Theory of surface phonons in superlattices

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We report the existence of surface-localized phonons for a superlattice consisting of alternating slabs (parallel to the surface) of two different crystals. The superlattice has a larger periodicity in the direction perpendicular to the slabs and therefore many phonon branches in the folded Brillouin zone. In the gaps existing between these phonon branches appear the surface localized modes. The theory is developed on a simple-cubic model. The simplicity of this model allows one to obtain in closed form the bulk and (001) surface Green's functions for this superlattice. The analytic knowledge of these functions enables us to study easily all the bulk and surface vibrational properties of a superlattice, which otherwise would require huge numerical calculations. We give here the analytic expression we obtained for the folded bulk phonon dispersion curves and also the expression that gives the surface-localized modes, which may appear within the extra gaps which exist between the folded bulk bands. A few figures for specific cases illustrate these results.

I. INTRODUCTION

A superlattice consists of alternating thin layers of two deposited compounds. Such systems have a new periodicity in the direction x_3 perpendicular to the layer. This produces a folding of the phonon dispersion curves in a reduced Brillouin zone and the opening of new gaps. The study of these bulk phonon properties of superlattices started recently with the help of linear chain models[1] treated numerically and also within the framework of the theory of elasticity.[2] Experimental investigations of GaAs-AlAs superlattices by Raman and infrared spectroscopy also appeared.[1,3]

Auld *et al.*[4] started the study of the transverse surface elastic vibrations by a purely numerical method. Analytical results for this problem were obtained recently by two different approaches: within the framework of elasticity theory[5,6] and as the elastic limit of a simple atomic model.[7] A short communication[7] about a few results due to this model was given at a recent conference. Here we will describe fully this atomic model and give all results obtained from it for the surface vibrations of a superlattice. The superlattice under study here is built up from alternately L_1 and L_2 (001) atomic planes of two different simple cubic lattices having the same lattice parameter a_0 and characterized by their atomic force constants (γ_1 and γ_2) and their atomic masses (M_1 and M_2). These alternating thin layers are bound together by a force constant γ between the interface atoms. This simple model enables one to obtain in closed form the bulk and (001) surface Green's function for this superlattice. The analytic knowledge of these functions enables us to study easily all the bulk and surface vibrational properties of a superlattice, which otherwise would require huge numerical calculations.

We will give here the analytic expressions we obtained for the Green's functions, for the folded bulk phonon dispersion curves and also for the surface-localized modes, which may appear within the extra gaps that exist between the folded bulk bands. These expressions enable us to discuss easily the effect of the physical parameters defined above. These surface phonons depend also on the kind of layer (1 or 2) being near the (001) surface. One simple limit of interest is the one where each layer can be treated as an elastic medium. This limit was studied also directly within the framework of elasticity theory[5,6] and the expressions found by a transfer-matrix method agree with the corresponding limit of our lattice dynamical results. Let us emphasize at this stage that both calculations apply only to the transverse vibration modes.

In Sec. II we obtain the bulk dynamical Green's function for the superlattice defined above. In Sec. III we give the corresponding surface Green's function. And in Secs. IV and V these results are used for the calculation of the bulk and surface phonons.

II. BULK DYNAMICAL GREEN'S FUNCTION FOR A SUPERLATTICE

In this section we obtain the bulk dynamical Green's function for a simple model[8,9] of a superlattice. We start from an infinite simple cubic lattice of atoms of mass M_1. Let $u_\alpha(l)$ denote the α component of the displacement of the atom at lattice site $\vec{x}(l)=a_0(l_1\hat{x}_1+l_2\hat{x}_2+l_3\hat{x}_3)$, where a_0 is the lattice parameter and $\hat{x}_1$, $\hat{x}_2$, and $\hat{x}_3$ unit vectors. The potential energy Φ associated with the lattice vibrations has the simple form

$$\Phi=\tfrac{1}{2}\gamma_1\sum_{l}\sum_{p}\sum_{\alpha}[u_\alpha(l)-u_\alpha(l+p)]^2\,, \tag{1}$$

where l ranges over all sites of the crystal, and p over the six nearest sites of the atom l.

This model is not rotationally invariant[10] 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 surfaces and interfaces[11] and in particular for the study of the transverse polarized surface modes we will consider here.

From the above form of the potential energy and by assuming a sinusoidal time dependence for the displacements, we obtain three uncoupled equations of motion, which we can write in the form

$$\overleftrightarrow{L}^{(B_1)}\cdot\vec{u}=\vec{0}\ , \tag{2}$$

where

$$L_{\alpha\beta}^{(B_1)}(l,l';\omega^2)=\delta_{\alpha\beta}\left[\left[\omega^2-6\frac{\gamma_1}{M_1}\right]\delta_{ll'}+\frac{\gamma_1}{M_1}\sum_p \delta_{l,l'+p}\right]. \tag{3}$$

We construct now out of this lattice a film of L_1 layers bounded by a pair of (001) surfaces. Each (001) atomic plane of this slab is labeled by

$$1\le l_3\le L_1\ . \tag{4}$$

The equations of motion of this film can be written as

$$\overleftrightarrow{L}^{(F_1)}\cdot\vec{u}=\vec{0} \tag{5}$$

and a Green's function $\overleftrightarrow{U}$ can be defined as

$$\overleftrightarrow{L}^{(F_1)}\cdot\overleftrightarrow{U}^{(F_1)}=\overleftrightarrow{I}\ . \tag{6}$$

$\overleftrightarrow{I}$ is a unit matrix with elements $\delta_{\alpha\beta}\delta_{ll'}$.

The advantage of this model is that this film Green's function $\overleftrightarrow{U}^{(F_1)}$ can be worked out in closed form,[12] once the corresponding surface Green's function is known.[13]

Taking advantage of the periodicity of the film in directions parallel to the surfaces, we introduce a two-dimensional position vector

$$\vec{x}_{||}(l)=a_0(l_1\hat{x}_1+l_2\hat{x}_2)\ , \tag{7}$$

a two-dimensional wave vector parallel to the surfaces

$$\vec{k}_{||}=k_1\hat{x}_1+k_2\hat{x}_2 \tag{8}$$

and

$$U_{\alpha\beta}^{(F_1)}(l,l';\omega^2)=\frac{\delta_{\alpha\beta}}{N^2}\sum_{\vec{k}_{||}}U_1(k_{||},\omega\,|\,l_3,l_3')\times e^{i\vec{k}_{||}\cdot[\vec{x}_{||}(l)-\vec{x}_{||}(l')]}\ , \tag{9}$$

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

The explicit expression of $U_1(k_{||},\omega\,|\,l_3,l_3')$ was worked out by Mazur and Maradudin[12] as a function of

$$\zeta_1=3-\cos(k_1a_0)-\cos(k_2a_0)-\frac{M_1}{2\gamma_1}\omega^2 \tag{10}$$

and

$$t_1=\begin{cases}\zeta_1-(\zeta_1^2-1)^{1/2}, & \zeta_1>1\\ \zeta_1+i(1-\zeta_1^2)^{1/2}, & -1<\zeta_1<1\\ \zeta_1+(\zeta_1^2-1)^{1/2}, & \zeta_1<-1\end{cases} \tag{11}$$

and is

$$U_1(\vec{k}_{||},\omega\,|\,l_3,l_3')=\frac{M_1}{\gamma_1}\frac{t_1^{|l_3-l_3'|+1}}{t_1^2-1}+\frac{M_1}{\gamma_1}\frac{t_1^{l_3+l_3'}}{t_1^2-1}+\frac{M_1}{\gamma_1}\frac{t_1}{t_1^2-1}\frac{t_1^{2L_1}}{1-t_1^{2L_1}}(t_1^{-l_3-l_3'+1}+t_1^{-l_3+l_3'}+t_1^{l_3-l_3'}+t_1^{l_3+l_3'-1})\ . \tag{12}$$

In the same manner we construct another film of L_2 (001) layers. In order to distinguish these two films one from the other, we will use an index $\kappa=1$ or 2. The corresponding Green's function $U_2(k_{||}\omega\,|\,l_3l_3')$ can be obtained from the above equations (10)—(12) by changing all indices 1 to 2. Let us also remark that for this $\kappa=2$ film, one has

$$1\le l_3\le L_2\ . \tag{13}$$

Let us now set this $\kappa=2$ film in epitaxy with the $\kappa=1$ film, but without any binding between the interface atom. We characterize this uncoupled double film by another integer n. An infinite repetition $-\infty<n<+\infty$ without coupling of this double film gives us our starting point for our model of a superlattice. One easily sees that the corresponding equations of motion and Green's function are

$$\overleftrightarrow{L}\cdot\vec{u}=\sum_{n=-\infty}^{+\infty}\sum_{\kappa=1}^{2}\overleftrightarrow{L}_n^{(F_\kappa)}\cdot\vec{u}(n\kappa)\ , \tag{14}$$

and

$$U(k_{||},\omega\,|\,n,\kappa,l_3;n',\kappa',l_3')=\delta_{nn'}\delta_{\kappa\kappa'}U_\kappa(\vec{k}_{||},\omega\,|\,l_3,l_3')\ , \tag{15}$$

such that

$$\overleftrightarrow{L}\cdot\overleftrightarrow{U}=\overleftrightarrow{I}\ . \tag{16}$$

Let us now couple all these alternating $\kappa=1$ and $\kappa=2$ films by a force interaction γ between the interface atoms facing each other. This creates the infinite superlattice we will study. Its equations of motion can be written as

$$(\overleftrightarrow{L}-\delta\overleftrightarrow{L})\cdot\vec{u}=\vec{0}\ , \tag{17}$$

where $\vec{u}$ stands for the vector whose elements are all the displacements $u_\alpha(n,\kappa,l_1,l_2,l_3)$ and

$$\delta L(n,\kappa,l_3;n',\kappa',l_3')=\gamma\sum_m\left[\frac{\delta_{nm}\delta_{\kappa 2}\delta_{l_3L_2}}{(M_2)^{1/2}}-\frac{\delta_{n,m+1}\delta_{\kappa 1}\delta_{l_3 1}}{(M_1)^{1/2}}\right]\left[\frac{\delta_{n'm}\delta_{\kappa' 2}\delta_{l_3'L_2}}{(M_2)^{1/2}}-\frac{\delta_{n',m+1}\delta_{\kappa' 1}\delta_{l_3' 1}}{(M_1)^{1/2}}\right]$$
$$+\gamma\sum_m\left[\frac{\delta_{nm}\delta_{\kappa 1}\delta_{l_3L_1}}{(M_1)^{1/2}}-\frac{\delta_{nm}\delta_{\kappa 2}\delta_{l_3 1}}{(M_2)^{1/2}}\right]\left[\frac{\delta_{n'm}\delta_{\kappa' 1}\delta_{l_3'L_1}}{(M_1)^{1/2}}-\frac{\delta_{n'm}\delta_{\kappa' 2}\delta_{l_3' 1}}{(M_2)^{1/2}}\right]. \quad (18)$$

We can now define a Green's function $\overleftrightarrow{D}$ for this infinite superlattice by

$$(\overleftrightarrow{L}-\delta\overleftrightarrow{L})\cdot\overleftrightarrow{D}=\overleftrightarrow{I} \quad (19)$$

and work out in closed form its elements $D(\vec{k}_{\parallel},\omega\mid n,\kappa,l_3;n',\kappa',l_3')$ with the help of the Dyson equation

$$\overleftrightarrow{D}=\overleftrightarrow{U}+\overleftrightarrow{U}\cdot\delta\overleftrightarrow{L}\cdot\overleftrightarrow{D}\,. \quad (20)$$

In what follows and for simplicity we will no longer write explicitly the dependence on $\vec{k}_{\parallel}$ and ω in the elements of $\overleftrightarrow{U}$ and $\overleftrightarrow{D}$.

After some algebra (see the Appendix for more details) we obtain the elements of $\overleftrightarrow{D}$ as functions of the q_κ defined in terms of the t_κ of Eq. (11) by

$$t_\kappa=e^{q_\kappa},\quad \kappa=1 \text{ or } 2 \quad (21)$$

and a new variable t defined by

$$t=\begin{cases}\eta-(\eta^2-1)^{1/2}, & \eta>1\\ \eta+i(1-\eta^2)^{1/2}, & -1<\eta<1\\ \eta+(\eta^2-1)^{1/2}, & \eta>-1\end{cases} \quad (22)$$

with

$$\eta=\frac{2\gamma_1\gamma_2}{\gamma^2}\tanh\left[\frac{q_1}{2}\right]\tanh\left[\frac{q_2}{2}\right]\sinh(q_1L_1)\sinh(q_2L_2)+\frac{2\gamma_1}{\gamma}\tanh\left[\frac{q_1}{2}\right]\sinh(q_1L_1)\frac{\cosh[q_2(L_2-\frac{1}{2})]}{\cosh(\frac{1}{2}q_2)}$$
$$+\frac{2\gamma_2}{\gamma}\tanh\left[\frac{q_2}{2}\right]\sinh(q_2L_2)\frac{\cosh[q_1(L_1-\frac{1}{2})]}{\cosh(\frac{1}{2}q_1)}+\frac{\gamma_2}{\gamma_1}\frac{\sinh[q_1(L_1-1)]}{\sinh q_1}\tanh\left[\frac{q_2}{2}\right]\sinh(q_2L_2)$$
$$+\frac{\gamma_1}{\gamma_2}\frac{\sinh[q_2(L_2-1)]}{\sinh q_2}\tanh\left[\frac{q_1}{2}\right]\sinh(q_1L_1)+\frac{\cosh[q_1(L_1-\frac{1}{2})]}{\cosh(\frac{1}{2}q_1)}\frac{\cosh[q_2(L_2-\frac{1}{2})]}{\cosh(\frac{1}{2}q_2)}\,. \quad (23)$$

Let us give the explicit expressions of the elements $D(k_{\parallel}\omega\mid n\kappa l_3,n'\kappa'l_3')$ of the superlattice Green's function $\overleftrightarrow{D}$.

The elements of $\overleftrightarrow{D}$ between different $\kappa=1$ and $\kappa=2$ films are

$$D(n,1,l_3;n',2,l_3')=\frac{t(M_1M_2)^{1/2}}{t^2-1}[K_{12}(l_3,l_3')t^{|n-n'|}+K_{12}(L_1-l_3+1,L_2-l_3'+1)t^{|n-n'-1|}]\,, \quad (24a)$$

where

$$K_{12}(l_3,l_3')=\frac{1}{\gamma}\frac{\cosh[q_1(l_3-\frac{1}{2})]}{\cosh(\frac{1}{2}q_1)}\frac{\cosh[q_2(L_2-l_3'+\frac{1}{2})]}{\cosh(\frac{1}{2}q_2)}+\frac{1}{\gamma_2}\frac{\cosh[q_1(l_3-\frac{1}{2})]}{\cosh(\frac{1}{2}q_1)}\frac{\sinh[q_2(L_2-l_3')]}{\sinh q_2}$$
$$+\frac{1}{\gamma_1}\frac{\cosh[q_2(L_2-l_3'+\frac{1}{2})]}{\cosh(\frac{1}{2}q_2)}\frac{\sinh[q_1(l_3-1)]}{\sinh q_1} \quad (24b)$$

and

$$D(n,2,l_3;n',1,l_3')=D(n,1,l_3';n',2,l_3)\,. \quad (24c)$$

The elements of $\overleftrightarrow{D}$ between the same κ films are

$$D(n,1,l_3;n',1,l_3')=\frac{M_1}{\gamma_1}t^{|n-n'|}\frac{\sinh[q_1(l_3-l_3')]}{\sinh q_1}\text{sgn}[L_1(n-n')+l_3-l_3']+\frac{M_1}{\gamma_1}\frac{t^{|n-n'|+1}}{t^2-1}K_{11}(l_3,l_3')\,, \quad (25a)$$

where

$$K_{11}(l_3,l_3')=\frac{1}{4\cosh^2(\frac{1}{2}q_1)}\{\cosh[q_1(L_1+l_3-l_3')]+\cosh[q_1(L_1-l_3+l_3')]+2\cosh[q_1(L_1-l_3-l_3'+1)]\}\ ,$$

$$\times\left[\frac{2\gamma_1\gamma_2}{\gamma^2}\tanh\left[\frac{q_2}{2}\right]\sinh(q_2L_2)+\frac{2\gamma_1}{\gamma}\frac{\cosh[q_2(L_2-\frac{1}{2})]}{\cosh(\frac{1}{2}q_2)}+\frac{\gamma_1}{\gamma_2}\frac{\sinh[q_2(L_2-1)]}{\sinh q_2}\right]$$

$$+\frac{1}{2\sinh q_1}\left\{\frac{\gamma_2}{\gamma_1}\tanh\left[\frac{q_2}{2}\right]\frac{\sinh(q_2L_2)}{\sinh q_1}\{\cosh[q_1(L_1+l_3-l_3'-1)]+\cosh[q_1(L_1+l_3'-l_3-1)]\right.$$

$$-2\cosh[q_1(L_1-l_3-l_3'+1)]\}$$

$$+\frac{2}{\cosh(\frac{1}{2}q_1)\cosh(\frac{1}{2}q_2)}\left[\sinh[q_1(L_1-\tfrac{1}{2})]\cosh[q_1(l_3-l_3')]\right.$$

$$\left.-\sinh\left[\frac{q_1}{2}\right]\cosh[q_1(L_1-l_3-l_3'+1)]\right]$$

$$\left.\times\left[\frac{2\gamma_2}{\gamma}\sinh\left[\frac{q_2}{2}\right]\sinh(q_2L_2)+\cosh[q_2(L_2-\tfrac{1}{2})]\right]\right\}\ . \tag{25b}$$

Thus $D(n,2,l_3;n',2,l_3')$ can be obtained from $D(n,1,l_3;n',1,l_3')$ by interchanging in γ_κ, M_κ, and L_κ all the indices $\kappa=1$ and 2. We now proceed to utilize these results for the calculation of the surface Green's functions.

III. SURFACE DYNAMICAL GREEN'S FUNCTION FOR A SUPERLATTICE

We will consider here two different cases, depending on the thickness of the last film near the free surface.

A. Surface film with same width as corresponding bulk films

We create two free surfaces by equating to zero all interactions between atoms in the plane ($n=0$, $\kappa=2$, $l_3=L_2$) and atoms in the plane ($n=1$, $\kappa=1$, $l_3=1$). The equation of motion (17) of the infinite superlattice is now changed to

$$(\overleftrightarrow{L}-\delta\overleftrightarrow{L}-\delta\overleftrightarrow{L}^{(s)})\cdot\vec{u}=\vec{0}\ , \tag{26}$$

where

$$\delta L^{(s)}(n,\kappa,l_3;n',\kappa',l_3')=-\gamma\left[\frac{\delta_{n0}\delta_{\kappa 2}\delta_{l_3L_2}}{(M_2)^{1/2}}-\frac{\delta_{n1}\delta_{\kappa 1}\delta_{l_3}}{(M_1)^{1/2}}\right]\left[\frac{\delta_{n'0}\delta_{\kappa' 2}\delta_{l_3'L_2}}{(M_2)^{1/2}}-\frac{\delta_{n'1}\delta_{\kappa' 1}\delta_{l_3'1}}{(M_1)^{1/2}}\right]. \tag{27}$$

We define a surface Green's function by

$$(\overleftrightarrow{L}-\delta\overleftrightarrow{L}-\delta\overleftrightarrow{L}^{(s)})\cdot\overleftrightarrow{G}=\overleftrightarrow{I}\ . \tag{28}$$

The Dyson relation between $\overleftrightarrow{G}$ and $\overleftrightarrow{D}$ enables us to find easily for n and $n'\geq 1$

$$G(n,\kappa,l_3;n',\kappa',l_3')=D(n,\kappa,l_3;n',\kappa',l_3')+\frac{\gamma}{\Delta_{S1}}\left[\frac{D(n,\kappa,l_3;0,2,L_2)}{(M_2)^{1/2}}-\frac{D(n,\kappa,l_3;1,1,1)}{(M_1)^{1/2}}\right]\frac{D(1,1,1;n',\kappa',l_3')}{(M_1)^{1/2}}\ , \tag{29}$$

where

$$\Delta_{S1}=1-\gamma\left[\frac{D(1,1,1;0,2,L_2)}{(M_1M_2)^{1/2}}-\frac{D(1,1,1;1,1,1)}{M_1}\right] . \tag{30}$$

Using Eqs. (24) and (25), one finds

$$\Delta_{S1}=\frac{1}{(t^2-1)}(-1+tA) , \tag{31a}$$

with

$$A=\frac{\cosh[q_1(L_1-\frac{1}{2})]}{\cosh(\frac{1}{2}q_1)\cosh(\frac{1}{2}q_2)}\left[\cosh[q_2(L_2-\tfrac{1}{2})]+\frac{2\gamma_2}{\gamma}\sinh\left[\frac{q_2}{2}\right]\sinh(q_2L_2)\right]$$
$$+\frac{2\gamma_2}{\gamma_1}\frac{\sinh(\frac{1}{2}q_2)}{\cosh(\frac{1}{2}q_2)}\sinh(q_2L_2)\frac{\sinh[q_1(L_1-1)]}{\sinh q_1} . \tag{31b}$$

B. Surface film with width smaller as corresponding bulk films

We now create two other free surfaces by equating to zero all interactions between atoms in the plane ($n=1$, $\kappa=1$, $l_3=l_0<L_1$) and atoms in the plane ($n=1$, $\kappa=1$, $l_3=l_0+1$).

The corresponding variation in the equations of motion [Eq. (29)] is now

$$\delta L^{(s)}(n,\kappa,l_3;n',\kappa',l_3')=-\frac{\gamma_1}{M_1}(\delta_{n1}\delta_{\kappa1}\delta_{l_3l_0}-\delta_{n1}\delta_{\kappa1}\delta_{l_3,l_0+1})(\delta_{n'1}\delta_{\kappa'1}\delta_{l_3',l_0}-\delta_{n'1}\delta_{\kappa'1}\delta_{l_3',l_0+1}) . \tag{32}$$

As above, one obtains the surface Green's function, which for n and $n'\geq 1$ and l_3 and $l_3'\geq l_0$ is

$$G(n,\kappa,l_3;n',\kappa',l_3')=D(n,\kappa,l_3;n',\kappa',l_3')+\frac{\gamma_1}{M_1\Delta_{S1}'}[D(n,\kappa,l_3;1,1,l_0)-D(n,\kappa,l_3;1,1,l_0+1)]D(1,1,l_0+1;n',\kappa',l_3') , \tag{33}$$

where

$$\Delta_{S1}'=1-\frac{\gamma_1}{M_1}[D(1,1,l_0+1;1,1,l_0)-D(1,1,l_0+1;1,1,l_0+1)] . \tag{34}$$

With the use of Eqs. (24) and (25), one finds

$$\Delta_{S1}'=\frac{1}{2}-\frac{t}{t^2-1}\frac{\sinh(\frac{1}{2}q_1)}{\cosh^2(\frac{1}{2}q_1)}\left[\frac{2\gamma_1\gamma_2}{\gamma^2}\tanh\left[\frac{q_2}{2}\right]\sinh(q_2L_2)+\frac{2\gamma_1}{\gamma}\frac{\cosh[q_2(L_2-\frac{1}{2})]}{\cosh(\frac{1}{2}q_2)}+\frac{\gamma_1}{\gamma_2}\frac{\sinh[q_2(L_2-1)]}{\sinh q_2}\right]$$
$$\times\left[\sinh\left[\frac{q_1}{2}\right]\cosh(q_1L_1)+\sinh[q_1(L_1-2l_0-\tfrac{1}{2})]\right]$$
$$-\frac{t}{t^2-1}\frac{1}{\sinh q_1\cosh(\frac{1}{2}q_1)\cosh(\frac{1}{2}q_2)}\left\{2\sinh^2\left[\frac{q_1}{2}\right]\{\sinh[q_1(L_1-\tfrac{1}{2})]-\sinh[q_1(L_1-2l_0-\tfrac{1}{2})]\}\right.$$
$$\times\left[\frac{2\gamma_2}{\gamma}\sinh\left[\frac{q_2}{2}\right]\sinh(q_2L_2)+\cosh[q_2(L_2-\tfrac{1}{2})]\right]$$
$$+\frac{\gamma_2}{\gamma_1}\sinh\left[\frac{q_2}{2}\right]\sinh(q_2L_2)\left[\sinh\left[\frac{q_1}{2}\right]\cosh[q_1(L_1-1)]\right.$$
$$\left.\left.-\sinh\,[q_1(L_1-2l_0-\tfrac{1}{2})]\right]\right\} . \tag{35}$$

We now proceed to use the results of Secs. II and III for the determination of the bulk and surface modes of a superlattice.

IV. BULK AND SURFACE PHONONS OF A SUPERLATTICE

The bulk phonons of our superlattice can be obtained from the knowledge of the bulk Green's function [Eqs. (24) and (25)]. Let us first recall that for the infinite simple cubic lattice described by Eqs. (1)—(11), the threefold degenerate bulk phonon dispersion curves can be obtained from Eqs. (11) and (10), and are given by

$$\zeta_1=\cos(k_3 a_0)\ , \tag{36}$$

where ζ_1 is given by Eq. (10), and k_3 is the propagation vector in the direction x_3 $(-\pi<k_3a_0<+\pi)$. In the same manner (see the Appendix), for the infinite superlattice described by Eqs. (17)—(23) we obtain the bulk phonons from Eqs. (22) and (23):

$$\eta=\cos[k_3(L_1+L_2)a_0]\ , \tag{37}$$

where η is given by Eq. (23) and because the periodicity in the direction x_3 is now given by $(L_1+L_2)a_0$, one has

$$-\pi<k_3(L_1+L_2)a_0<+\pi\ . \tag{38}$$

Because of this larger periodicity in the direction x_3, one has a folding of the phonon dispersion curves in a reduced Brillouin zone specified by Eq. (38) and an opening of new gaps between these folded dispersion curves (see Figs. 1 and 2).

In these gaps, new surface phonons may appear; they can be found from the new poles in the surface Green's functions [Eqs. (29) and (33)] due to the creation of the free surface. Let us illustrate this in the case for which the surface film has the same width as the corresponding bulk films [Eq. (29)]. We work out explicitly the diagonal element of the Green's function $\overleftrightarrow{G}$ on the surface plane. From Eq. (29) we obtain

$$G(1,1,1;1,1,1)=\frac{D(1,1,1;1,1,1)}{\Delta_{S1}}\ . \tag{39}$$

In Δ_{S1} we have a square root of $\pm(1-\eta^2)^{1/2}$ introduced by t [Eq. (22)]. In order to remove it, we multiply the numerator and denominator of Eq. (39) by $(-1+A/t)$. Then Eq. (39) reads

$$G(1,1,1;1,1,1)=\frac{t^2-1}{t}(t-A)\frac{D(1,1,1;1,1,1)}{(2\eta-A)A-1}\ . \tag{40}$$

From Eq. (25) one obtains

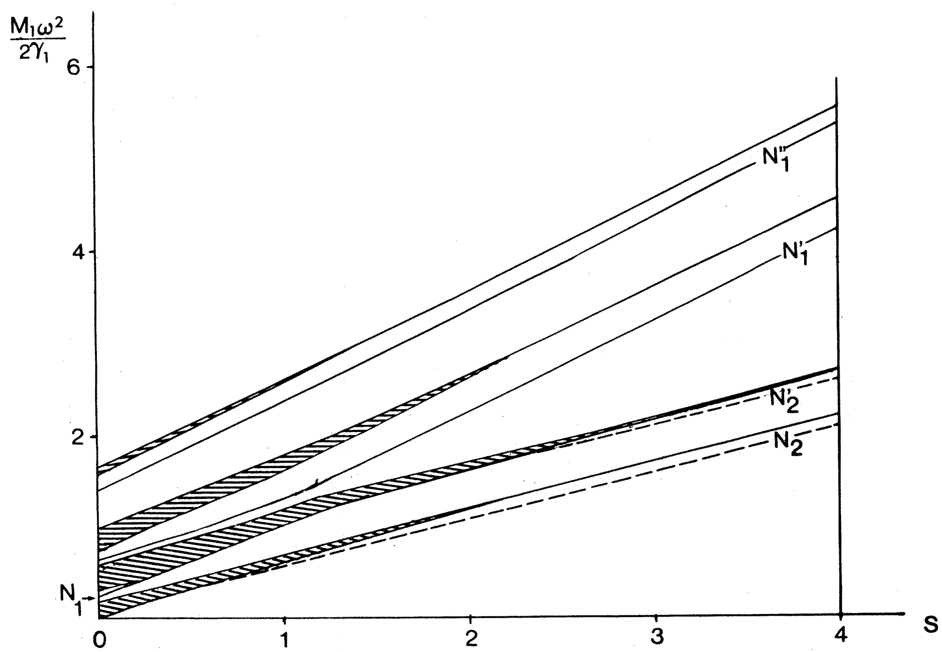

FIG. 1. Band structure of a superlattice with $L_1=L_2=2$ in function of the parameter $S=2-\cos(k_xa_0)-\cos(k_ya_0)$. The two crystals differ by their atomic masses ($M_1=2M_2$ or $M_1=M_2/2$) while the force constants are identical everywhere ($\gamma_1=\gamma_2=\gamma$). The film labeled by the index $\kappa=1$ is at the surface. The shaded areas represent the bulk bands (four of each value of $k_{\|}$). Surface modes are present for $e_0=2$ (same thickness of the crystal film at the surface as in the bulk) but not for $e_0=1$. The N_1,N_1',N_1'' modes (——) refer to a surface film of lighter mass ($M_1=M_2/2$), the N_2,N_2' modes (— — —) to a surface film of heavier mass ($M_1=2M_2$).

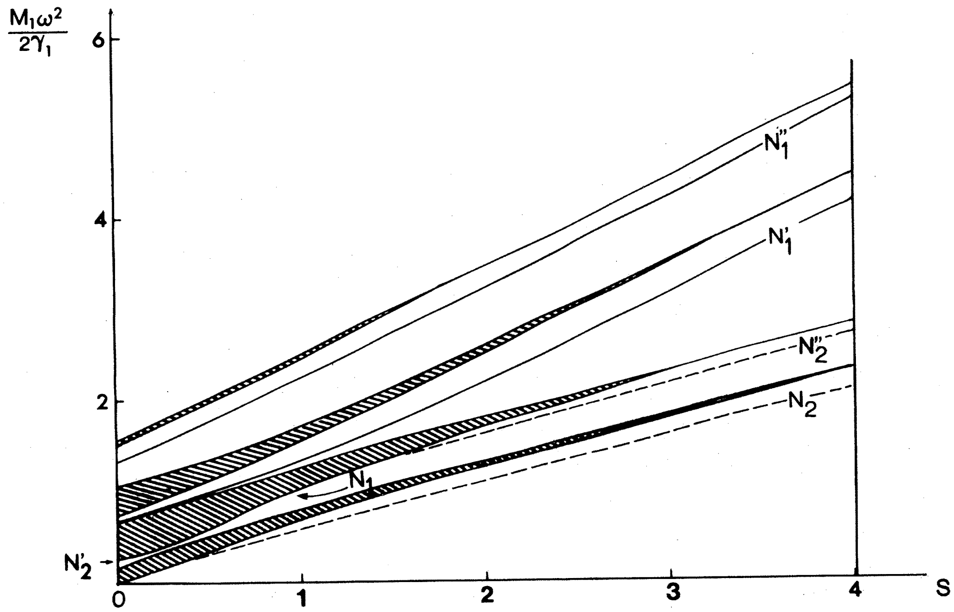

FIG. 2. Same as in Fig. 1 but with crystals of the same atomic mass ($M_1=M_2$) and different force constants ($\gamma_1=2\gamma_2$ or $\gamma_1=\gamma_2/2$). The force constant at the interfaces are assumed to be $\gamma=(\gamma_1+\gamma_2)/2$. The N_1,N_1',N_1'' modes (——) refer to $\gamma_1=2\gamma_2$, the N_2, N_2',N_2'' modes to $\gamma_1=\gamma_2/2$.

$$D(1,1,1;1,1,1)=\frac{M_1}{2\gamma_1}\frac{t}{(t^2-1)}\frac{F(\omega^2)}{\sinh(\tfrac{1}{2}q_1)\cosh(\tfrac{1}{2}q_1)\cosh(\tfrac{1}{2}q_2)}\,, \tag{41}$$

where

$$\begin{aligned}F(\omega^2)=&\frac{4\gamma_1\gamma_2}{\gamma^2}\sinh\left[\frac{q_1}{2}\right]\sinh\left[\frac{q_2}{2}\right]\sinh(q_2L_2)\cosh[q_1(L_1-\tfrac{1}{2})]+\frac{4\gamma_1}{\gamma}\sinh\left[\frac{q_1}{2}\right]\cosh[q_1(L_1-\tfrac{1}{2})]\cosh[q_2(L_2-\tfrac{1}{2})]\\&+\frac{2\gamma_2}{\gamma}\sinh\left[\frac{q_2}{2}\right]\sinh(q_2L_2)\sinh[q_1(L_1-1)]+\frac{\gamma_1}{\gamma_2}\frac{\sinh(\tfrac{1}{2}q_1)}{\sinh(\tfrac{1}{2}q_2)}\cosh[q_1(L_1-\tfrac{1}{2})]\sinh[q_2(L_2-1)]\\&+\cosh[q_2(L_2-\tfrac{1}{2})]\sinh[q_1(L_1-1)]\,.\end{aligned} \tag{42}$$

In addition, Eqs. (23) and (31b) provide us with

$$(2\eta-A)A-1=\frac{F(\omega^2)D(\omega^2)}{\gamma_1\cosh^2(\tfrac{1}{2}q_1)\cosh^2(\tfrac{1}{2}q_2)\sinh(\tfrac{1}{2}q_1)}\,, \tag{43}$$

where

$$\begin{aligned}D(\omega^2)=&\frac{2\gamma_1\gamma_2}{\gamma}\sinh\left[\frac{q_1}{2}\right]\sinh\left[\frac{q_2}{2}\right]\sinh(q_1L_1)\sinh(q_2L_2)+\gamma_2\sinh\left[\frac{q_2}{2}\right]\cosh[q_1(L_1-\tfrac{1}{2})]\sinh(q_2L_2)\\&+\gamma_1\sinh\left[\frac{q_1}{2}\right]\sinh(q_1L_1)\cosh[q_2(L_2-\tfrac{1}{2})]\,.\end{aligned} \tag{44}$$

And finally, by combining Eqs. (40)–(44),

$$G(1,1,1;1,1,1)=\frac{t-A}{2}\frac{M_1\cosh(\tfrac{1}{2}q_1)\cosh(\tfrac{1}{2}q_2)}{D(\omega^2)}\,. \tag{45}$$

From the poles of this Green's function, we find the frequencies ω_S of the surface modes localized at the free surface ($\eta=1$, $\kappa=1$, $l_3=1$) and decaying inside the bulk situated at n and $l_3>1$. They are given by

$$D(\omega_S^2)=0\,. \tag{46a}$$

Let us call A_S and t_S the corresponding values of A and t [Eq. (31)]. From Eq. (31a), we must have for these surface modes

$$t_S = \frac{1}{A_S} .$$

Since t_S is the exponential decaying factor for these surface modes [see Eq. (25)], the frequencies of these modes are given by Eq. (46a) with the following condition,

$$|A_S| > 1 ,$$

which, with the help of Eqs. (46a) and (31b), can be rewritten as

$$\frac{\gamma_2}{\gamma_1} \left| \frac{\sinh(q_2 L_2)\tanh(\frac{1}{2}q_2)}{\sinh(q_1 L_1)\tanh(\frac{1}{2}q_1)} \right| > 1 . \qquad (46b)$$

V. A FEW APPLICATIONS

A. The biatomic linear chain

The simplest possible example is the one of a linear biatomic chain, which can be obtained as a limit of our model with $L_1 = L_2 = 1$, $\gamma_1 = \gamma_2 = \gamma$, and $k_{||} = 0$. Then one easily finds from Eqs. (46)

$$\omega^2 = \gamma \left[\frac{1}{M_1} + \frac{1}{M_2} \right] \qquad (47a)$$

and

$$\frac{M_2}{M_1} > 1 , \qquad (47b)$$

which is the well-known result for the surface optical mode of a biatomic linear chain with a free surface on the atom of mass M_1.

B. The elastic limit

The bulk dispersion relation [Eqs. (36) and (10)] of the simple cubic Montroll-Potts model, reduces in the limit of long wavelength to

$$\omega^2 = \frac{\gamma_1}{M_1} a_0^2 k^2 = c_{t_1}^2 k^2 + \cdots , \qquad (48)$$

where

$$k^2 = k_1^2 + k_2^2 + k_3^2 .$$

In this limit ζ_κ ($\kappa = 1$ or 2) given by Eq. (10) reduces to

$$\zeta_\kappa = 1 + \frac{a_0^2}{2} \alpha_\kappa^2 + \cdots , \qquad (49a)$$

where

$$\alpha_\kappa^2 = \kappa_{||}^2 - \frac{\omega^2}{c_{t\kappa}^2} . \qquad (49b)$$

Recalling that $t_\kappa = e^{q_\kappa} = 1 + q_\kappa + \cdots$ [Eq. (21)], we see with the help of Eq. (11) that

$$t_\kappa = 1 - \alpha_\kappa a_0 + \cdots ,$$

for $\zeta_\kappa > 1$ and $-1 < \zeta_\kappa + 1$, which are the two regions of interest in the elastic limit. Then

$$q_\kappa = -\alpha_\kappa a_0 \ll 1 . \qquad (50)$$

We can now take the elastic limit of Eqs. (46) giving the surface modes. Keeping only the terms of first order in q_κ and neglecting the term in $q_1 q_2$, we obtain

$$F \tanh(\alpha_1 L_1) + \tanh(\alpha_2 L_2) = 0 \qquad (51a)$$

and

$$\left| \frac{\cosh(\alpha_2 L_2)}{\cosh(\alpha_1 L_1)} \right| > 1 , \qquad (51b)$$

where

$$F = \frac{\alpha_1 \rho_1 c_{t_1}^2}{\alpha_2 \rho_2 c_{t_2}^2} , \qquad (51c)$$

and where ρ_1 and ρ_2 are the mass densities. This result was simultaneously obtained with the help of the elasticity theory.[5,6]

C. A few examples of the bulk and surface phonons

In Figs. 1 and 2 we present the results for the band structure and surface modes of a superlattice with two atomic planes in each film ($L_1 = L_2 = 2$). We first assume (Fig. 1) that the two crystals differ only by their atomic masses, while the force constants are identical everywhere ($\gamma_1 = \gamma_2 = \gamma$). Figure 1 presents the surface modes when the crystal film at the surface is either the type 1 (mass M_1) or of type 2 (mass M_2). However, all these modes refer to a thickness of the crystal at the surface equal to two atomic layers ($e_0 = 2$). For $e_0 = 1$ there is no more surface mode. This sensitivity of the surface modes to the surface film thickness was already pointed out in the elastic limit.[5,6] Let us stress that care must be taken to obtain the surface modes, in the case $e_0 < L_1$, from the roots of Δ'_{S1} [Eq. (35)]. As a matter of fact, some of the roots of Δ'_{S1} appear as well as roots in the numerator of this Green's function in Eq. (33) and then should be disregarded.

We next assume (Fig. 2) that all the atoms have the same mass ($M_1 = M_2$) but the force constants are different in the two crystals. The force constants at the interface is taken to be $\gamma = \frac{1}{2}(\gamma_1 + \gamma_2)$. It is worthwhile to note that the band structure is sensitive to the choice of γ, whose effect becomes more important as the film thicknesses decrease. As in Fig. 1, the surface modes are represented for the crystal film at the surface either of type 1 or of type 2. Moreover, the surface modes are only present for $e_0 = 2$, in these simple examples.

VI. CONCLUSIONS

In this paper we obtained for the first time surface phonons on a simple three-dimensional atomic model of a superlattice. The simplicity of this model enables us to derive in closed form the bulk and surface dynamical

Green's functions for a superlattice. From the poles of these Green's functions we obtained analytic expressions for the bulk and surface phonons of a superlattice. We analyzed also the case for which the width of the last surface slab was smaller than that of the corresponding bulk slabs. A few specific examples illustrate these general results.

A mathematical transposition of this theory to electrons in a bimetallic superlattice is underway, within the tight-binding approximation.[14] Another extension to phonons in superlattices formed out of two different biatomic slabs is also under study. Finally let us also mention our interest in liquid superlattices, following the theory of liquid surfaces done before.[15]

APPENDIX

In this appendix, we will explain how one can obtain the bulk Green's function $\overleftrightarrow{D}$ of a superlattice. We start with the Dyson equation (20) relating $\overleftrightarrow{D}$ to the Green's function $\overleftrightarrow{U}$ of the same infinite set of uncoupled films. $\overleftrightarrow{U}$ was obtained in closed form [Eqs. (15) and (12)]. The perturbation $\delta\overleftrightarrow{L}$ which couples all these films together is given by Eq. (18).

For the evaluation of the elements of $\overleftrightarrow{D}$, it is helpful to define

$$f(n,n')=\frac{1}{(M_1)^{1/2}}D(n,1,L_1;n',\kappa',l_3')-\frac{1}{(M_2)^{1/2}}D(n,2,1;n',\kappa',l_3')\,, \tag{A1}$$

$$g(n,n')=\frac{1}{(M_2)^{1/2}}D(n-1,2,L_2;n',\kappa',l_3')-\frac{1}{(M_1)^{1/2}}D(n,1,1;n',\kappa',l_3')\,, \tag{A2}$$

and

$$\Delta=1-\gamma\left[\frac{U_1(1,1)}{M_1}+\frac{U_2(1,1)}{M_2}\right]. \tag{A3}$$

When use is made of the Dyson equation (20), one easily finds two coupled equations for $f(n,n')$ and $g(n,n')$

$$\Delta g(n,n')+\frac{\gamma}{M_1}U_1(1,L_1)f(n,n')+\frac{\gamma}{M_2}U_2(1,L_2)f(n-1,n')=\delta_{n-1,n'}\frac{\delta_{2\kappa'}}{(M_2)^{1/2}}U_2(L_2,l_3')-\delta_{n,n'}\frac{\delta_{1\kappa'}}{(M_1)^{1/2}}U_1(1,l_3') \tag{A4}$$

and

$$\Delta f(n,n')+\frac{\gamma}{M_1}U_1(1,L_1)g(n,n')+\frac{\gamma}{M_2}U_2(1,L_2)g(n+1,n')=\delta_{n,n'}\left[\frac{\delta_{1\kappa'}}{(M_1)^{1/2}}U_1(L_1,l_3')-\frac{\delta_{2\kappa'}}{(M_2)^{1/2}}U_1(1,l_3')\right]. \tag{A5}$$

By elimination of $f(n,n')$, one obtains from the above two equations

$$Bg(n,n')+\widetilde{A}[g(n+1,n')+g(n-1,n')]=C_2\delta_{n,n'}+C_1\delta_{n-1,n'}\,, \tag{A6a}$$

where

$$B=\Delta-\frac{\gamma^2}{\Delta}\left[\frac{U_1^2(1,L_1)}{M_1^2}+\frac{U_2^2(1,L_2)}{M_2^2}\right], \tag{A6b}$$

$$\widetilde{A}=-\frac{\gamma^2}{M_1M_2\Delta}U_1(1,L_1)U_2(1,L_2)\,, \tag{A6c}$$

$$C_2=-\delta_{1\kappa'}\frac{U_1(1,l_3')}{(M_1)^{1/2}}-\frac{\gamma}{\Delta}\frac{U_1(1,L_1)}{M_1}\left[\delta_{1\kappa'}\frac{U_1(L_1,l_3')}{(M_1)^{1/2}}-\delta_{2\kappa'}\frac{U_2(1,l_3')}{(M_2)^{1/2}}\right], \tag{A6d}$$

and

$$C_1=\delta_{2\kappa'}\frac{U_2(L_2,l_3')}{(M_2)^{1/2}}-\frac{\gamma}{\Delta}\frac{U_2(1,L_2)}{M_2}\left[\delta_{1\kappa'}\frac{U_1(L_1,l_3')}{(M_1)^{1/2}}-\delta_{2\kappa'}\frac{U_2(1,l_3')}{(M_2)^{1/2}}\right]. \tag{A6e}$$

Then Eq. (A6) can be solved, remembering that the solution of

$$h(n+1,n')+h(n-1,n')-2\eta h(n,n')=\delta_{n,n'}\,, \tag{A7a}$$

where

$$2\eta=-\frac{B}{\widetilde{A}}\,, \tag{A7b}$$

is

$$h(n,n')=\frac{t^{|n-n'|+1}}{t^2-1}\ , \tag{A8a}$$

where

$$t=\begin{cases}\eta-(\eta^2-1)^{1/2}, & \eta>1\\ \eta+i(1-\eta^2)^{1/2}, & -1<\eta<1\\ \eta+(\eta^2-1)^{1/2}, & \eta<-1.\end{cases} \tag{A8b}$$

Returning to Eq. (A6) one obtains

$$g(n,n')=\frac{C_2}{\widetilde{A}}\frac{t^{|n-n'|+1}}{t^2-1}+\frac{C_1}{\widetilde{A}}\frac{t^{|n-n'-1|+1}}{t^2-1}\ . \tag{A9}$$

Then Eq. (A5) provides us with $f(n,n')$ and the Dyson equation (20) gives

$$D(n,\kappa,l_3;n',\kappa',l_3')=\delta_{n,n'}\delta_{\kappa\kappa'}U_\kappa(l_3,l_3')+\gamma\left\{\frac{\delta_{\kappa1}}{(M_1)^{1/2}}[U_1(l_3,L_1)f(n,n')-U_1(l_3,1)g(n,n')]\right.$$
$$\left.+\frac{\delta_{\kappa2}}{(M_2)^{1/2}}[U_2(l_3,L_2)g(n+1,n')-U_2(l_3,1)f(n,n')]\right\}\ . \tag{A10}$$

And finally after some algebra, we obtain the $D(n,\kappa,l_3;n',\kappa',l_3')$ in the form given by Eqs. (27) and (28).

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