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Vibrational properties of the modulated spring chain: neutron cross section and dual spectrum

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Abstract. The inelastic neutron cross section is calculated for a modulated spring chain, which models an incommensurate composite system. We obtain a difference equation for the neutron cross section which has an exact and analytic solution for a periodic modulation. The structure of the neutron spectrum is studied as functions of wavevector and energy transfers for a wide range of model parameters. In general the spectra consist of bands of intensity, except at special wavevectors where bands collapse to a singular structure characteristic of normal modes of vibration.

1. Introduction

A harmonic chain of atoms with modulated springs (stretching forces) has been examined, during the past decade, with a view to understanding excitations in incommensurate phases of solids (Currat and Janssen 1988). The modulation of the springs models the change in local environment of the other atoms which surround a chain in an incommensurate composite system. In this and companion papers (Lovesey 1989, Lovesey and Westhead 1990 (hereafter referred to as paper I)) we report an extensive study of the vibrational properties of the modulated spring chain that exploits analytic methods which produce exact results. One value of our findings is a reference against which to assess the physical significance of measured features. Discrepancies will arise from effects due to inter-chain couplings, anharmonicity and coupling to the energy density, for example.

Here we focus attention on the vibrational spectrum observed in the so-called inelastic one phonon coherent neutron scattering cross section. For a regular crystal this cross section, in the harmonic approximation, vanishes unless there is conservation of energy and wavevector for annihilation or creation of a single phonon (Lovesey 1987), and measurements yield phonon dispersion curves. Modulation of the springs in a harmonic system is shown to radically change the vibrational spectrum. A review of previous work is provided by Currat and Janssen (1988): see also Garcia *et al* (1989).

At a technical level, the spectrum reported here is for the dual model of the one examined in paper I. In the latter we provide the density of vibrational states, and local thermodynamic quantities, whereas the neutron scattering cross section is the vibrational spectrum of spatial Fourier components u_q where q is the wavevector. Duality and the metal insulator transition is reviewed by Sokoloff (1985a, b).

Another interesting aspect is the comparison of neutron cross sections for the modulated spring chain and an analogous magnetic system in which atomic moments vary in magnitude from site to site (Lovesey 1988, Brackstone and Lovesey 1989, Lantwin 1990). Even though in both situations scattering events come from a coherent spatial superposition of atoms on a regular lattice there are basic differences in the spectra. These arise because in scattering from lattice vibrations the lattice structure is directly involved, whereas in scattering from magnetic moments it is the magnetic configuration which counts. In particular, the vibrational spectrum is singular when the neutron wavevector q coincides with a reciprocal lattice vector of the system with periodic springs. This arises because full translational invariance is restored to the scattering problem for these particular values of the wavevector. However, magnetic scattering does not assume a special form for this choice of wavevector; there is strong amplitude variation with changes in q but minimal energy dispersion.

The model that we solve was introduced by de Lange and Janssen (1981) and it possesses a simple cosine modulation of the springs as a function of the equilibrium atomic site positions. Results from this form of modulation enable us to readily solve the model with squared-cosine modulation (Cohan and Weissmann 1983). We assume that the atoms are equally spaced in the equilibrium state. While this is not consistent with Hooke's law it has strong appeal because the resulting model can be solved exactly using simple analytic methods.

We couch our discussion in terms of the lattice displacement Green function defined such that it is directly related to the one-phonon neutron cross section. It is constructed from an equation of motion, which has the form of a difference equation in reciprocal (wavevector) space. This equation, for a cosine modulation, is provided in section 2, together with some frequency sum rules that are useful in understanding the basic features of the spectrum. Explicit results for the Green function are given in section 3 for q equal to a reciprocal lattice vector. The general result is contained in section 4, and specific examples of the spectrum are discussed in some detail in section 5. In section 6 we discuss the squared cosine modulation. Conclusions and comments are gathered at the end of the paper.

2. Equation of motion and sum rules

The basic model is the same as the one studied in paper I. Atoms of mass m are bound harmonically and the equilibrium configuration is a regular chain with unit lattice spacing (the vibrational spectrum of atoms on a disordered chain is considered by Kim and Nelkin 1973). The potential energy is

$$\frac{1}{2} \sum_n \alpha_n (u_n - u_{n-1})^2. \quad (1)$$

Here, u_n is the displacement (assumed to be small compared to the lattice spacing) of the atom at the site labelled by integer n , and α_n is the spring constant

$$\alpha_n = m\{\alpha - \gamma \cos(nQ + \Delta)\} \quad (2)$$

in which the strength γ , phase Δ and wavevector of magnitude Q specify the sinusoidal modulation.

We choose to obtain the spectrum of spontaneous fluctuations of the spatial Fourier components $\{u_q\}$ from a Green function $G(q, \varepsilon)$ where ε is the energy parameter; the spectrum is the dual of the density of states provided in paper I. In a neutron scattering experiment, q and $\omega = \varepsilon^{1/2}$ are the changes in wavevector and energy in the scattering process.

The effect of the sinusoidal modulation in the equation of motion is to couple u_q with $u_{q \pm Q}$, i.e. it has the form of a difference equation. In view of this, we are led to consider a Green function $P(n, \varepsilon)$ that yields the temporal Fourier transform of the displacement correlation function $\langle u_q^* u_{q+nQ}(t) \rangle$. By construction $P(0, \varepsilon) = G(q, \varepsilon)$ is the Green function of interest in calculating the neutron cross section, proportional to (Lovesey 1987)

$$-(1/\pi)\{1 - \exp(-\omega/T)\}^{-1} \text{Im } G(q, \varepsilon) \quad (3)$$

where T is the temperature and the imaginary part of G is obtained with $\varepsilon = \omega^2$, $\omega + i\eta$ and $\eta \rightarrow 0^+$. In the absence of spring modulation ($\gamma = 0$),

$$mG(q, \varepsilon) = 1/(\varepsilon - \omega_q^2) \quad (4)$$

and

$$\omega_q = \omega_0 |\sin(q/2)| \quad (5)$$

where $\omega_0^2 = 4\alpha$ is the phonon dispersion in a chain of atoms.

The equation of motion for $P(n, \varepsilon)$ is directly obtained from the equation for u_q constructed by Fourier transformation of the real space equation of motion. We find

$$E_n P(n, \varepsilon) = (\delta_{n,0}/m) + T_n P(n-1, \varepsilon) + T_{n+1} P(n+1, \varepsilon) \quad (6)$$

where

$$E_n = \varepsilon - \omega_{q+nQ}^2 \quad (7)$$

and

$$T_n = -2\gamma \sin\{\frac{1}{2}(q+nQ)\} \sin\{\frac{1}{2}(q+(n-1)Q)\}. \quad (8)$$

Note that the phase Δ in the spring constant (2) does not appear in the equation for $P(n, \varepsilon)$. This is because the equation is constructed for Q an irrational multiple of 2π , i.e. for an incommensurate system. Change of the phase Δ is a continuous symmetry of such a system and therefore we expect that the neutron cross section will be independent of Δ .

Subsequent sections describe solutions of (6) for rational $Q = 2\pi M/N$ (where M and N are coprime integers) obtained by algebraic methods. At this juncture it is to be noted that (6) is the dual of the real-space Green function equation of motion. It is constructed for Q irrational since this is the situation of physical interest. The fact that we exploit the periodicity which results from taking Q rational to construct algebraic solutions is a mathematical trick unconnected to the physical properties of interest. Of course, an irrational value of Q can be approached to arbitrary accuracy using a sequence of rationals, for example a Fibonacci sequence, though we do not

pursue this topic. In the next section we describe solutions for special values of q and then report the general solution.

Before this we report sum rules which can be constructed from the equation of motion. These are useful in unravelling general features of the spectrum, as well as gauging its shape and sensitivity to the spring modulation. We find for the integral

$$-\frac{m}{\pi} \int_0^\infty d\varepsilon \varepsilon^n \operatorname{Im} G(q, \varepsilon)$$

the results

$$\begin{aligned} 1 & \quad \text{for } n = 0 \\ \omega_q^2 & \quad \text{for } n = 1 \\ \omega_q^4 + T_0^2 + T_1^2 & \quad \text{for } n = 2. \end{aligned} \tag{9}$$

From these we conclude that the spectrum is normalised to unity and the mean value (i.e. the sum rule for $N = 1$) is independent of the modulation when this takes the form of a simple cosine as in (2). Furthermore, intensity shifts to higher frequencies with increasing q and the modulation influences the high frequency components more strongly than the low frequency components. Note that $T_0^2 + T_1^2$ is proportional to ω_q^2 and vanishes in the long-wavelength limit. Dependence on the N -fold periodicity occurs in the second frequency sum rule. This is finite in the limit $N \rightarrow \infty$ indicating a bounded spectrum which is an envelope of N bands having a mean square width $(2\gamma/\omega_0)^2 \omega_q^2 [\sin^2(\{q + Q\}/2) + \sin^2(\{q - Q\}/2)]$.

3. Special cases; $q = 2\pi s/N$, $s = 0, 1, 2, \dots$

In the next section we obtain an expression for $G(q, \varepsilon)$ by iteration of the infinite set of difference equations (6). This produces infinite continued fractions for the quantities $P(\pm 1, \varepsilon)/P(0, \varepsilon)$. In the general case this yields a spectrum which consists of N finite bands of intensity located where the Green function given by (18) is pure imaginary. A special form for the expression for G arises in the case when one or more of the parameters $\{T_n\}$ is zero because in this case the iteration procedure stops after a finite number of steps. This of course gives finite continued fractions and yields a spectrum which is singular, consisting of a number of delta functions rather than finite bands. Consideration of these special cases first gives valuable insight into the structure of the spectrum.

Observing the form of T_n given in (8) we see that if we set $q = 2\pi s/N$ with $s = 0, 1, 2, \dots$ (i.e. q is equal to a reciprocal lattice vector) then T_n and T_{n+1} will be zero if we can find an integer p such that

$$pN - nM = s \tag{10}$$

We are interested in solutions of (10) for the case where N and M have no common factors, i.e. they are coprime. When this condition holds a standard theorem in number theory shows that (10) will have integer solutions in the range $0 \leq p \leq (M-1)$, $0 \leq n \leq (N-1)$, for any integer s . So, for q equal to any reciprocal lattice vector there exists n such that $T_n = T_{n+1} = 0$. The resulting singular structure in the spectrum at

the reciprocal lattice vectors allows us to determine the periodicity of the modulation from the dynamics.

Let us examine the case of $s = 0$, i.e. $q = 0$. The solution of (10) is then $p = n = 0$, so $T_0 = T_1 = 0$. We can obtain $G(q, \varepsilon)$ directly by setting $n = 0$ in equation (6):

$$E_0 G(q, \varepsilon) = (1/m) + T_0 P(-1, \varepsilon) + T_1 P(1, \varepsilon) \tag{11}$$

for $s = 0$ this gives

$$mG(0, \varepsilon) = 1/\varepsilon. \tag{12}$$

This shows that the spectrum is singular and independent of the spring modulation. Physically this stems from the fact that it costs no energy to make a uniform translation of the atoms.

A second special q of interest is $q = Q$ which corresponds to setting $s = M$ in (10). In this case solution of (10) yields the result $T_0 = T_{N-1} = 0$. The equation for G is then

$$E_0 - \{1/mG(Q, \varepsilon)\} = \{T_1^2/(E_1 - T_2^2/[E_2 - \dots T_{N-2}^2/E_{N-2}])\}. \tag{13}$$

The finite continued fraction on the right-hand side is readily expressed in compact and convenient form.

To this end we introduce two sets of functions $\{p_n\}$ and $\{q_n\}$ which are constructed recursively from

$$E_n p_n = T_n p_{n-1} + T_{n+1} p_{n+1} \tag{14}$$

with the same equation for q_n , starting from the initial values $p_0 = q_1 = 0$ and $p_1 = q_0 = 1$. It is easy to demonstrate that

$$\{T_1^2/(E_1 - T_2^2/[E_2 - \dots T_n^2/(E_n - h)])\} = \frac{-T_1(q_{n+1}T_{n+1} - q_n h)}{p_{n+1}T_{n+1} - p_n h}. \tag{15}$$

The expression on the right-hand side of (13) is obtained with $h = 0$ and $n = (N - 2)$; the general form of (15) is required in the following section.

From (13) and (15) we arrive at the result

$$mG(Q, \varepsilon) = \frac{1}{E_0 + (T_1 q_{N-1}/p_{N-1})}. \tag{16}$$

Since p_n and q_n are polynomials of order $(n - 1)$ and $(n - 2)$ respectively, the denominator of $G(Q, \varepsilon)$ is a polynomial of order $(N - 1)$. Hence, the spectrum $\text{Im } G(Q, \varepsilon)$ will in general consist of $(N - 1)$ delta functions. A specific example is provided in section 5.

If we apply the same method to a general s we obtain a Green function whose denominator is a polynomial of order at most $(N - 1)$. The spectrum is therefore singular, consisting of at most $(N - 1)$ delta functions.

We conclude that the spectrum is singular for q equal to a reciprocal lattice vector. In the next section we construct the spectrum for arbitrary q and find that it consists of N bands.

4. Arbitrary q

For this case to obtain $G(q, \epsilon)$ it is necessary to construct continued fractions for $P(\pm 1, \epsilon)/P(0, \epsilon)$. Because they are periodic $T_{n+N} = T_n$ and $E_{n+N} = E_n$, it is possible using identities like (15) to express them in simple algebraic forms in terms of the polynomials $\{p_n\}$ and $\{q_n\}$. For example, $h = P(1, \epsilon)/P(0, \epsilon)$ is the fixed point of the linear transformation (15) evaluated for $n = N$. A similar scheme exists for $P(-1, \epsilon)/P(0, \epsilon)$. The results, together with the Wronskian relation

$$p_{n+1}q_n - p_nq_{n+1} = (T_1/T_{n+1}) \tag{17}$$

enable us to obtain a closed algebraic form for $G(q, \epsilon)$. Further details of the mathematical steps involved can be found in Lovesey (1988) and paper I.

Assembling results, the required expression is

$$mG(q, \epsilon) = \frac{(\pm)p_N/T_1}{\sqrt{(p_{N+1} + q_N)^2 - 4}}. \tag{18}$$

Except at $q = 2\pi s/N$, $s = 0, 1, 2, \dots$, the spectrum $\text{Im}G(q, \epsilon)$ is finite within bands for which

$$(p_{N+1} + q_N)^2 - 4 < 0. \tag{19}$$

The function on the left hand side is a polynomial in ϵ of order $2N$. Stable solutions occur for $\epsilon = \omega^2 > 0$. The choice of sign in (18) is related to the convergence of the continued fractions and yields a spectrum which is strictly positive within the bands, as demonstrated in paper I.

In describing the spectrum it is useful to consider periodic $\{\lambda_i\}$ and antiperiodic $\{\mu_i\}$ roots:

$$p_{N+1}(\lambda_i) + q_N(\lambda_i) - 2 = 0 \tag{20}$$

$$p_{N+1}(\mu_i) + q_N(\mu_i) + 2 = 0 \tag{21}$$

where $i = 1, 2, \dots, N$. It can be shown that the inequality (4.3) is satisfied for ϵ within a band specified by a pair λ_i, μ_i and gaps are alternatively specified by pairs μ_i, μ_{i+1} and λ_i, λ_{i+1} . A simple example is displayed in figure 1, about which we will have more to say in the next section.

The results

$$\sum_i \mu_i = \sum_i \lambda_i = (N/2) \tag{22}$$

are analogous to the first frequency moment of the spectrum and demonstrate that its width is insensitive to the spring modulation. The results

$$\prod_i \lambda_i = (-1)^N (T_0 T_1 \dots T_{N-1}) (p_{N+1} + q_N - 2)|_{\epsilon=0} \tag{23}$$

$$\prod_i \mu_i = (-1)^N (T_0 T_1 \dots T_{N-1}) (p_{N+1} + q_N + 2)|_{\epsilon=0} \tag{24}$$

can be useful in certain calculations.

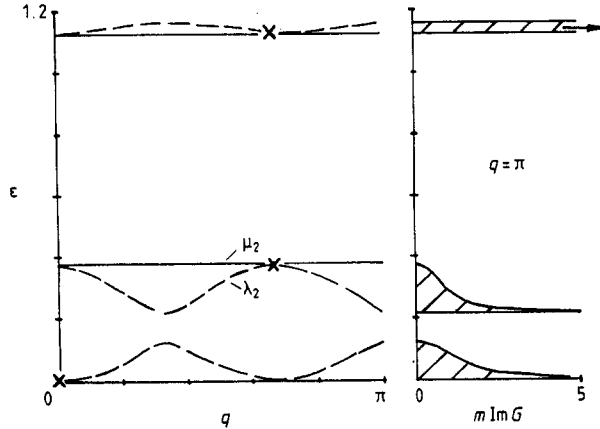


Figure 1. The energy band structure and neutron scattering spectrum are shown for periodicity $N = 3$ and $\nu = \frac{1}{2}$. Crosses in the band structure, shown on the left-hand side, denote positions of the delta functions which occur in the spectrum for $q = 0$ and $q = 2\pi/3$. The band edges $\{\mu_i\}$ are at $0, 0.375$ and 1.125 , and the band edges $\{\lambda_i\}$ are shown as broken curves. The neutron spectrum is shown for $q = \pi$ on the right-hand side of the figure. The actual quantity shown is $m \text{Im} G(q, \varepsilon)$ for which the total area is π . The minimum value of the high-frequency contribution to the spectrum is 49.98 , and the band edges ε_i are $(0, 0.125)$, $(0.215, 0.375)$ and $(1.125, 1.159)$.

5. Examples

In this section we report specific examples of the neutron cross section for the modulated spring chain. Particular attention is given to the case $N = 3$, $Q = 2\pi M/3$ where $M = 1, 2$. This is sufficiently simple to record in detail, yet nicely displays the essential features of the spectrum of a modulated chain. The size and complexity of the polynomials $\{p_n\}$ and $\{q_n\}$ for large n , greater than $n = 5$ say, precludes analytic manipulations for large periodicities. However, the spectrum is easily constructed numerically as we shall see.

The Green function for $N = 3$ is obtained from the general result (18) using

$$p_3/T_1 = (E_1 E_2 - T_2^2) \left(\prod_i T_i \right)^{-1} \quad (25)$$

and

$$p_4 + q_3 = (E_1 E_2 E_3 - T_1^2 E_2 - T_2^2 E_3 - T_3^2 E_1) \left(\prod_i T_i \right)^{-1} \quad (26)$$

where $\prod_i T_i = T_0 T_1 T_2$. At this point we can note that the effect of changing $M = 1$ to $M = 2$ is simply to permute the parameters T_n and E_n : $T_1 \rightarrow T_3$, $T_2 \rightarrow T_2$, $T_3 \rightarrow T_1$, and, $E_1 \rightarrow E_2$, $E_2 \rightarrow E_1$, $E_3 \rightarrow E_3$. This permutation leaves the right-hand side of both (25) and (26) unchanged so that the Green function is invariant under $M = 1$ going to $M = 2$.

Using (6) and (7) we obtain from (26)

$$\prod_i T_i(p_4 + q_3 \pm 2) = \varepsilon^3 - \frac{3}{2}\varepsilon^2 + \frac{9}{16}(1 - \nu^2)\varepsilon - (1 \mp 2\nu)(1 \pm \nu)^2 [\sin(q/2)\{1/4 - \cos^2(q/2)\}]^2 \quad (27)$$

where $\nu = (\gamma/2\alpha)$ and ε is measured in units of the maximum phonon frequency ω_0 . Note that the factors of $\prod_i T_i$ cancel in the result for $G(q, \varepsilon)$ and that quantities $\prod_i \lambda_i$ and $\prod_i \mu_i$ are given by the appropriate constant term in (28).

For $q = 0$,

$$E_1 E_2 - T_2^2 = \varepsilon^2 - \frac{3}{2}\varepsilon + \frac{9}{16}(1 - \nu^2)$$

so $mG(0, \varepsilon) = 1/\varepsilon$ in accord with general result obtained in section 3.

The constant term on the right-hand side of (28) also vanishes for $q = 2\pi/3$. This corresponds to $q = Q$ for $M = 1$ or to $q = Q/2$ for $M = 2$. For this value of q the spectrum is singular. From either (25) and (28) evaluated for $q = 2\pi/3$, or (16), we find,

$$2mG(2\pi/3, \varepsilon) = \frac{1}{\varepsilon - \varepsilon_1} + \frac{1}{\varepsilon - \varepsilon_2} \quad (28)$$

where $\varepsilon_1 = 3(1 - \nu)/4$ and $\varepsilon_2 = 3(1 + \nu)/4$. Evidently $\text{Im } G(2\pi/3, \varepsilon)$ is the sum of two delta functions located at ε_1 and ε_2 which are equidistant above and below the phonon dispersion at $q = 2\pi/3$ in an unmodulated chain. In figure 1 these are indicated by crosses together with the singular contribution at $\varepsilon = 0$. The result (28) satisfies the sum rules provided in section 2.

For other values of q the spectrum consists of three bands; results for $\nu = \frac{1}{2}$ are displayed in figure 1. With $\nu = \frac{1}{2}$, the maximum value consistent with stability, the band edge frequencies $\{\mu_i\}$ are independent of q and have values 0 , $\frac{3}{8}$ and $\frac{9}{8}$. The band edge frequencies $\{\lambda_i\}$ are the roots of (28) taken with the lower sign. Note that the coefficient of ε^2 is $-\frac{3}{2}$. In consequence, the sum of the band edge frequencies is $\frac{3}{2}$, consistent with (22). From figure 1 it is seen that $\{\lambda_i\}$ display strong dispersion.

The actual spectrum is shown in figure 1 for $q = \pi$. It consists of three bands; the two low frequency bands tend smoothly to zero at their high frequency edges, whereas a square root singularity, characteristic of one dimensional motion, occurs at the other four band edges. The low frequency bands are weak in intensity compared with the high frequency component, which is expected at this value of q given that the spectrum is normalised to π and the first frequency moment is $\pi\omega_q^2$. Decreasing q leads to a shift in intensity to the low frequency bands, with a total concentration at $\varepsilon = 0$ in the limit $q \rightarrow 0$.

Examples of spectra for $N = 4$ and $N = 5$ are shown in figure 2. These are generated numerically from (18) and (14). We chose values of q which are at the Brillouin zone edges where the bands are widest. Each spectrum consists of N bands, the width of the bands decreases as N increases.

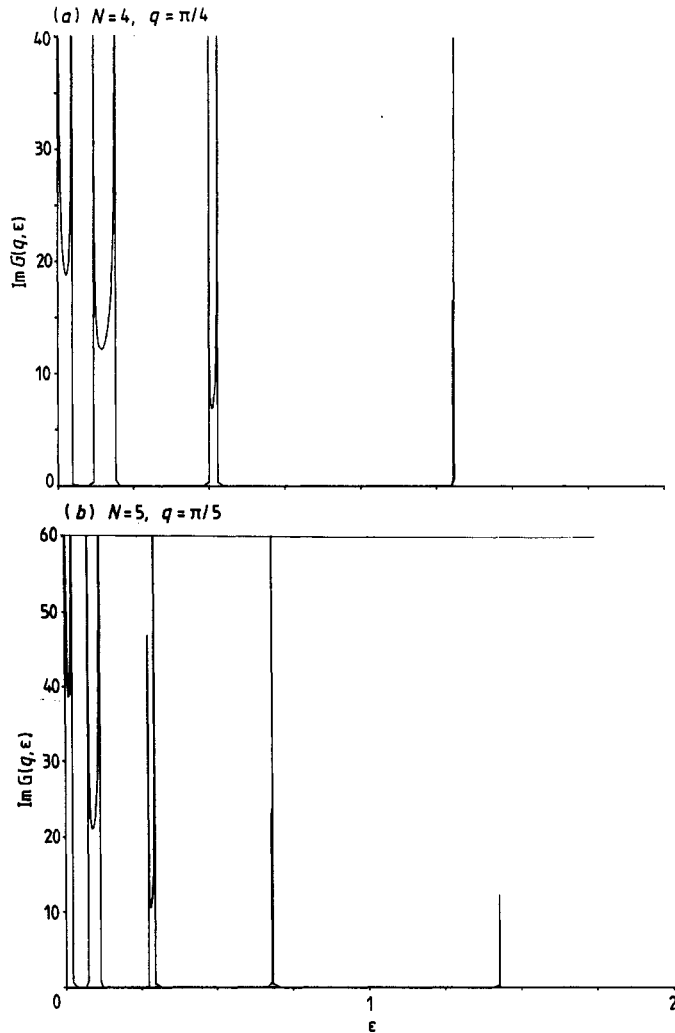


Figure 2. The neutron scattering spectrum is shown for periodicity in (a) $N = 4$, $q = \pi/4$ and (b) $N = 5$, $q = \pi/5$. Both cases have $M = 1$ and $\nu = \frac{1}{2}$ as in figure 1, and $\alpha = \gamma = 0.25$. The spectrum is shown as a function of ϵ for values of q that lie at the Brillouin zone edges, where the bands are widest.

6. Squared-cosine spring modulation

The paper by Cohen and Weissmann (1983) motivates us to consider different forms of the spring modulation, particularly powers of the simple cosine wave. We note here that the squared-cosine modulation can be solved easily by using the above work and the identity $2 \cos^2(\theta) = 1 + \cos(2\theta)$.

We consider the spring constants to take the form

$$\alpha_n = m\{\alpha' - \gamma' \cos^2(nQ' + \Delta')\} \quad (29)$$

where $Q' = \pi M/N$, so that we have a system of periodicity N . Using the above identity we see that this system can be transformed into a system with cosine modulation where

the parameters in (2) become

$$\begin{aligned}\alpha &= \alpha' - (\gamma'/2) & \gamma &= \gamma'/2 \\ \Delta &= 2\Delta' & Q &= 2Q'\end{aligned}\tag{30}$$

Using this transformation the full solution for the system with squared-cosine modulation is obtained.

7. Conclusions

The work reported provides a comprehensive algebraic solution for the dual spectrum, and corresponding inelastic neutron cross section, of the (harmonic) modulated spring chain. For a modulation wavevector $Q = 2\pi M/N$ the spectrum consists of N bands for an arbitrary external wavevector q . In general, the spectrum displays inverse square root singularities at band edges, characteristic of a one dimensional model, but this feature in the neutron cross section is absent for some high symmetry values of q .

Bands collapse to produce singular (delta function) contributions in the cross section for special values of q . One such value is $q = 0$ for which the spectrum, as expected, consists of a single delta function at zero energy, independent of the spring modulation. A second expected special case is when q matches the harmonic of the modulation, $q = Q$. For this wavevector the spectrum consists of $(N - 1)$ delta functions located at energies which depend explicitly on the form of the spring modulation. Similar singular behaviour occurs whenever q is equal to any reciprocal lattice vector.

These singular features of the dual spectrum do not arise in the spectrum for scattering from transverse spin fluctuations in a longitudinally modulated magnet. This result, mentioned already in the introduction, stems from fundamental differences in the nature of coherent scattering processes from lattice vibrations and spin fluctuations.

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