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LETTER TO THE EDITOR

Exact properties of the mixed mass modulated spring constant model

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Abstract. Properties of the modulated spring constant model, which has been used to explore the dynamic features of incommensurate crystal phases, are expressed in analytic form. Results for the density of states, average energy and mean square velocity are given. Particular attention is given to the effects and properties of a mass defect.

The lattice vibrations of a compound in an incommensurate structural state are radically different from those of a crystal with lattice periodicity. For one thing, there is no Brillouin zone and the wavevector is not a good quantum number. A model used to explore the intriguing dynamics of an incommensurate system consists of a chain of particles with harmonic interactions that vary from site to site. The modulation of the spring constant is incommensurate with the chain, so the model does not possess translational invariance. It mimics an anisotropic compound in which some atoms, in an otherwise periodic crystal, form one-dimensional chains.

Previous studies of the model, reviewed by Currat and Janssen (1988), have used numerical techniques to reveal properties of the energy spectrum, and it has many features in common with the Hofstadter butterfly energy spectrum of electrons in a tight binding model subject to an external magnetic field. Such studies are, of course, restricted to commensurate configurations. If the modulation wavevector $Q = 2\pi M/N$ the number of band is proportional to the periodicity integer N. An irrational number can be approached for suitably large N (using a Fibonacci series, for example). Since the width of the spectrum is independent of N it becomes increasingly fragmented with increasing N. Therefore the precise form of the spectrum is really merely a mathematical curiosity given the finite accuracy available in experimental and numerical studies. From this standpoint, there is much interest in obtaining analytic expressions for physical properties of models with relatively low-order periodicity.

We have used the analytic technique developed in previous papers (Lovesey 1988a, b) to obtain physical properties of the periodic modulated spring model, including the effect of a defect. Here we report results for the density of states, average energy, and the mean square velocity observed in the second-order Doppler shift of a Mössbauer peak.

The quantities of interest are obtained from a displacement Green function, which is conveniently defined in terms of normal coordinates. Let these be chosen real, and

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denoted by $f_\sigma(n)$ where σ is a normal coordinate label and the integer n defines an atomic site. The orthonormality and closure relations are

$$\sum f_\sigma(n) f_{\sigma'}(n) m_n = \delta_{\sigma,\sigma'}$$
$$\sum_\sigma f_\sigma(n) f_\sigma(n') = \delta_{n,n'}/m_n \tag{1}$$

where m_n is the particle mass, and the lattice displacement

$$u_n = \sum_\sigma f_\sigma(n)(b_\sigma^+ + b_\sigma)/(2\omega_\sigma)^{1/2} \tag{2}$$

in which b_σ, b_σ^+ are standard Bose operators and ω_σ is the eigenfrequency in the diagonalised Hamiltonian. With these definitions, the Green function

$$G(n, n'; \mu) = \sum_\sigma f_\sigma(n) f_\sigma(n')/(\mu - \omega_\sigma^2) \tag{3}$$

satisfies the equation of motion

$$(\mu m_n - \alpha_n - \alpha_{n+1}) G(n, n'; \mu) = \delta_{n,n'} - \alpha_n G(n-1, n'; \mu) - \alpha_{n+1} G(n+1, n'; \mu). \tag{4}$$

For the modulated spring model

$$\alpha_n = m(\alpha - \gamma \cos(nQ + \Delta)). \tag{5}$$

Here, γ is the strength of the modulation of the force constants attributed to changes in the local environment of atoms along the chain.

If all particles in the chain have mass m except at the site labelled s where there is a mass $m' = m(1 - \lambda)$ then (see, for example, Lovesey 1986)

$$G(n, n'; \mu) = P(n, n'; \mu) + m\lambda\mu P(n, s; \mu) P(s, n'; \mu)/(1 - m\lambda\mu P(s, s; \mu)). \tag{6}$$

In this expression, which can be deduced from the identity

$$\mu \sum m_n G(n, n'; \mu) = 1$$

P is the Green function for the perfect chain, i.e. the solution of (4) for $m_n = m$ for all n. An appropriate expression for P is obtained, using the method described previously (Lovesey 1988a, b), in terms of polynomials in p_n, q_n which satisfy the recursion relation (p_n, $q_n \equiv R_n$)

$$R_{n+2} + R_{n+1}(\mu - \alpha_n - \alpha_{n+1}) + R_n \alpha_n^2 = 0 \tag{7}$$

with the initial conditions $p_0 = q_1 = 0$, $p_1 = q_0 = 1$.

Let us now record expressions for quantities of physical interest, and then present some specific results. The density of states $Z(\omega)$, normalised to unity for $\omega \geqslant 0$, is

$$z(\omega) = (-2\omega/\pi N) \sum_n m_n \operatorname{Im} G(n, n; \omega^2 + \mathrm{i}0^+). \tag{8}$$

While the mean square velocity $\langle v_n^2 \rangle$ and the average energy can be obtained from this expression, it is more convenient to exploit a result obtained from (2) and (3)

$$m_n \langle v_n^2 \rangle = T\left(1 + 2\sum_{k=1}^{\infty} (1 + m_n \theta_k G(n, n; -\theta_k))\right). \tag{9}$$

Here, T is the temperature ($\hbar = k_B = 1$) and $\theta_k = (2\pi k T)^2$. The defect energy, defined as the change in the average energy per particle

$$\sum_{n=0}^{N-1} m_n \langle v_n^2 \rangle / N$$

due to the defect, is readily shown to be

$$\varepsilon(s) = (2T/N) \sum_{k=1}^{\infty} \theta_k \frac{\mathrm{d}}{\mathrm{d}\mu} \ln(1 - m\lambda\mu P(s, s; \mu))|_{\mu = -\theta_k}. \tag{10}$$

The technique for calculating P outlined here and developed in detail in previous papers applies to modulation wavevectors $Q = 2\pi M/N$ with arbitrary integers M, N. We report results for $N = 3$. This simple case illustrates the main features of the modulated spring constant model with a mass defect. Some features are admittedly exaggerated by the small periodicity number and might not be quite so significant in applications to realistic situations modelled by larger N. A more extensive investigation of the N-dependence of the modulated spring model will be prepared.

To evaluate the expressions provided for the physical quantities we require explicit expressions for $P(s, s; \mu) = P(s; \mu)$. The two identities

$$m \sum_{s=0}^{N-1} P(s; \mu) = (p'_{N+1} + q'_N)/[(p_{N+1} + q_N)^2 - (2\alpha_0 \ldots \alpha_{N-1})^2]^{1/2} \tag{11}$$

and

$$m \sum_{n=0}^{N-1} P(s, n; \mu) P(n, s; \mu) = -P'(s; \mu) \tag{12}$$

are also useful; the prime denotes differentiation with respect to μ. We find for $N = 3$

$$p_4 + q_3 \mp 2\alpha_0\alpha_1\alpha_2 = \begin{cases} J(\mu) = \mu(-\mu^2 + 6\alpha\mu - 9\alpha^2 + \tfrac{9}{4}\gamma^2) \\ J(\mu) + C \end{cases} \tag{13}$$

where $C = 4\alpha^3 - 3\alpha\gamma^2 - \gamma^3 \cos 3\Delta$ and

$$mP(s; \mu) = [J'(\mu) - 3\mu\gamma \cos(\Delta + (s-1)Q)]/\{3[J(\mu)(J(\mu) + C)]^{1/2}\}. \tag{14}$$

Combining the various expressions, we find that the density of states of the pure system is

$$z(\omega) = (2\omega/3\pi)|J'|[-J(J + C)]^{-1/2} \tag{15}$$

for $J(J + C) < 0$ and zero otherwise; the prime denotes differentiation with respect to $\mu = \omega^2$. Solutions of $J(J + C) = 0$ for which $\mu \geqslant 0$ give the band edges. The periodic roots are solutions of $J = 0$, namely $\mu = 0, 3(2\alpha \pm |\gamma|)/2$ from which we see that $2\alpha > |\gamma|$. An additional constraint on the magnitude of γ is obtained from the antiperiodic roots $J + C = 0$, and this reduces to the condition $C \geqslant 0$, i.e. the range of allowed values of γ depends on the phase Δ and reflects the requirement that $\{\alpha_n\}$ are positive for a stable system.

The average energy in the pure system shows some variation as a function of the phase Δ, although this is not significant at high tempertures where the energy approaches T. The minimum energy occurs at $\Delta = 0, 2\pi/3$ for $0 < \gamma < \alpha$ and at $\pi/3, \pi$ for $-\alpha < \gamma < 0$, and the variation is less than 5% at a temperature = 0.02 in units of $\alpha^{1/2}$. For allowed states with $|\gamma| > \alpha$ the minimum energy occurs for $\gamma = \gamma(\Delta)$ which is a

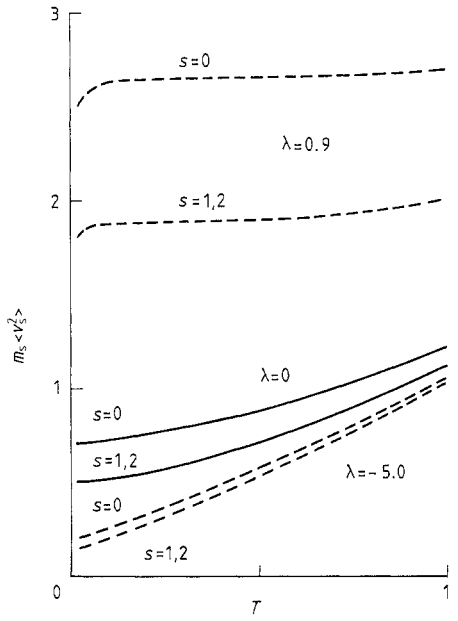

Figure 1. The mean square velocity is displayed as a function of temperature (in units of $\alpha^{1/2}$) for $\Delta = 2\pi/3$, $\gamma = 0.95$ in a pure system (full curves) and for mass defects characterised by $\lambda = 0.9$ (upper curves) and $\lambda = -5.0$. For $\Delta = 2\pi/3$ the sites $s = 1, 2$ are equivalent.

solution of the cubic equation $C = 0$. Figure 1 shows the mean square velocity of a particle in the pure system as a function of temperature for $\Delta = 2\pi/3$ and $\gamma = 0.95\alpha$. For this choice of Δ there are just two distinct sites, and the difference in $\langle v_s^2\rangle$ for these sites is minimal beyond about $T = 1$ at which the values are essentially classical.

In contrast to this, classical values of $\langle v_s^2\rangle$ for a light mass defect are achieved at much higher temperatures as illustrated in figure 1. There is a slight variation of $\langle v_s^2\rangle$ for $\lambda = 0.9$ with temperature up to $T = 1$, while a heavy mass $\lambda = -5.0$ has a comparatively much smaller velocity and a temperature variation akin to that obtained in a pure system. The Δ-variation of $\langle v_s^2\rangle$ for $T = 0.02$ is shown in figure 2. As might be expected, the phase dependence is quite pronounced for the light mass defect.

A heavy mass defect decreases the normal mode frequencies from their values in the corresponding pure case, whereas a light mass defect increases the frequencies and generates localised modes outside the band intervals of the pure density of states. In consequence, the defect energy, as defined here, is negative for $\lambda < 0$ and positive for $0 < \lambda < 1$. The Δ-dependence of $\varepsilon(s)$ for $\lambda = 0.9$ is shown in figure 2 for $s = 0$ and it possesses a strong minimum at $2\pi/3$; values of $\varepsilon(s)$ for $s = 1, 2$ are shifted relative to those for $s = 0$ such that the minima occur at $\Delta = \pi/2, \pi/3$ respectively. The temperature dependence of the average energy of a pure system and $\varepsilon(s)$ for $\lambda = 0.9, -5.0$ is shown in figure 3. A heavy defect reduces the energy of the system, but the reduction is essentially independent of m' for $m' > 6m$.

The strong λ-dependence of quantities shown in figures 1–3 is evident in the corresponding high-temperature approximations. We find that the mean square velocity of a mass defect approaches

$$\langle v_s^2\rangle = (T/m')\{1 + (\alpha_s + \alpha_{s+1})/[12mT^2(1-\lambda)]\}$$

and the corresponding defect energy is

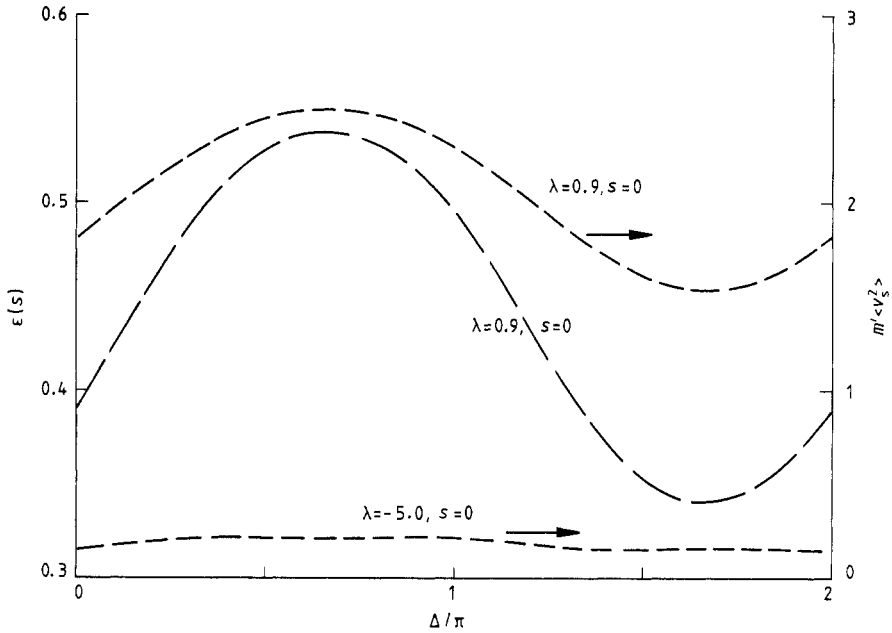

Figure 2. Defect energy and mean square velocity are shown as functions of Δ at a reduced temperature = 0.02, $\gamma = 0.95$. The defect energy is for $\lambda = 0.9$, and $m'\langle v_s^2 \rangle$ is given for $s = 0$ and $\lambda = 0.9$ (upper curve) and $\lambda = -5.0$.

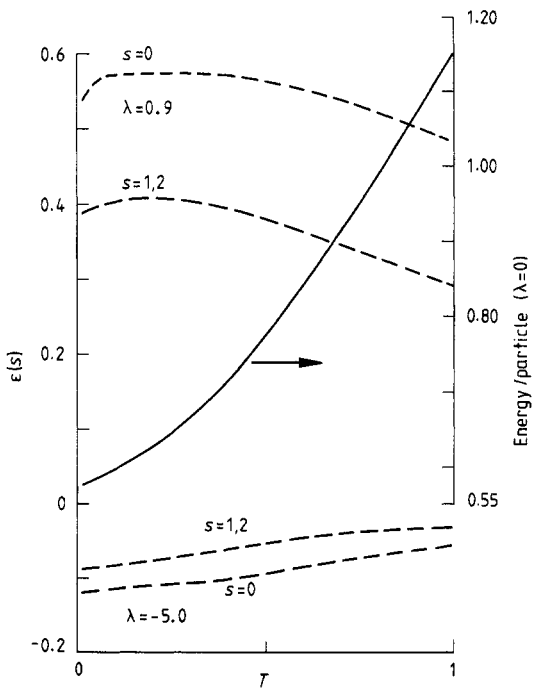

Figure 3. The average energy per particle is shown as a function of reduced temperature in the range 0.02 to 1.0 for $\Delta = 2\pi/3$ and $\gamma = 0.95$. Defect energies for $\lambda = 0.9$ and -5.0 are displayed for the two distinct sites $s = 0, 1$.

$$\varepsilon(s) = \lambda(\alpha_s + \alpha_{s+1})/[12mTN(1-\lambda)].$$

These results are derived from (9) and (10) using the large-μ form of $P(s;\mu)$ which is readily obtained from (3).

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