Effect of Randomness in Layer Thickness on the Phonon Structure of Superlattices

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Abstract

A new type of disorder may occur in artificial superlattices, arising from the randomness in the layer thicknesses. We treat here its effect on the phonon structure. A modification of the infinite order perturbation theory by Wu *et al.* (1974*a*, 1974*b*) serves well to give satisfactory solutions of the problem.

1. Introduction

A lack of complete control over the preparation of samples often brings about defects and irregularities in the structure of artificial superlattices (Schuller 1980). Among the various factors of disorder, there is one unique to such layered materials—the randomness in the layer thicknesses. Continuing our studies of the influences of various types of disorder on the electron and phonon structures of superlattices (Xiong 1985*a*, 1985*b*; Pang *et al.* 1985, 1986, 1987), we treat here the effect of this new factor of randomness on the spectrum of superlattice vibrations. Here we point out that the infinite order perturbation theory of Wu *et al.* (1974*a*, 1974*b*) modified to perform the computation in a self-consistent manner provides a satisfactory way to a solution of the problem. For this purpose, we adopt in this paper only a simplified dynamical model for the system, although it will not be difficult to generalise the discussion to more complicated situations (Xiong *et al.* 1982; Sun and Tsai 1982).

2. The Model

We consider the layered material to consist of alternating crystalline films A and B. We assume both A and B layers to have simple cubic structure with identical lattice constants, and with the interfaces between adjacent layers perpendicular to the (001) direction. Let the number of atomic planes contained in an A or B layer be p or q respectively, so that the following distributions are satisfied:

$$P(p) = \sum_i P_A(i)\,\delta_{p,i}, \qquad P(q) = \sum_i P_B(i)\,\delta_{q,i}, \tag{1a, b}$$

where $i = 1, 2, ..., r$, with r an integer. We consider p and q to be statistically uncorrelated.

We adopt for each layer a simple elastic model described by the potential function (Xiong *et al.* 1982)

$$\Phi = \Phi_0 + \tfrac{1}{2} \sum_{a,a'} K_{a,a'} \{ u(a) - u(a') \}^2 , \tag{2}$$

where Φ_0 is a constant, $u(a)$ the displacement of an atom at the site a, and $K_{a,a'}$ the force constants. We assume isotropy and homogeneity in each film, and include only nearest and next-to-nearest interactions with force constants K_1 and K_2 respectively.

In the mixed Bloch-site representation (Xiong *et al.* 1982), the matrix elements of the phonon Greenian satisfy

$$D(n, n'; k) = D_0(n, k) + \sum_{n''} D_0(n, k) \, T(n, n''; k) \, D(n'', n'; k), \tag{3}$$

where n, n', n'' label the atomic planes, k is a Bloch vector in a plane parallel to the interfaces, while

$$D_0(n, k) = M_A^{-1} \Gamma(k) - \omega^2, \qquad \text{for } n \text{ in an A layer}$$

$$= M_B^{-1} \Gamma(k) - \omega^2, \qquad \text{for } n \text{ in a B layer}, \tag{4}$$

$$T(n, n'; k) = M_A^{-1} \gamma(k), \qquad \text{for } n, n' \text{ in an A layer}$$

$$= M_B^{-1} \gamma(k), \qquad \text{for } n, n' \text{ in a B layer}, \tag{5}$$

with $M_{A(B)}$ the masses of the atoms in the A(B) layers, and

$$\Gamma(k) = 2K_1 \{ -3 + \cos(k_x d) + \cos(k_y d) \}$$

$$+ 4K_2 \{ -3 + \cos(k_x d) \cos(k_y d) \} , \tag{6}$$

$$\gamma(k) = K_1 + 2K_2 \{ \cos(k_x d) + \cos(k_y d) \} . \tag{7}$$

One also needs the matrix elements of T between atomic planes on both sides of an interface. Generally speaking, the elastic coupling between two unlike atoms A and B should be different from those between like atoms A and A, or B and B, but, for simplicity, we overlook such differences. In fact, we have assumed already in equations (4)–(7) that the interactions between a pair of A atoms and between a pair of B atoms are identical, so that only two force constants K_1 and K_2 are used to characterise the elastic couplings between any pair of nearest or next-to-nearest neighbouring atoms. Thus, the two sorts of atoms are distinguished only by their different masses, and

$$T(n, n'; k) = (M_A M_B)^{-\tfrac{1}{2}} \gamma(k) \tag{8}$$

for two adjacent atomic planes n, n' on both sides of an interface.

3. Method of Computation

Our aim is to evaluate the phonon density of states (DOS) given by

$$\rho(\omega^2) = -\pi^{-1} \sum_k \sum_n \{\text{Im}\langle D(n, n; k)\rangle\}, \tag{9}$$

where $\langle ... \rangle$ signifies a configurational average over the random distribution of layer thickness. Following Wu *et al.* (1974a, 1974b), $D(n, n)$ can be expressed as

$$D(n, n) = (1 - Z_{n, n+1} - Z_{n, n-1})^{-1} D_0(n), \tag{10}$$

where $Z_{n, n\pm 1}$ satisfy

$$D_0^{-1}(n) Z_{n, n\pm 1} = T(n, n\pm 1) T(n\pm 1, n)\{D_0^{-1}(n\pm 1) - D_0^{-1}(n\pm 1) Z_{n\pm 1, n\pm 2}\}^{-1}. \tag{11}$$

Hereafter, the parameter k, which is a good quantum number, is suppressed to make the equations resemble those for a linear chain.

Equation (11) gives $Z_{n, n\pm 1}$ as infinite continued fractions, so that approximations are unavoidable for their evaluation. Let us consider a certain arbitrary finite segment of the chain. We use two 'mean field' parameters T_A and T_B to describe the influences on the segment by the rest of the infinite chain connected to its two ends. For any specified distributions $P(p)$ and $P(q)$, equations can be written for T_A and T_B, so that they can be computed self-consistently. With these, the $D(n, n)$ and its configurational average are then calculated exactly for the segment.

The probability for the occurrence of an A-layer of p_1 planes of A atoms, a B-layer of q_1 planes of B atoms, a second A-layer of p_2 planes, a second B-layer of q_2 planes, ..., an sth A-layer of p_s planes and an sth B-layer of q_s planes, in succession, is $P_A(p_1) P_B(q_1) P_A(p_2) P_B(q_2) ... P_A(p_s) P_B(q_s)$. Writing T_B and T_A for $D_0^{-1}(1) Z_{1,0}$ at the first atomic plane of the first A-layer and $D_0^{-1}(m) Z_{m, m+1}$ at the last (the q_sth) atomic plane of the sth B-layer respectively, where $m = p_1 + q_1 + p_2 + q_2 + ... + p_s + q_s$, the configurational average is then

$$\langle D(n, n)\rangle = \sum_{p_1} \sum_{q_1} \sum_{p_2} \sum_{q_2} ... \sum_{p_s} \sum_{q_s} \left(\prod_{i=1}^{s} P_A(p_i) P_B(q_i) m^{-1} \sum_{n=1}^{m} D(n, n)\right), \tag{12}$$

with $D(n, n)$ given by (10) and (11). The $Z_{n, n\pm 1}$ are ultimately expressed in terms of $T(n, n\pm 1)$ and T_B and T_A, the latter parameters being defined by

$$T_A = \sum_{p_1} \sum_{q_1} \sum_{p_2} \sum_{q_2} ... \sum_{p_s} \sum_{q_s} \prod_{i=1}^{s} P_A(p_i) P_B(q_i) F_A(p_1, q_1, ..., p_s, q_s; T_A), \tag{13}$$

$$T_B = \sum_{p_1} \sum_{q_1} \sum_{p_2} \sum_{q_2} ... \sum_{p_s} \sum_{q_s} \prod_{i=1}^{s} P_A(p_i) P_B(q_i) F_B(p_1, q_1, ..., p_s, q_s; T_B), \tag{14}$$
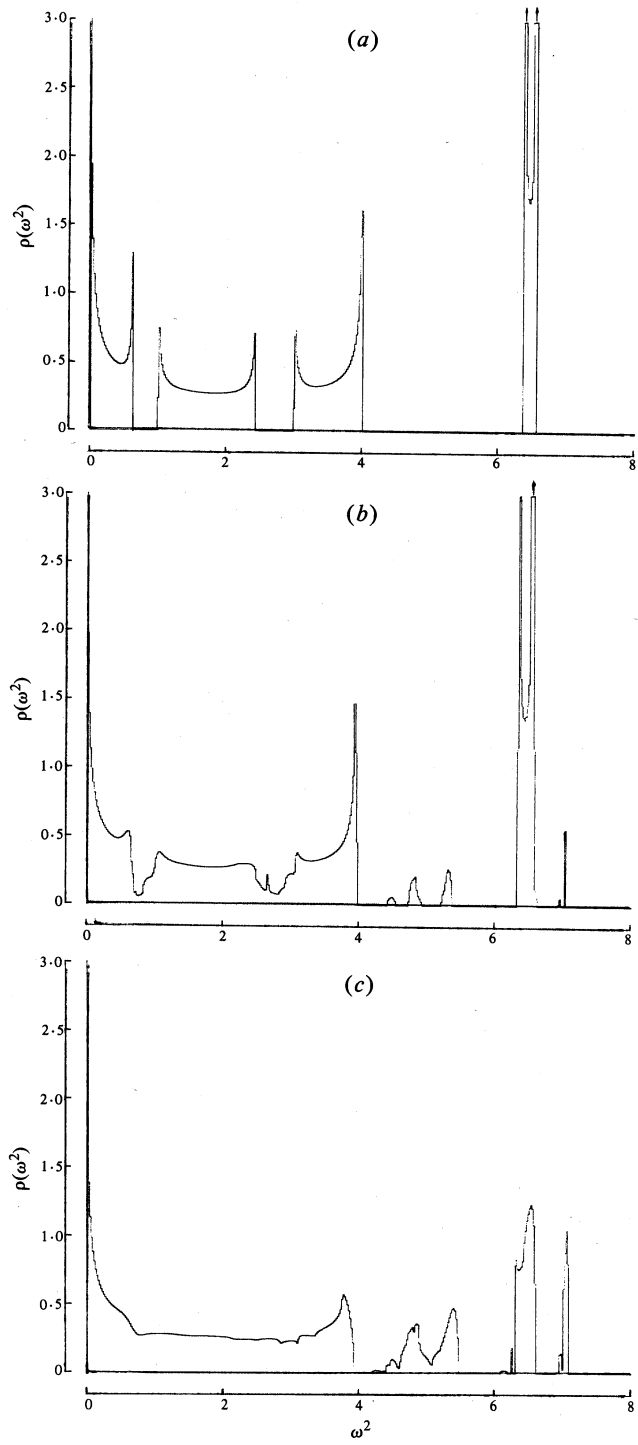
Fig. 1. Phonon DOS at the centre of the two-dimensional Brillouin zone for different degrees of randomness in layer thickness: (*a*) perfect superlattice ($p = 0$); (*b*) $p = 0.05$; and (*c*) $p = 0.20$.
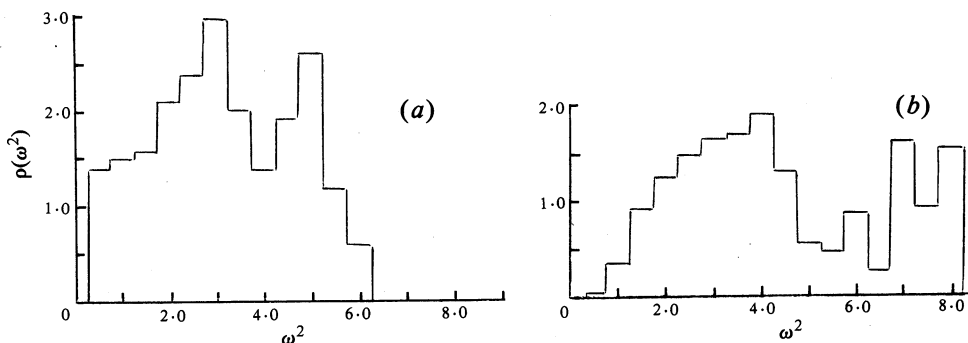
Fig. 2. Total phonon DOS for different degrees of randomness in layer thickness: (a) perfect superlattice ($p = 0$) and (b) $p = 0 \cdot 20$.

where

$$F_A(p_i, q_i; T_A) = D_0^{-1}(0) \, Z_{0,1} = \cfrac{|T(0,1)|^2}{D_0^{-1}(1) - \cfrac{|T(1,2)|^2}{D_0^{-1}(2) - \cdot \atop \qquad \cdot \cfrac{|T(m-1, m)|^2}{D_0^{-1}(m) - T_A}}}, \qquad (15)$$

$$F_B(p_i, q_i; T_B) = D_0^{-1}(m+1) \, Z_{m+1, m} = \cfrac{|T(m+1, m)|^2}{D_0^{-1}(m) - \cfrac{|T(m, m-1)|^2}{D_0^{-1}(m-1) - \cdot \atop \qquad \cdot \cfrac{|T(1,0)|^2}{D_0^{-1}(1) - T_B}}} . \qquad (16)$$

4. Numerical Example

Equations (9)–(16) constitute our working scheme. The more layers (larger s) included in the computation, the better the approximation. As an illustration, we take

$$M_A^{-1} K_1 = \tfrac{2}{3}, \quad M_A^{-1} K_2 = \tfrac{1}{3}, \quad M_B = 2M_A, \quad s = 1, \qquad (17)$$

$$P_A(i) = P_B(i) = p, \qquad i = 1, 3$$

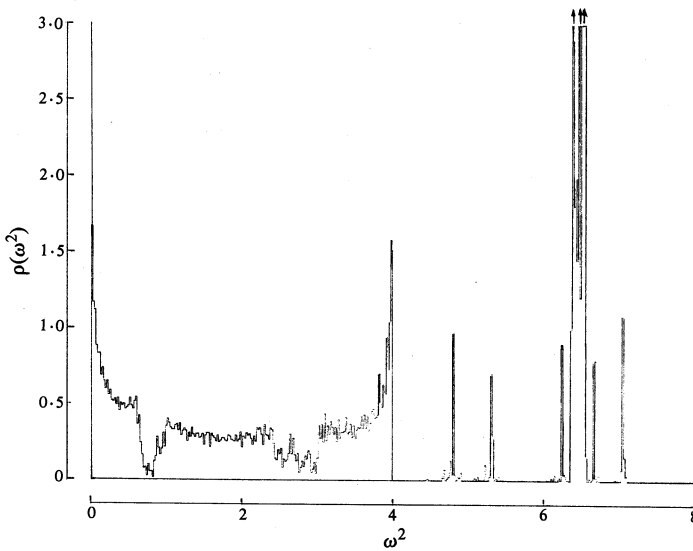$$= 1 - 2p, \quad i = 2 \quad (r = 3). \qquad (18)$$

Fig. 3. Phonon density of states at the centre of the two-dimensional Brillouin zone by Monte Carlo simulation for $p = 0.05$.

Fig. 1 displays the phonon DOS at the centre of the two-dimensional Brillouin zone $k = 0$, with (a) $p = 0$, (b) $p = 0.05$, and (c) $p = 0.20$, while Fig. 2 shows the total DOS with (a) $p = 0$ and (b) $p = 0.20$. In Figs 1a and 2a, the curves correspond to the case of a perfect superlattice.

As a check, we have performed a Monte Carlo simulation for $p = 0.05$ with randomly generated samples consisting of 600 atoms. The number of eigenfrequencies between ω and $\omega + \Delta\omega$ ($\Delta\omega = 0.01$) has been computed and averaged over the samples; the result is shown in Fig. 3. (We found that 600 atoms are sufficient to ensure the stability of the results.) The good agreement between Figs 1b and 3 clearly illustrates the merit of the theory, even though s only has the value 1.

Figs 1 and 2 exhibit the effects of randomness in layer thickness on the phonon structure. With increasing disorder, new modes appear within the forbidden bands, while the DOS profile smooths down with more and more fine structure wiped out and the whole frequency range broadened.

Acknowledgments

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