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1974 J. Phys. A: Math. Nucl. Gen. 7 410

(http://iopscience.iop.org/0301-0015/7/3/012)

J. Phys. A: Math., Nucl. Gen., Vol. 7, No. 3, 1974. Printed in Great Britain.

The observation of sidebands produced when monochromatic radiation passes through a vibrated resonant medium

J Asher†, T E Cranshaw‡ and D A O'Connor§

† Nuclear Physics Laboratory, University of Oxford, Oxford, UK
‡ Nuclear Physics Division, AERE, Harwell, UK
§ Department of Physics, University of Birmingham, Birmingham, UK

Received 13 July 1973, in final form 27 September 1973

Abstract. Monochromatic (Mössbauer) γ rays are passed through a medium containing resonant nuclei. When the medium is vibrated at ultrasonic frequencies, the emerging radiation analysed by a Mössbauer spectrometer contains sidebands separated from the original frequency by multiples of the ultrasonic frequency. The conditions for the production of these sidebands and their intensities are discussed.

1. Introduction

Several investigations are reported in the literature of the effects of ultrasonic vibration on Mössbauer spectra. These experiments are of two types. Either the source is subjected to the ultrasonic vibration (Ruby and Bolef 1960) and a single line absorber used to measure the Fourier components of the emitted radiation, or the vibration is applied to an absorber and a line source used to observe its absorption spectrum (Cranshaw and Reivari 1967). Usually a piezo-electric crystal is used to supply the ultrasonic motion, but in some experiments the effects of magnetostriction of an absorber in a RF field have been studied (Pfeiffer *et al* 1972). In most cases, a description in terms of the Fourier components of a frequency modulated resonance has been adequate to account for the observations. A quantum-mechanical description leads to the same results.

In the present work, we investigate a different point. Radiation from a Mössbauer single line source is passed through a filter containing resonant nuclei (not necessarily at the same frequency as the source) to which ultrasonic vibration can be applied by a piezo-electric crystal. A second stationary resonant absorber is now used to measure the Fourier components of the radiation which emerges from the filter.

At first sight, it may be somewhat surprising that the transmitted radiation contains frequencies other than those in the source. However, such is the case, and once again a classical description of a resonant absorber in ultrasonic motion adequately accounts for the effects observed.

2. The experiments

A schematic diagram of the experimental configuration is shown in figure 1. In the

experiments, the resonant filter was vibrated at 7·6 MHz, using a piezo-electric crystal (figure 2). The analysing absorber was a thin 1 % ^{57}Fe in Pd foil.

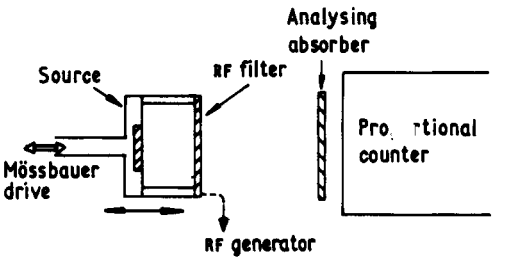

Figure 1. Diagram of the experiment layout.

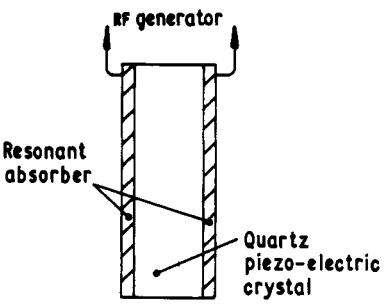

Figure 2. Detail of the ultrasonic drive for the filter.

Observations were made with two experimental arrangements.

(i) The filter consists of 7 % ^{57}Fe in Pd fixed on each side of the crystal with Durofix adhesive. Spectra showing sidebands with a spacing equal to 7·6 MHz are obtained with RF voltage between 0 and 60 V. In this case the filter is resonant with the source radiation, and the centre line of the spectrum is heavily absorbed for low RF voltages, making the sidebands appear prominently (figure 3).

(ii) The filter consists of enriched ^{57}Fe foils. There is here a difference of 7·6 MHz between the source frequency and the frequency of one of the inner lines of the iron hyperfine spectrum due to the combined effect of the magnetic splitting and the isomer shift. At low RF voltages, there is very little absorption of the source radiation. As the RF voltage is increased, sidebands appear in the emerging radiation, and the centre line intensity decreases (figure 4).

The spectra were fitted empirically for sideband intensity and also, in the PdFe case, for absorption of the centre line (characterized by a thickness parameter T so that the centre line depth is given by e^{-T} when the source line depth is unity). These results are summarized in table 1.

An interesting result is that, whereas the PdFe sidebands are symmetrically produced, the ^{57}Fe sidebands appear *asymmetrically* in intensity, although the spacing is uniform, at 7·6 MHz, the modulation frequency. This asymmetry decreases at higher voltage levels, but is still apparent.

No effect was seen when a 'dummy' filter (two layers of Al sandwiching the piezo-electrical crystal) with an RF voltage applied, was used.

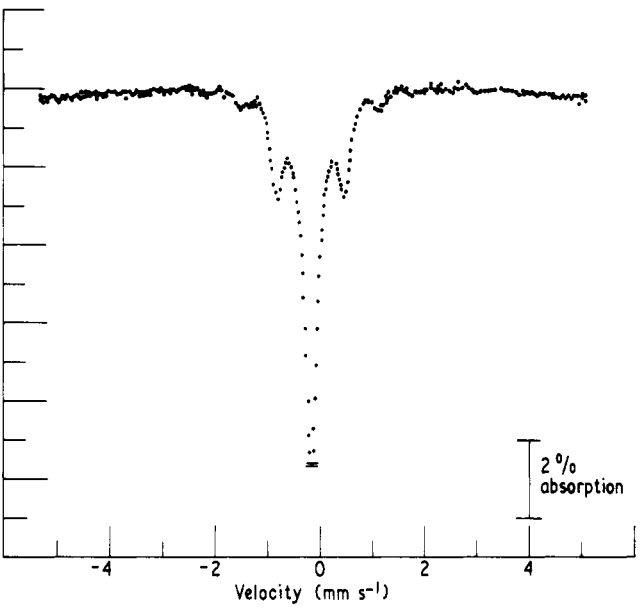

Figure 3. Sideband spectrum observed with PdFe filter.

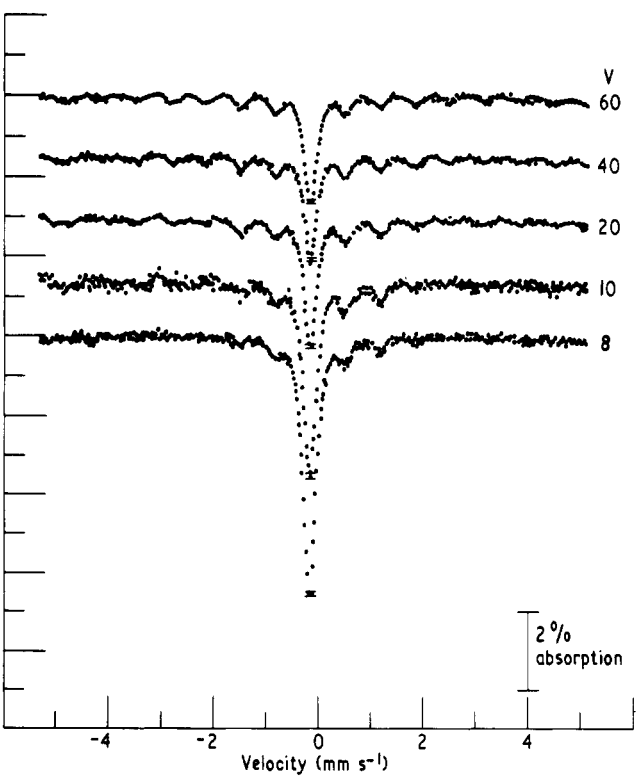

Figure 4. Sideband spectrum observed with ^{57}Fe filter, showing the development of the sidebands with increase in RF voltage.

Table 1.

Filter	RF voltage†	Sideband intensities‡				Error	Thickness parameter
		1st	2nd	3rd	4th		
^{57}Fe	8	L 0·069	0·055	0·032	0·027	±0·01	—
		R 0·096	0·078	0·042	0·024		
	10	L 0·096	0·087	0·029	0·025	±0·015	—
		R 0·139	0·115	0·052	0·023		
	20	L 0·136	0·148	0·065	0·060	±0·006	—
		R 0·181	0·150	0·094	0·043		
	40	L 0·161	0·153	0·098	0·096	±0·007	—
		R 0·178	0·143	0·129	0·079		
	60	L 0·160	0·142	0·110	0·097	±0·008	—
		R 0·163	0·138	0·122	0·089		
PdFe	20	L 0·0199	—	—	—	±0·0015	3·26
		R 0·0246	—	—	—		
	40	L 0·0443	0·0078	0·0041	0·0021	±0·0016	2·17
		R 0·0456	0·0054	0·0045	0·0028		
	50	L 0·0672	0·0124	0·0061	0·0029	±0·001	1·43
		R 0·0688	0·0132	0·0063	0·0034		
	55	L 0·0496	0·0032	0·0009	—	±0·0010	1·55
		R 0·0508	0·0042	0·0010	—		

† Voltage error is ±5%.
‡ Centre line intensity is defined to be 1. Sideband intensity is normalized to the unabsorbed centre line intensity (which is $e^T \times$ (measured centre line intensity)).

In this experiment, the analysing absorber is stationary, while the low frequency Doppler motion is applied to source and filter together. This is equivalent to a hypothetical case in which the source is stationary, the filter undergoes vibration at RF frequency and the Doppler motion is applied only to the analysing absorber. It is this second arrangement which forms the basis for the rest of the discussion.

A qualitative explanation can be given by considering the rest frame of the resonant absorbing nucleus, in which the incident single line gamma ray is frequency modulated by the (apparently) vibrating source, to produce the centre line with equally spaced sidebands (figure 5(*a*)). The resonant nucleus now absorbs only the Fourier component corresponding to its own absorption frequency. The resulting amplitude component spectrum (figure 5(*b*)) is equivalent to a superposition of the original Fourier spectrum (A), and the single absorbed component, 180° out of phase (B) (figure 5(*c*)).

Returning to the lab frame, ie demodulating the spectra, (A) reverts to the original single line frequency, but (B) is now frequency modulated, at the RF vibration frequency (figure 5(*d*)). The result is that the transmitted radiation has superimposed sidebands. Figure 5(*e*) shows the resultant spectrum.

With the PdFe filter, the centre line is resonant and the transmitted sidebands should appear to be symmetric. However, the ^{57}Fe filter is different since the first sideband of the 'incident spectrum' is absorbed and demodulation results in asymmetric sidebands, with the larger sidebands occurring on the side with the resonant frequency component. Since the filter is 'thick' this effect will occur for each of the sidebands produced, which will, in turn, generate further second order sidebands (with the same

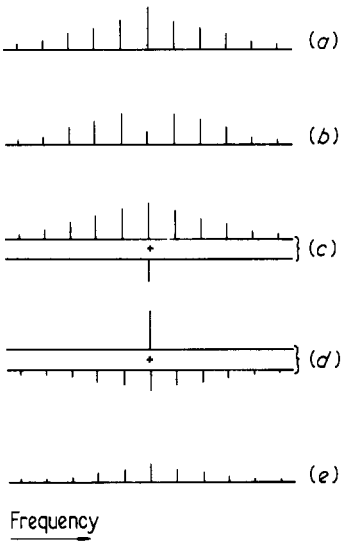

Figure 5. (*a*) Radiation from source as seen by filter; (*b*) radiation after absorption by filter; (*c*) equivalent to (*b*); (*d*) (*c*) in source frame; (*e*) resultant intensity has sidebands.

frequency separation) and tend to dilute the asymmetry. (For ^{57}Fe follow through figure 6).

In a quantum-mechanical treatment, the nuclei in a vibrating absorber can be represented as having their ground and excited state energy levels split into two sets of energy levels, with equal spacing $\hbar\omega_1$, by a sharp peak in the phonon spectrum at angular frequency ω_1. In its ground state, an absorber nucleus occupies one of the ground state levels, according to the number of quanta $\hbar\omega_1$ of energy of vibration.

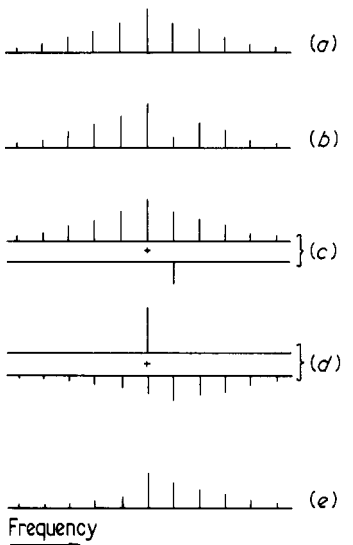

Figure 6. As for figure 5, when the filter is ^{57}Fe.

A transition between a ground state level and the corresponding excited state level involves no change in phonon number and has probability P_0. A transition involving a change in phonon number of $\pm n$ has a probability of $P_{\pm n}$, where $P_{+n} = P_{-n}$. Thus a photon of energy corresponding to the absorber resonance energy will produce forward scattered photons of the same energy with probability P_0^2 and photons with energy shifted by $\pm\hbar\omega_1$, with probability P_0P_1. This results in symmetric sidebands in the scattered photon spectrum.

If the source energy $\hbar\omega_s = \hbar(\omega_a - \omega_1)$, where $\hbar\omega_a$ is the absorber resonance energy, then the forward scattered photons, of energy $\hbar\omega_s$, have associated probability P_1^2, the sideband at $\omega_s + \omega_1$ has a probability P_0P_1, since it involves the annihilation of one phonon, and the sideband at $\omega_s - \omega_1$ has a probability P_1P_2, since it involves the annihilation of one phonon followed by the creation of two phonons. This difference in the probabilities results in asymmetry in the scattered photon sidebands (see figure 7).

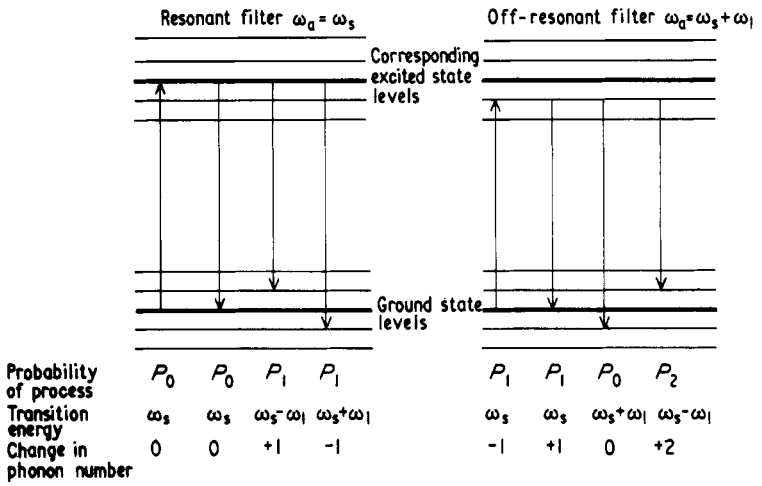

Figure 7. Diagram showing the splitting of the ground and excited state levels by phonons of angular frequency ω_1.

3. Theory

We use the optical theorem (see, for example, Burcham 1963). Assume a wave of amplitude A_0 is incident on a slice dx containing N scatterers per unit volume, of forward scattering amplitude a. Then the transmitted amplitude is

$$A_0(1 - iN\lambda a\,dx)$$

where λ is the wavelength. Let a resonant absorber be at rest, and consider the Fourier component of the source radiation of frequency ω and amplitude $A_\omega(x)$, at depth x in the absorber. The response of the resonant absorber of resonant frequency ω_a and damping constant γ is

$$\gamma a[(\omega_a - \omega) + i\gamma]^{-1}\lambda A_\omega(x)$$

and

$$\frac{dA_\omega(x)}{dx} = -iN\lambda\gamma a[(\omega_a - \omega) + i\gamma]^{-1}A_\omega(x).$$

Thus

$$A_\omega(x) = A_\omega(0)\exp\left(\frac{-iN\lambda\gamma ax}{\omega_a-\omega+i\gamma}\right),$$

and for the intensity, we have

$$I_\omega(x) = I_\omega(0)\exp\left(\frac{-Nx\sigma}{1+S_a^2}\right)$$

where $\sigma = 2\lambda a$, the peak cross section, and $S_a = (\omega_a-\omega)/\gamma$. The source amplitude can be written

$$A_\omega(0) = \frac{\gamma}{\sqrt{\pi}}\frac{1}{(\omega_s-\omega)+i\gamma},$$

where ω_s is the source centre frequency, and if $S = (\omega_s-\omega)/\gamma$

$$\int A_\omega(0)A_\omega^*(0)\,dS = 1.$$

The total transmitted intensity is then

$$\frac{1}{\pi}\int\frac{dS}{1+S^2}\exp\left(-Nx\sigma\frac{1}{1+(S-S_0)^2}\right)$$

where $(\omega_s-\omega_a)/\gamma = S_0$. This is the usual expression for an absorber at rest. We now apply a vibration of frequency ω_1 and peak amplitude u common to all nuclei. The incident wave $A_\omega(x)\,e^{i\omega t}$ now looks to the absorber like

$$A_\omega(x)\exp[i\omega t+iZ\cos(\omega_1 t+\phi)] = A_\omega(x)\,e^{i\omega t}\sum_{p=-\infty}^{\infty}(i)^pJ_p(Z)\exp[ip(\omega_1 t+\phi)]$$

where $Z = (2\pi/\lambda)u$, and ϕ is an arbitrary phase angle. The response of the absorber between x and $x+dx$ is now

$$-iN\lambda a\gamma\,dxA_\omega(x)\exp(i\omega t)\sum_{p=-\infty}^{\infty}(i)^p\frac{J_p(Z)\exp[ip(\omega_1 t+\phi)]}{(\omega_a-p\omega_1-\omega)+i\gamma}.$$

We transform back to the laboratory system by multiplying by $\exp[-iZ\cos(\omega_1 t+\phi)]$. The forward scattered component is then

$$-iN\tfrac{1}{2}\sigma\gamma\,dxA_\omega(x)\exp(i\omega t)\sum_p\sum_q(-1)^q\frac{(i)^{p+q}J_p(Z)J_q(Z)\exp[i(p+q)(\omega_1 t+\phi)]}{\omega_a-p\omega_1-\omega+i\gamma}.$$

Thus the incident wave of frequency ω produces forward scattered waves of frequency $\omega+(p+q)\omega_1$. Denote the amplitude of the wave of frequency $\omega+n\omega_1$ by A_n. Then including all possible processes in which A_p is scattered into A_n, we can write:

$$\frac{dA_n}{dx} = -iN\tfrac{1}{2}\sigma\gamma\sum_p\sum_q\frac{A_p(-1)^q(i)^{n-p}J_{n-p-q}J_q}{\omega_a-(n-q)\omega_1-\omega+i\gamma} \tag{1}$$

with the boundary conditions $A_p = 0$, $x = 0$, $p \neq 0$, $A_0 = A_{00}$ at $x = 0$.

Thus we have an infinity of differential equations to solve. We have to restrict n and make some assumptions about the relative strengths of the A_n.

Assume $A_0 \gg A_n$, which is certainly true for thin absorbers. In the equation for dA_0/dx let us retain only the terms in A_0:

$$\frac{dA_0}{dx} = -iN\tfrac{1}{2}\sigma\gamma A_0 \sum_q \frac{J_q^2}{\omega_a + q\omega_1 - \omega + i\gamma}. \tag{2}$$

Writing out the first terms:

$$\frac{dA_0}{dx} = -iN\tfrac{1}{2}\sigma\gamma A_0\left(\frac{J_0^2}{\omega_a-\omega+i\gamma}+\frac{J_1^2}{\omega_a-\omega_1-\omega+i\gamma}+\frac{J_1^2}{\omega_a+\omega_1-\omega+i\gamma}\right).$$

This is what we expect. The first term is the normal absorption reduced by J_0^2 because the absorber is vibrating. The second term is the absorption by the component of the absorber centred at $(\omega_a-\omega_1)$, of strength J_1^2, etc. Next assume that for any A_n the important terms are its production from A and its self-absorption:

$$\frac{dA_n}{dx} = -iN\tfrac{1}{2}\sigma\gamma A_0 \sum_q \frac{(-1)^q i^n J_{n-q} J_q}{\omega_a-(n-q)\omega_1-\omega+i\gamma} - iN\tfrac{1}{2}\sigma\gamma A_n \sum_q \frac{J_q^2}{\omega_a-(n-q)\omega_1-\omega+i\gamma}. \tag{3}$$

Equation (2) can be solved independently of equation (3) and then equation (3) can be solved. We consider some particular cases.

Case A. Let the source and absorber have the same resonant frequency. Then

$$(\omega_a-\omega) \ll (\omega_a-\omega+p\omega_1)$$

and we can retain terms in $(\omega_a-\omega+i\gamma)^{-1}$ only. Equation (2) then gives

$$A_0(x) = A_{00}\exp\left(-iN\tfrac{1}{2}\sigma\gamma\frac{J_0^2}{(\omega_a-\omega)+i\gamma}x\right).$$

This is the usual absorption reduced by J_0^2. Equation (3) gives for A_1

$$\frac{dA_1}{dx} = -iN\tfrac{1}{2}\sigma\gamma\left(\frac{-iA_0J_1J_0}{\omega_a-\omega+i\gamma}+\frac{J_1^2A_1}{\omega_a-\omega+i\gamma}\right)$$

$$A_1 = iA_{00}\frac{J_1J_0}{J_0^2-J_1^2}\left[\exp\left(-iN\tfrac{1}{2}\sigma\gamma\frac{J_0^2x}{\omega_a-\omega+i\gamma}\right)-\exp\left(-\frac{iN\tfrac{1}{2}\sigma\gamma J_1^2x}{\omega_a-\omega+i\gamma}\right)\right].$$

The intensities are

$$I_0 = |A_0|^2 = |A_{00}|^2\exp\left(-N\sigma J_0^2x\frac{\gamma^2}{(\omega_a-\omega)^2+\gamma^2}\right)$$

$$= |A_{00}|^2\exp\left(-\frac{N\sigma x J_0^2}{1+S_a^2}\right)$$

$$I_1 = |A_{00}|^2\frac{J_1^2J_0^2}{(J_0^2-J_1^2)^2}\left\{\exp\left(-\frac{N\sigma x}{1+S_a^2}J_0^2\right)+\exp\left(-\frac{N\sigma x}{1+S_a^2}J_1^2\right)\right.$$

$$\left.-2\exp\left[\frac{N\sigma x}{1+S_a^2}\left(\frac{J_0^2+J_1^2}{2}\right)\cos\left(\frac{N\sigma x S_a(J_0^2-J_1^2)}{2(1+S_a^2)}\right)\right]\right\}.$$

I_{-1}, the other side band, has the same intensity as I_{+1}. We now have to substitute for

$$|A_{00}|^2 = \frac{1}{\pi}\frac{1}{S^2+1}$$

and integrate with respect to S. For this case $S = S_a$.

Case B. Source and absorber shifted, $S \neq S_a$. Assume that we choose ω_1 so that $\omega_a - \omega_1 = \omega_s$. Then for incident frequencies $\omega \sim \omega_s$, we have that

$$\omega_a - \omega_1 - \omega \ll \omega_a - p\omega_1 - \omega,$$

$p \neq 1$ and terms in $(\omega_a - \omega_1 - \omega + i\gamma)^{-1}$ are more important than all others.

The equation for A_0 is

$$\frac{dA_0}{dx} \simeq -iN\tfrac{1}{2}\sigma\gamma A_0 \frac{J_1^2}{\omega_a - \omega_1 - \omega + i\gamma}$$

For A_1,

$$\frac{dA_1}{dx} = \tfrac{1}{2}N\sigma\gamma A_0 \frac{J_0 J_1}{\omega_a - \omega_1 - \omega + i\gamma} - i\tfrac{1}{2}N\sigma\gamma A_1 \frac{J_1^2}{\omega_a - \omega_1 - \omega + i\gamma}.$$

For A_{-1},

$$\frac{dA_{-1}}{dx} = \tfrac{1}{2}N\sigma\gamma A_0 \frac{J_1 J_2}{\omega_a - \omega_1 - \omega + i\gamma} - i\tfrac{1}{2}N\sigma\gamma A_{-1} \frac{J_2^2}{\omega_a - \omega_1 - \omega + i\gamma}.$$

Note the asymmetry. The production term for A_1 depends on $J_0 J_1$, which is normally larger than $J_1 J_2$ for A_{-1}. Then

$$A_0(x) = A_{00} \exp\left(-i\tfrac{1}{2}N\sigma\gamma x \frac{J_1^2}{\omega_a - \omega_1 - \omega + i\gamma} \right)$$

$$A_1(x) = A_{00} \frac{N\sigma\gamma x}{2(\omega_a - \omega - \omega_1 + i\gamma)} J_0 J_1 \exp\left(-i\tfrac{1}{2}N\sigma \frac{\gamma x J_1^2}{\omega_a - \omega_1 - \omega + i\gamma} \right)$$

$$A_{-1}(x) = A_{00} \frac{N\sigma\gamma x}{2(\omega_a - \omega - \omega_1 + i\gamma)} \frac{J_1 J_2}{J_1^2 - J_2^2} \left[\exp\left(-i\tfrac{1}{2}N\sigma \frac{\gamma x J_1^2}{\omega_a - \omega_1 - \omega + i\gamma} \right) - \exp\left(-i\tfrac{1}{2}N\sigma \frac{\gamma x J_2^2}{\omega_a - \omega_1 + \omega + i\gamma} \right) \right].$$

$|A_1|^2$ is obviously greater than $|A_{-1}|^2$ for large x. For small x

$$A_{-1}(x) = AJ_1 J_2 \frac{N\sigma\gamma x}{2(\omega_a - \omega_1 - \omega + i\gamma)}$$

which is again smaller in the ratio $J_1 J_2 / J_0 J_1$ than A_1.

The course of a quantum-mechanical treatment can be found in a paper by Visscher (1960). Visscher shows (equation (5)) that the resonance scattering amplitude involving a transition from a state with α phonons to a state with β phonons is

$$W = \sum_n \frac{\langle \beta | \exp(-ip \cdot x) | n \rangle \langle n | \exp(-ip \cdot x) | \alpha \rangle}{\omega_a - \omega + (n - \alpha)\omega_1 + i\gamma}.$$

By using equations (8) of Visscher's paper, this has the time dependence

$$\exp\{i[\omega+(\beta-\alpha)\omega_1]t\},$$

which is the same as the classical expression.

Acknowledgments

It is a pleasure to thank P Schofield for helpful discussions.

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