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A. Rodriguez, A. Noguera, T. Szwacka, J. Mendiàldua, Leonard Dobrzynski. Theory of phonons in three- and four-layer superlattices. *Physical Review B*, 1989, 39 (17), pp.12568-12574. 10.1103/PhysRevB.39.12568 . hal-04070519

HAL Id: hal-04070519

<https://hal.science/hal-04070519>

Submitted on 15 Apr 2023

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Theory of phonons in three- and four-layer superlattices

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(Received 7 December 1988; revised manuscript received 7 February 1989)

A theory of bulk and surface phonons in N -layer superlattices is presented. As an application we give, for the first time, closed-form expressions for surface phonons in three-layer superlattices and bulk phonons for four-layer superlattices, together with a few phonon dispersion curves.

I. INTRODUCTION

Phonons in two-layer superlattices have been studied theoretically and experimentally by Raman and Brillouin spectroscopies for several years. The aim of this paper is not to review the existing literature,¹ but rather to address phonons in N -layer superlattices. N -layer superlattices are formed out of a periodic repetition of a unit cell containing N ($N > 2$) different slabs. There was progress recently in producing such materials² and in studying them experimentally by Raman spectroscopy.³ We hope that the present theoretical paper will stimulate further experimental investigation of phonons in N -layer superlattices. In Sec. II, we first present briefly a simple three-dimensional phonon model and then give the surface-response operators necessary for a theoretical investigation of N -layer superlattice phonons. We show in Sec. III how the interface response theory⁴ enables us to calculate the response function and the bulk superlattice phonons. In Sec. IV, we give closed-form expressions for bulk phonons in three-layer and four-layer superlattices and surface phonons in three-layer superlattices.

II. BULK-PHONON MODEL AND THE SURFACE-RESPONSE OPERATORS

The potential energy ϕ_i associated with the lattice vibrations of an infinite simple-cubic lattice of type i (Ref. 5) considered here has the form

$$\phi_i = \frac{1}{2} \beta_i \sum_l \sum_p \sum_\alpha [u_\alpha(l) - u_\alpha(l+p)]^2, \quad (1)$$

where l ranges over all sites of the crystal, p ranges over the six nearest sites of the atom at l , and $u_\alpha(l)$ with $\alpha = 1, 2, 3$ denotes the component of the displacement of the atom at site l . This model is not rotationally invariant and does not give rise to Rayleigh surface waves on a (001) surface. Nevertheless, these deficiencies are unimportant for the qualitative study of many physical properties of surfaces⁶ and superlattices^{7,8} and, in particular, for the study of the transverse polarized phonons we will consider here. Let us recall also that most of the experi-

mental results for phonons in two-layered superlattices are still interpreted^{1,3} with the help of even simpler models (linear chain or transverse elastic vibrations). So the results of the present paper can be considered realistic for phonons polarized perpendicularly to the sagittal plane and do not address the phonons polarized within the sagittal plane nor Rayleigh waves polarized within this plane.

The bulk vibrational properties of the above crystal can be studied with the help of its bulk-response function \vec{G}_{0i} defined by

$$\vec{H}_{0i} \vec{G}_{0i} = \vec{I}, \quad (2)$$

where \vec{I} stands for the unit matrix and \vec{H}_{0i} for the bulk dynamical matrix. Taking advantage of the periodicity of the system in directions parallel to the (001) plane, we introduce the following two-dimensional vectors:

$$\begin{aligned} \mathbf{x}_\parallel(l) &= a_0(l_1 \hat{\mathbf{x}}_1 + l_2 \hat{\mathbf{x}}_2), \\ \mathbf{k}_\parallel(l) &= k_1 \hat{\mathbf{x}}_1 + k_2 \hat{\mathbf{x}}_2, \end{aligned} \quad (3)$$

(where a_0 is the lattice parameter and $\hat{\mathbf{x}}_1, \hat{\mathbf{x}}_2$ are unit vectors), and a Fourier transformation of the response function,

$$\begin{aligned} G_{0i}(l, l'; \omega^2) &= \frac{1}{N^2} \sum_{\mathbf{k}_\parallel} G_{0i}(l_3, l'_3; \mathbf{k}_\parallel \omega^2) \\ &\times \exp\{i \mathbf{k}_\parallel \cdot [\mathbf{x}_\parallel(l) - \mathbf{x}_\parallel(l')]\}, \end{aligned} \quad (4)$$

where N^2 is the number of atoms in a (001) plane.

The corresponding bulk-response function is⁶

$$G_{0i}(l_3, l'_3; \mathbf{k}_\parallel \omega^2) = \frac{M_i}{\beta_i} \frac{t_i^{|l_3 - l'_3| + 1}}{t_i^2 - 1}, \quad (5)$$

where M_i is the mass of the atoms.

Let us now establish a slab by removing in the infinite crystal all interactions between the atoms situated in the $l_3 = 0$ and $l_3 = 1$ planes and also between those situated in the $l_3 = L_i$ and $l_3 = L_i + 1$. The corresponding cleavage operator is^{7,8}

$$\begin{aligned}
V_{0i}(l_3, l'_3) = & \frac{\beta_i}{M_i} (\delta_{l_3,0} \delta_{l'_3,0} + \delta_{l_3,1} \delta_{l'_3,1} \\
& - \delta_{l_3,0} \delta_{l'_3,1} - \delta_{l_3,1} \delta_{l'_3,0}) \\
& + \frac{\beta_i}{M_i} (\delta_{l_3,L_i} \delta_{l'_3,L_i} + \delta_{l_3,L_i+1} \delta_{l'_3,L_i+1} \\
& - \delta_{l_3,L_i} \delta_{l'_3,L_i+1} - \delta_{l_3,L_i+1} \delta_{l'_3,L_i}). \quad (6)
\end{aligned}$$

The elements of the surface-response operator \vec{A}'_{si} formed⁸ out of the elements within the slab of \vec{A}_{0i} = $\vec{G}_{0i} \vec{V}_{0i}$ are

$$\begin{aligned}
A'_{si}(l_3 l'_3) = & -\frac{1}{t_i + 1} (\delta_{l'_3,1} t_i^{l_3} + \delta_{l'_3,L_i} t_i^{L_i - l_3 + 1}) \\
& \text{with } 1 \leq l_3, l'_3 \leq L_i. \quad (7)
\end{aligned}$$

III. THE RESPONSE FUNCTION AND THE N -LAYER SUPERLATTICE PHONONS

Define for the N -layer superlattice a reference response function \vec{G} as a block-diagonal matrix formed out of only the elements of the bulk-response functions \vec{G}_{0i} contained within the space of definition of each slab, namely, with the notation

$$m \equiv nN + i \equiv (n, i), \quad (8)$$

where n labels the unit cell formed out of N different slabs, $1 \leq i \leq N$ the position of the i th slab in this unit cell, and l_3 the atomic (001) plane within the i th slab:

$$\begin{aligned}
G(m, l_3; m', l'_3) = & \delta_{mm'} G_m(l_3, l'_3) \\
& \text{with } 1 \leq l_3, l'_3 \leq L_m, \quad (9)
\end{aligned}$$

where $\delta_{mm'}$ is the usual Kronecker symbol.

The response function \vec{g} of the N -layer superlattice can be calculated directly from the reference response function \vec{G} through the universal relation⁴

$$(\vec{I} + \vec{A}') \vec{g} = \vec{G}, \quad (10)$$

where the interface operator \vec{A}' is defined⁴ by

$$\vec{A}' = \vec{A}'_s + \vec{G} \vec{V}_I. \quad (11)$$

\vec{A}' is the block-diagonal surface-response operator formed out of the $A'_s(m, l_3; m', l'_3)$ given by

$$\vec{A}'_s(m, l_3; m', l'_3) = \delta_{mm'} A'_{sm}(l_3, l'_3) \quad (12)$$

and \vec{V}_I is the coupling operator

$$\begin{aligned}
V_I(m, l_3; m', l'_3) = & \delta_{mm'} (\delta_{l_3, L_m} \delta_{l'_3, L_m} \beta_{m, m+1} \\
& + \delta_{l_3, 1} \delta_{l'_3, 1} \beta_{m-1, m}) \\
& - \delta_{m+1, m'} \delta_{l_3, L_m} \delta_{l'_3, 1} \beta_{m, m+1} \\
& - \delta_{m, m'-1} \delta_{l_3, 1} \delta_{l'_3, L_{m-1}} \beta_{m, m-1}, \quad (13a)
\end{aligned}$$

where the force constants $\beta_{m, m'}$ coupling the interface atoms are taken as mean values between the bulk force constants of the adjacent layers

$$\beta_{m, m'} = \frac{\beta_m + \beta_{m'}}{2}. \quad (13b)$$

The general solution of Eq. (10) for any model of an N -layer superlattice was given before.⁹ For the present model, the elements $g(m, l_3; m', l'_3)$ are scalars and can be calculated in closed form with the help of the following (2×2) matrices:

$$\vec{K}(m) = \begin{bmatrix} A'(m, 1; m, L_m) & A'(m, 1; m+1, 1) \\ 1 + A'(m, L_m; m, L_m) & A'(m, L_m; m+1, 1) \end{bmatrix}, \quad (14)$$

$$\vec{H}(m) = \begin{bmatrix} A'(m, 1; m-1, L_{m-1}) & 1 + A'(m, 1; m, 1) \\ A'(m, L_m; m-1, L_{m-1}) & A'(m, L_m; m, 1) \end{bmatrix}, \quad (15)$$

$$\vec{P}(m) = -\vec{K}^{-1}(m) \vec{H}(m), \quad (16)$$

and

$$\vec{R} \equiv \vec{R}(N, 0) = \vec{P}(N) \vec{P}(N-1) \cdots \vec{P}(1). \quad (17)$$

When calculating $\vec{P}(m)$ it is helpful to define

$$t_m = e^{q_m}, \quad (18)$$

$$A_m = \frac{1}{\beta_m} \frac{\sinh[(L_m - 1)q_m]}{\sinh(q_m)}, \quad (19)$$

$$B_m = \frac{\cosh[(L_m - \frac{1}{2})q_m]}{\cosh(q_m/2)}, \quad (20)$$

and

$$C_m = 2\beta_m \tanh(q_m/2) \sinh(q_m L_m). \quad (21)$$

The $\vec{P}(m)$ is, for the present phonon model, given by

$$\vec{P}(m) = \begin{bmatrix} -\beta_{m-1, m} A_m & \beta_{m-1, m} A_m + B_m \\ -\beta_{m-1, m} A_m - \frac{\beta_{m-1, m}}{\beta_{m, m+1}} B_m & \beta_{m-1, m} A_m + \left[1 + \frac{\beta_{m-1, m}}{\beta_{m, m+1}} \right] B_m + \frac{1}{\beta_{m, m+1}} C_m \end{bmatrix}. \quad (22)$$

With the help of these expressions, Eq. (10) can be written¹⁰ as

$$\vec{\mathbf{K}}(m) \begin{bmatrix} g(m, L_m; m', l') \\ g(m+1, 1; m', l') \end{bmatrix} = -\vec{\mathbf{H}}(m) \begin{bmatrix} g(m-1, L_{m-1}; m', l') \\ g(m, 1; m', l') \end{bmatrix} + \delta_{mm'} \begin{bmatrix} G_m(1, l') \\ G_m(L_m, l') \end{bmatrix}. \quad (23)$$

Using N times this equation, one obtains

$$\begin{bmatrix} g(n, N, L_N; n', i', l') \\ g(n+1, 1, 1; n', i', l') \end{bmatrix} = \vec{\mathbf{R}}(N, 0) \begin{bmatrix} g(n-1, N, L_N; n', i', l') \\ g(n, 1, 1; n', i', l') \end{bmatrix} + \delta_{nn'} \vec{\mathbf{R}}(N, i') \vec{\mathbf{K}}^{-1}(i') \begin{bmatrix} G_{i'}(1, l'_3) \\ G_{i'}(L_{i'}, l'_3) \end{bmatrix}. \quad (24)$$

The dispersion relation for the eigenvalues of the N -layer superlattices is shown to be^{9,10}

$$2 \cos(k'_3 a_u) = \text{Tr} \vec{\mathbf{R}}(N, 0), \quad (25a)$$

where

$$a_u = a_0 \sum_{i=1}^N L_i \quad (25b)$$

is the width of the unit cell of the superlattice and k'_3 is the component of the propagation vector perpendicular to the interfaces.

From Eq. (24) one easily obtains

$$\begin{bmatrix} g(n, N, L_N; n', i', l'_3) + g(n-2, N, L_N; n', i', l'_3) \\ g(n+1, 1, 1; n', i', l'_3) + g(n-1, 1, 1; n', i', l'_3) \end{bmatrix} = (R_{11} + R_{22}) \vec{\mathbf{I}} \begin{bmatrix} g(n-1, N, L_N; n', i', l'_3) \\ g(n, 1, 1; n', i', l'_3) \end{bmatrix} + \delta_{nn'} \vec{\mathbf{B}} - \delta_{n-1, n} \vec{\mathbf{R}}^{-1} \vec{\mathbf{B}}, \quad (26)$$

where

$$\vec{\mathbf{B}} = \vec{\mathbf{R}}(N, i') \vec{\mathbf{K}}^{-1}(i') \begin{bmatrix} G_{i'}(1, l'_3) \\ G_{i'}(L_{i'}, l'_3) \end{bmatrix}. \quad (27)$$

The general solution of Eq. (26) is^{10,11}

$$\begin{bmatrix} g(n-1, N, L_N; n', i', l'_3) \\ g(n, 1, 1; n', i', l'_3) \end{bmatrix} = \frac{t}{t^2-1} (\vec{\mathbf{I}} t^{|n-n'|} - \vec{\mathbf{R}}^{-1} t^{|n-n'-1|}) \vec{\mathbf{B}}, \quad (28)$$

with

$$t = \begin{cases} \eta - (\eta^2 - 1)^{1/2}, & \eta > 1 \\ \eta + i(1 - \eta^2)^{1/2}, & -1 < \eta < 1 \\ \eta + (\eta^2 - 1)^{1/2}, & \eta < -1 \end{cases} \quad (29)$$

and

$$2\eta = R_{11} + R_{22}. \quad (30)$$

With the help of the definitions (14) and (17), and using Eq. (23), one also obtains¹⁰

$$\begin{bmatrix} g(n, i, L_i; n', i', l'_3) \\ g(n, i+1, 1; n', i', l'_3) \end{bmatrix} = \left[\frac{t}{t^2-1} [\vec{\mathbf{R}}(i, 0) \vec{\mathbf{R}}(N, i') t^{|n-n'|} - \vec{\mathbf{R}}(i, 0) \vec{\mathbf{R}}^{-1}(i', 0) t^{|n-n'-1|}] + \delta_{nn'} \vec{\mathbf{R}}(i, i') \right] \times \vec{\mathbf{K}}^{-1}(i') \begin{bmatrix} G_i(1, l'_3) \\ G_i(L_i, l'_3) \end{bmatrix}. \quad (31)$$

IV. THREE- AND FOUR-LAYER SUPERLATTICES

A. Bulk dispersion relations

The dispersion relation for the three-layer superlattice was presented before.⁸ We recall it here, as we will need it for the study of surface phonons in three-layer superlattices,

$$2 \cos(k'_3 a_u) = 2B_1 B_2 B_3 + \frac{C_1 C_2 C_3}{\beta_{12} \beta_{23} \beta_{31}} + |\varepsilon_{ijk}| \left[B_i A_j C_k + \frac{1}{2} B_i B_j C_k \left(\frac{1}{\beta_{12}} + \frac{1}{\beta_{23}} + \frac{1}{\beta_{31}} \right) \right. \\ \left. + \frac{1}{2\beta_{jk}} A_i C_j C_k + \frac{1}{2\beta_{jk}} B_i C_j C_k \left(\frac{1}{\beta_{ki}} + \frac{1}{\beta_{ij}} \right) \right], \quad (32)$$

where

$$a_u = a_0 \sum_{i=1}^3 L_i, \quad (33)$$

and bearing in mind the Einstein summation rule (over repeated indices) for tensors and the Levi-Civita completely antisymmetric tensor of rank 3,

$$\varepsilon_{ijk} = \begin{cases} 1 \\ -1 \\ 0 \end{cases} \text{ if } i, j, k \text{ is an } \begin{cases} \text{even} \\ \text{odd} \\ \text{otherwise} \end{cases} \text{ permutation of } i, j, k = 1, 2, 3. \quad (34)$$

Equation (32) can be solved easily numerically. In Fig. 1 we present the results for the band structure of a superlattice with two atomic planes in each layer. We assume that the three crystals differ only by their atomic masses, while the force constants are identical ($\beta_1 = \beta_2 = \beta_3$). The bulk phonons of such a superlattice are inside the shaded areas. Between these shaded areas are gaps in which are represented surface phonons. We will discuss them in Sec. IV B.

We have also calculated the dispersion relation for a four-layer superlattice and it is given by

$$2 \cos(k'_3 a_u) = 2B_1 B_2 B_3 B_4 + \frac{C_1 C_2 C_3 C_4}{\beta_{12} \beta_{23} \beta_{34} \beta_{14}} + C_1 C_3 A_2 A_4 + A_1 A_3 C_2 C_4 \\ + \sum_{\substack{i \neq j \neq k \neq l \\ i < j}} B_i B_j A_k C_l + \sum_{\substack{i \neq j \neq k \neq l \\ i < j < k}} \left[C_i C_j C_k C_l \left(\frac{1}{\beta_{ij} \beta_{jk} \beta_{kl}} + \frac{1}{\beta_{li} \beta_{ij} \beta_{jk}} \right) + C_i C_j C_k C_l \frac{1}{\beta_{l+1, l+2} \beta_{l+2, l+3}} \right. \\ \left. + B_i B_j B_k C_l \left(\frac{1}{\beta_{12}} + \frac{1}{\beta_{23}} + \frac{1}{\beta_{34}} + \frac{1}{\beta_{14}} \right) \right] \\ + \sum_{\substack{i \neq j \neq k \neq l \\ i < j \\ k < l}} B_i B_j C_k C_l \left\{ \left(\frac{1}{\beta_{ik}} + \frac{1}{\beta_{il}} \right) \left(\frac{1}{\beta_{jl}} + \frac{1}{\beta_{jk}} \right) \delta_{i+1, j-1} \right. \\ \left. + \frac{1}{\beta_{kl}} \left[\frac{1}{\beta_{ij}} + \frac{\delta_{l+1, i} + \delta_{i+1, l}}{\beta_{li}} + \frac{\delta_{j+1, l} + \delta_{j, l+1}}{\beta_{jl}} \right. \right. \\ \left. \left. + \frac{\delta_{j+1, k} + \delta_{k+1, j}}{\beta_{kj}} + \frac{\delta_{k+1, i} + \delta_{i+1, k}}{\beta_{ki}} \right] (\delta_{i+1, j} + \delta_{i, j+1}) \right\} \\ + \sum_{\substack{i \neq j \neq k \neq l \\ i < j}} C_i C_j A_k B_l \left[\frac{\delta_{i+1, j}}{\beta_{ij}} + \left(\frac{1}{\beta_{lj}} + \frac{1}{\beta_{li}} \right) \delta_{k+1, l-1} \right], \quad (35)$$

where $a_u = a_0 \sum_{i=1}^4 L_i$. In Figs. 2 and 3 we present the results for the band structure of a superlattice with two atomic planes in each film. We first assume (Fig. 2) that the four materials differ only by their masses, while the force constants are identical. We next assume (Fig. 3) that all the atoms have the same mass, but the force constants are different and at the interfaces these constants are taken to be as defined by Eq. (13b).

The long-wavelength limit of these bulk phonons for three- and four-layer superlattices can be compared with

the transverse polarized vibrations in such superlattices reported and discussed before.¹¹ The results presented here give, in fact, the departure from linear elasticity of the transverse polarized vibrations of three- and four-layer superlattices.¹¹

B. Surface phonons for a three-layer superlattice

In order to obtain a semi-infinite three-layer superlattice with a free surface, we cleave the infinite superlattice

by removing all interactions between two adjacent slabs. This cleavage procedure enables us to calculate the response function \vec{g}_s of the semi-infinite superlattice from the knowledge of the response function \vec{g} of the infinite superlattice. For the purpose of studying surface-localized modes, we need only to calculate one element of \vec{g} . For a semi-infinite three-layer superlattice having a slab of type 1 at the surface and below slab 2 and then slab 3, and so on, one finds, with the help of Eq. (31),

$$g_s(1, 1, 1; 1, 1, 1) = -\frac{A_s(\omega^2) - t}{D(\omega^2)}, \quad (36)$$

where

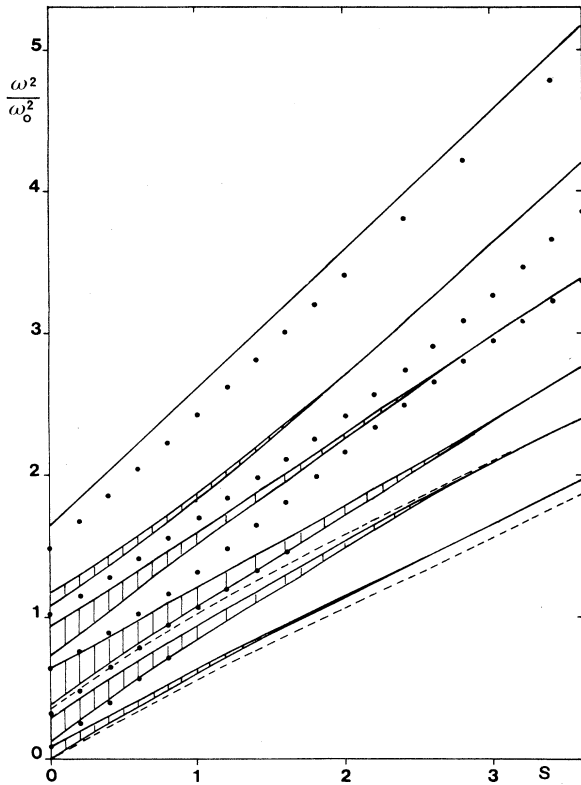


FIG. 1. Phonon band structure ω^2/ω_0^2 with $\omega_0^2 = 2\beta_1/M_1$ of a three-layer superlattice (with two atomic planes in each layer) as a function of the parameter $S = 2 - \cos k_1 a_0 - \cos k_2 a_0$. The three crystals differ by their atomic masses ($M_2 = 1.5M_1$ and $M_3 = 2M_1$), while the force constants are identical everywhere ($\beta_1 = \beta_2 = \beta_3 = \beta_{12} = \beta_{23} = \beta_{31}$). The five shaded areas and the curve highest in frequency represent the bulk bands. In the gaps between these bulk bands are represented the surface-phonon dispersion curves. The four highest curves represented by the dotted lines are for a surface terminated by two planes of crystal 1 having below them two planes of crystal 2 and then two planes of crystal 3, and so on. The two lowest curves represented by the dashed lines are for a surface terminated by two planes of crystal 3 having below them two planes of crystal 1 and then two planes of crystal 2, and so on.

$$A_s(\omega^2) = B_1 B_2 B_3 + A_1 C_2 B_3 + A_1 B_2 C_3 + B_1 A_2 C_3 + \frac{1}{\beta_{23}} (A_1 C_2 C_3 + B_1 B_2 C_3) + \frac{1}{\beta_{12}} (B_1 C_2 B_3 + B_1 B_2 C_3) + \frac{B_1 C_2 C_3}{\beta_{12} \beta_{31}}, \quad (37)$$

$$D(\omega^2) = B_1 B_2 C_3 + B_1 C_2 B_3 + C_1 B_2 B_3 + C_1 A_2 C_3 + \frac{1}{\beta_{23}} (C_1 B_2 C_3 + B_1 C_2 C_3) + \frac{1}{\beta_{12}} (C_1 C_2 B_3 + C_1 B_2 C_3) + \frac{C_1 C_2 C_3}{\beta_{12} \beta_{23}}. \quad (38)$$

From the poles of this response function, we find the frequencies ω_s of the surface modes localized at the free surface ($n=1, i=1, l_3=1$) and decaying inside the bulk situated at $n > 1$ and $l_3 > 1$. They are given by

$$D(\omega_s^2) = 0 \quad (39)$$

with the following condition,

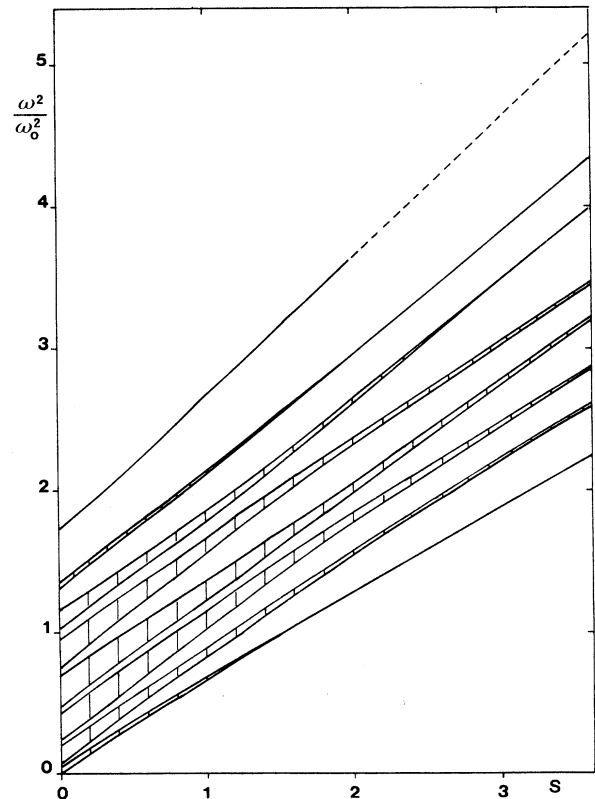


FIG. 2. Phonon band structure ω^2/ω_0^2 with $\omega_0^2 = 2\beta_1/M_1$ of a four-layer superlattice (with two atomic planes in each layer) as a function of the parameter $S = 2 - \cos k_1 a_0 - \cos k_2 a_0$. The four crystals differ by their atomic masses ($M_2 = 1.25M_1$, $M_3 = 1.5M_1$, and $M_4 = 1.75M_1$), while the force constants are identical everywhere.

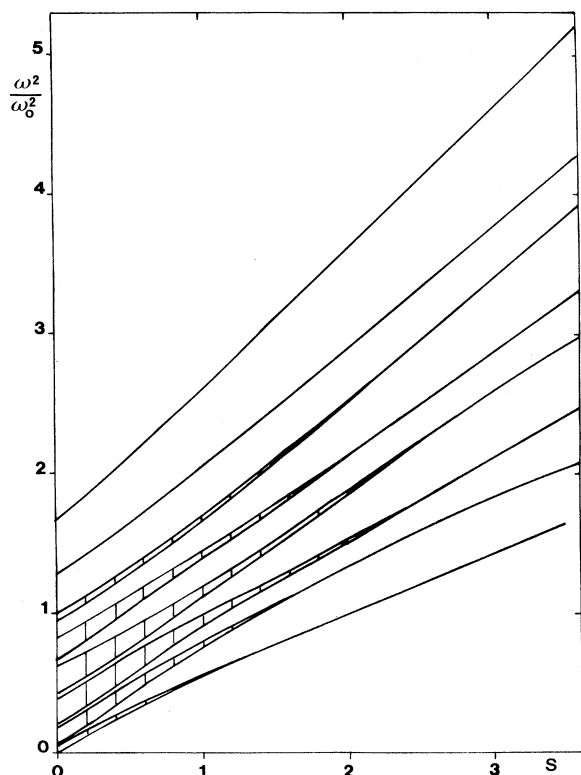


FIG. 3. Phonon band structure ω^2/ω_0^2 with $\omega_0^2=2\beta_1/M_1$ of a four-layer superlattice (with two atomic planes in each layer) as a function of the parameter $S=2-\cos k_1 a_0-\cos k_2 a_0$. The four crystals differ by their force constants [$\beta_1=2.5\beta_4$, $\beta_2=2.0\beta_4$, $\beta_3=1.5\beta_4$, and $\beta_{ij}=(\beta_i+\beta_j)/2$], while the masses are identical everywhere.

$$|A_s| > 1, \quad (40)$$

which ensures⁷ that these modes decay inside the bulk of the superlattice.

In order to illustrate this result, we investigated for the three-layer superlattice described in Sec. IV A and Fig. 1 the occurrence of surface phonons in the gaps between the bulk bands. We have first chosen the case for which the lighter slab 1 is at the surface. In that case, four branches of surface phonons were found and are

represented by dotted lines. Then we chose the case for which the heavier slab 3 is at the surface. For this case two branches of surface phonons appear and are represented by dashed lines.

The surface mode lying below the lowest bulk band is similar in the long-wavelength limit to Love waves in several respects. This surface wave exists when the outermost medium has the lower transverse sound velocity. In the large- k_{\parallel} limit the velocity of this mode approaches the transverse sound velocity in the outermost (slower) medium.

In contrast, the surface modes that lie between the gaps of the bulk modes may also exist even if the outermost medium has a higher transverse sound velocity.

The long-wavelength limit of these surface modes can also be compared to the transverse elastic waves reported before for two-layer¹² and three- and four-layer¹³ superlattices. The results presented here also provide the departure from linear elasticity of the surface transverse elastic waves reported before¹³ for three- and four-layer superlattices.

V. CONCLUSIONS

In this paper we obtained for the first time bulk phonons for a four-layer superlattice and surface phonons for a three-layer superlattice. These results were obtained with the help of a three-dimensional lattice model, which addresses only transversely polarized lattice vibrations in such superlattices. Although simple, the model is sufficient to illustrate the salient features. In more realistic models, the coupling between transverse and longitudinally polarized modes will have to be included, especially if one would like to study the Rayleigh waves on such materials.¹⁴ A complete list of all other possible improvements to the present work would be very lengthy and will have to be considered in relation with the forthcoming experimental results.³

ACKNOWLEDGMENTS

Three of us (A.R., J.M., and L.D.) would like to acknowledge the support of the Consejo Nacional de Investigaciones Científicas y Tecnológicas, Venezuela, and of the Centre National de la Recherche Scientifique, France, through their exchange program. The Laboratoire de Dynamique des Cristaux Moléculaires is Unité Associée au Centre National de la Recherche Scientifique No. 801.

¹See, for example, for a literature review, B. Djafari-Rouhani and J. Sapriel, Surf. Sci. Rep. (to be published); J. H. Worlock, *Proceedings of the Second International Conference on Phonon Physics*, edited by J. Kollar *et al.* (World Scientific, Singapore, 1985), p. 506; M. V. Klein, IEEE J. Quantum Electron. **QE-22**, 1970 (1986).

²L. Esaki, L. L. Chang, and E. E. Mendez, Jpn. J. Appl. Phys. **20**, L529 (1981).

³D. J. Lockwood, M. W. C. Dharma-wardana, J. M. Baribeau, and D. C. Houghton, Phys. Rev. B **35**, 2243 (1987); D. J.

Lockwood (private communication).

⁴L. Dobrzynski, Surf. Sci. Rep. **6**, 119 (1986).

⁵H. B. Rosenstock and G. F. Newell, J. Chem. Phys. **21**, 1607 (1953); E. N. Montroull and R. B. Potts, Phys. Rev. **102**, 72 (1956).

⁶See, for example, A. A. Maradudin, R. F. Wallis, and L. Dobrzynski, in *Handbook of Surfaces and Interfaces*, edited by L. Dobrzynski (Garland Scientific, Technical, Professional, and Medical, New York, 1980), Vol. 3, p. 327.

⁷L. Dobrzynski, B. Djafari-Rouhani, and O. Hardouin-Duparc,

- Phys. Rev. B **29**, 3138 (1984).
- ⁸T. Szwacka, A. Noguera, A. Rodriguez, J. Mendiola, and L. Dobrzynski, Phys. Rev. B **37**, 8451 (1988).
- ⁹L. Dobrzynski, Prog. Surf. Sci. **26**, 103 (1987).
- ¹⁰P. Masri and L. Dobrzynski, Surf. Sci. **198**, 285 (1988).
- ¹¹B. Djafari-Rouhani and L. Dobrzynski, Solid State Commun. **62**, 609 (1987).
- ¹²R. E. Camley, B. Djafari-Rouhani, L. Dobrzynski, and A. A. Maradudin, Phys. Rev. B **27**, 7318 (1983).
- ¹³J. Mendiola, T. Szwacka, A. Rodriguez, and L. Dobrzynski, Phys. Rev. B **39**, 10 674 (1989).
- ¹⁴B. Djafari-Rouhani, L. Dobrzynski, O. Hardouin Duparc, R. E. Camley, and A. A. Maradudin, Phys. Rev. B **28**, 1711 (1983).