate in Sect. 5.

Assembling the results for the phonon band and resonance contribution $G''(\omega)$ leads to the following expression for the displacement correlation function that features in $S(k,\omega)$, namely,

$$\begin{aligned}2M\omega_0\langle xx(t)\rangle &= J(\omega_r,t)/\{1-g^2\Delta'(\omega_r)\}\\ &+\int_0^{\omega_m}(d\omega/\pi)\Gamma(\omega)J(\omega,t)/\{[\omega-\omega_0-g^2\Delta(\omega)]^2\\ &+[\Gamma(\omega)]^2\}.\end{aligned} \tag{4.5}$$

In this expression, $\Gamma(\omega)$ is defined in (3.11) and ω_m is the phonon band cut-off, and

$$\begin{aligned}S(k,\omega)&=\exp\{-2W(k)\}\int_{-\infty}^{\infty}(dt/2\pi)\\ &\cdot\exp\{-i\omega t+k^2\langle xx(t)\rangle\}.\end{aligned} \tag{4.6}$$

In view of the fact that the displacement correlation function (4.5) is a sum of resonance mode and phonon band contributions, we express $S(k,\omega)$ as a convolution of response functions for each contribution. The Fourier transform in the resonance mode response is readily accomplished with the aid of (3.9), and it leads to a response that is the sum of delta functions located at multiples of ω_r. The corresponding amplitudes are obtained from (3.10) on making the replacements $\omega_0\to\omega_r$ and $M\to M\{1-g^2\Delta'(\omega_r)\}$. The final result for the scattering response function is

$$\begin{aligned}S(k,\omega)&=\exp\{-2W_r(k)\}\sum_{n=-\infty}^{\infty}\exp(n\omega_r/2T)I_n(y_r)\\ &* S_l(k,\omega-n\omega_r),\end{aligned} \tag{4.7}$$

where $S_l(k,\omega)$ is the response generated by the phonon band contribution to $\langle xx(t)\rangle$. From (4.7) we deduce immediately that the phonon bath induces structure in the resonance mode lineshapes. This structure is a subject of the next section in which we present numerical results for $S(k,\omega)$, using a frequency scale in which $\omega_m=1$.

5. Numerical Results and Discussion

We have calculated $S(k,\omega)$ as a function of ω for a wide range of k, and two densities of states, namely, the Debye approximation and the realistic model, of three dimensional lattice vibrations, displayed in

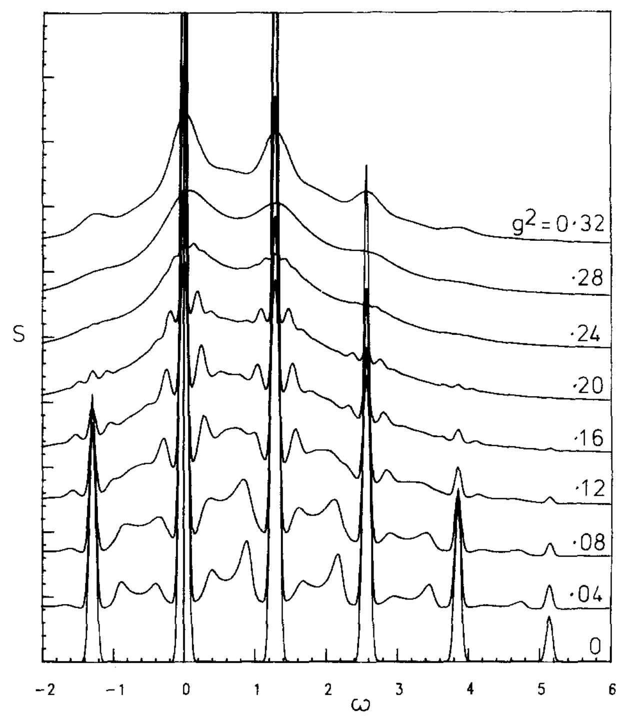

Fig. 3. $S(k,\omega)$ is shown as a function of ω for $k^2/2M=1$, $T=1$ and $\omega_r=1.3$, for a range of values of g^2. The base lines are shifted upwards to clarify the presentation

Figs. 1 and 2. The method of numerical analysis is described in detail in [5].

The variation of $S(k, \omega)$ with g^2 displayed in Fig. 3, for $\omega_r = 1.3$ and fixed wave vector and temperature, can be understood largely by examining the structure in $G''(\omega)$. In the extreme case $g^2 = 0$ we have $\omega_0 = \omega_r$, $G''(\omega)$ is compatible with an Einstein density of states, and $S(k, \omega)$ is the sum of delta functions. With regard to Fig. 3, in order to facilitate visual presentation of $S(k, \omega)$ we have convoluted it with a gaussian function, and this accounts for the finite linewidths for $g^2 = 0$.

Let us now consider the three cases $g^2 = 0.04$, 0.12 and 0.20 since we find that they illustrate the three basic forms of the phonon contribution to $G''(\omega)$ shown in Fig. 2. For both $g^2 = 0.04$ and 0.20 the phonon contribution contains a single, well defined peak, centred at about 0.9 and 0.2, respectively, whereas $g^2 = 0.12$ is an intermediate case with two weak, but well separated, peaks. Well defined peaks in the phonon contribution to $G''(\omega)$ can generate satellite structure in the resonance contributions to $S(k, \omega)$, as is evident from (4.7). Thus, for $g^2 = 0.04$ there are principal satellites at frequencies $= (m\omega_r \pm 0.9)$, where m is an integer. The satellite frequency increment decreases with increasing g^2, and for $g^2 = 0.20$ the principal satellites are located at $(m\omega_r \pm 0.2)$. As g^2 is allowed to approach its maximum value, the satellite frequency increment approaches zero, and the satellite structure eventually coalesces with the resonance contributions yielding broad peaks devoid of the fine structure evident with small g^2.

Results for $S(k, \omega)$ based on a Debye phonon density of states are quite similar to those displayed in Fig. 3, and the evolution of the satellite structure with increasing g^2 is certainly much the same. Small differences exist at intermediate g^2 because the phonon contribution to $G''(\omega)$ for a Debye model possesses just one peak for all values of the coupling strength, and no singular features.

Figures 4 and 5 show, for $\omega_r = 1.3$ and $g^2 = 0.3$, the variation of $S(k, \omega)$ with wave vector and temperature. It is interesting to observe in Fig. 4 that the amplitude of the fundamental contribution to $S(k, \omega)$ is more or less independent of k^2. The significant changes are the decrease in the amplitude of the diffuse background with decreasing k^2, which is required by the f-sum rule, and the increase in the elastic amplitude. The temperature variation of $S(k, \omega)$ illustrated in Fig. 5 is more pronounced, and the first overtone emerges as a distinct feature at the lowest temperature. Figure 6 gives the variation of $S(k, \omega)$ with k^2 for a g^2 that is smaller ($g^2 = 0.20$)

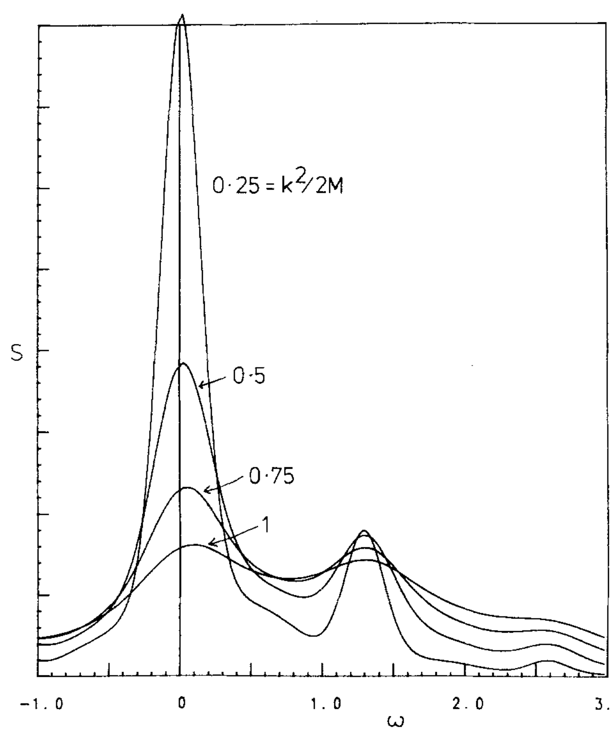

Fig. 4. $S(k, \omega)$ for $\omega_r = 1.3$, $g^2 = 0.3$ and $T = 1$ is shown for various $k^2/2M$

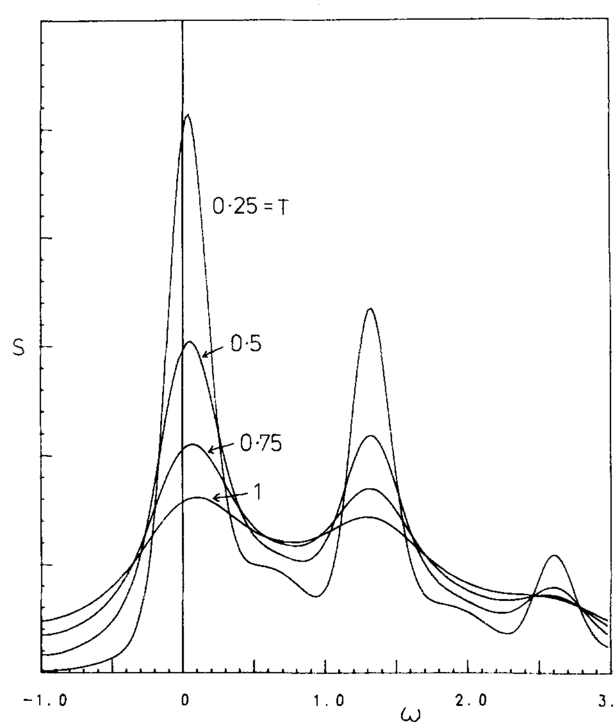

Fig. 5. $S(k, \omega)$ for $\omega_r = 1.3$, $g^2 = 0.3$ and $k^2/2M = 1$ is shown for various temperatures

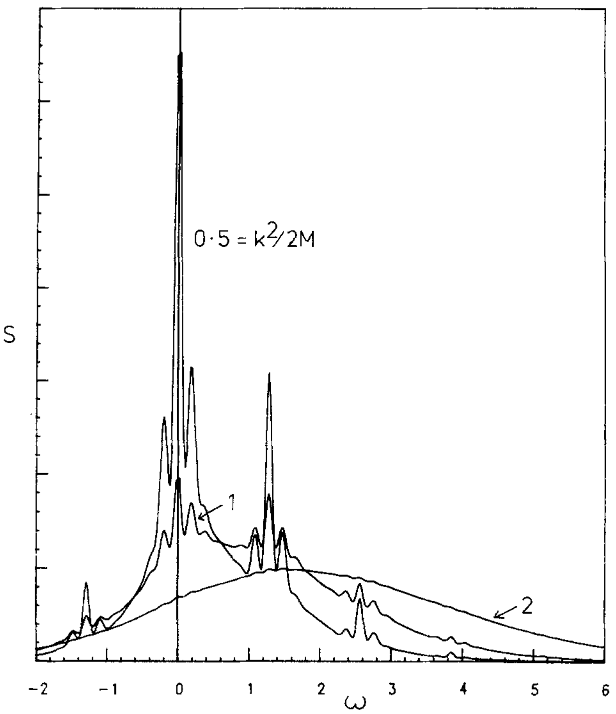

Fig. 6. $S(k, \omega)$ for $\omega_r = 1.3$, $g^2 = 0.2$ and $T = 1$ is shown for various $k^2/2M$

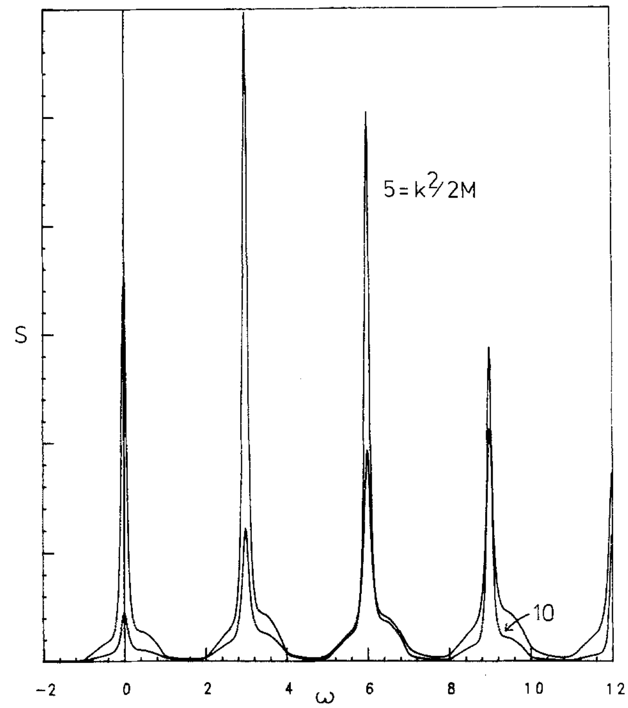

Fig. 7. $S(k, \omega)$ for $\omega_r = 3.0$, $g^2 = 1.3$ and $T = 1$ is shown for two values of the recoil energy $k^2/2M$

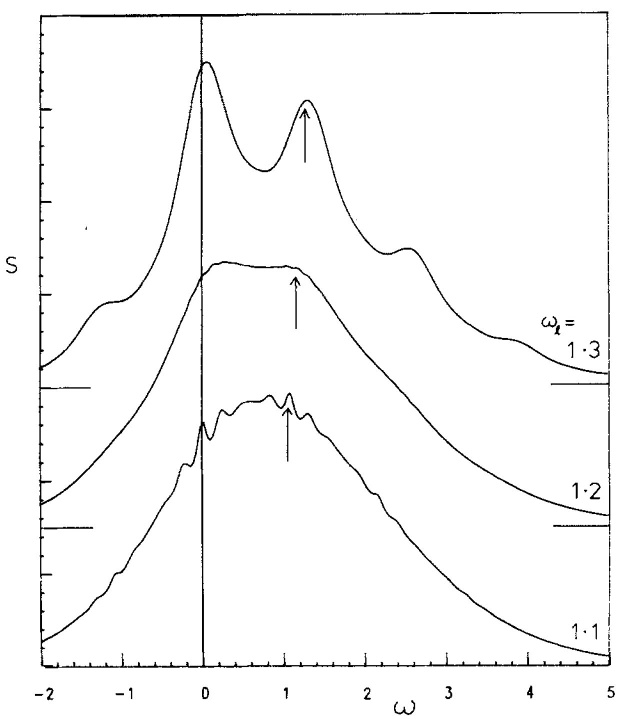

Fig. 8. The g^2 variation of $S(k, \omega)$ for fixed ω_0 is illustrated for $\omega_0 = 0.7$, $k^2/2M = T = 1$. The base lines are shifted for clarity. The values of ω_r are: $g^2 = 0.107$, $\omega_r = 1.1$; $g^2 = 0.200$, $\omega_r = 1.2$; $g^2 = 0.313$, $\omega_r = 1.3$

than the value used in Fig. 4. It serves to illustrate the degradation of fine structure with increasing k^2, for in this case the response with $k^2/2M = 1$ is a gaussian centred at the recoil energy $k^2/2M$, to a good approximation.

Phonon induced structure in $S(k, \omega)$ becomes relatively weaker with increasing ω_r. This feature of the scattered response, and the strong wave vector dependence of mode amplitudes, are illustrated in Fig. 7, which contains $S(k, \omega)$ for $\omega_r = 3.0$, $g^2 = 1.3$ and two values of $k^2/2M$.

Finally, we give an example of the variation in $S(k, \omega)$ with g^2 for fixed ω_0 (and k and T) whereas in previous examples we have kept ω_r fixed. Figure 8 shows $S(k, \omega)$ for $\omega_0 = 0.7$, and $k^2/2M = 1$ and $T = 1$, for three values of g^2. For the smallest value of g^2 the response contains weak structure on a gaussian background centred at the recoil energy. The resonance frequency for this case $\omega_r = 1.1$, while for the largest g^2, $\omega_r = 1.3$ is well separated from the top of the phonon band and the fundamental mode in $S(k, \omega)$ is distinct.

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