Interface response theory of phonons in N-layer superlattices

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Bulk phonons were obtained previously for a two-layer superlattice, using slab response functions. We show that they can be obtained more directly using bulk response functions and the interface response theory. This new approach enables us to give for the first time a general theory of phonons in N-layer superlattices. As an illustration of the theory, we give a closed-form expression for phonons in three-layer superlattices.

I. INTRODUCTION

Phonons in two-layer superlattices are of interest theoretically[1] and also have been studied experimentally by Raman and Brillouin spectroscopies for several years. N-layer superlattices are formed out of a periodic repetition of a unit cell containing N ($N>2$) different slabs. There has been progress recently in producing such materials[2] and we hope that the present theoretical paper will stimulate experimental investigations of phonons in N-layer superlattices. In the next section, we first present 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 theory[3] enables us to calculate the response function and the superlattice phonons. A closed-form expression for the phonons in a three-layer superlattice is presented here for the first time. Finally, we discuss some extensions of the present results.

II. BULK PHONON MODEL AND THE SURFACE REPONSE OPERATORS

A. Bulk phonon model

We start from an infinite simple-cubic lattice of atoms of mass M_i. Let $u_\alpha(l)$ denote the α (=1, 2, or 3) component of the displacement of the atom at lattice site

$$\mathbf{x}(l)=a_0(l_1\hat{\mathbf{x}}_1+l_2\hat{\mathbf{x}}_2+l_3\hat{\mathbf{x}}_3) , \tag{1}$$

where a_0 is the lattice parameter and $\hat{\mathbf{x}}_1$, $\hat{\mathbf{x}}_2$, and $\hat{\mathbf{x}}_3$ are unit vectors. The potential energy Φ_i associated with the lattice vibrations of the model[4,5] considered here has the form

$$\Phi_i=\tfrac{1}{2}\beta_i\sum_l\sum_p\sum_\alpha[u_\alpha(l)-u_\alpha(l+p)]^2 , \tag{2}$$

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. Nevertheless, this deficiency is unimportant for the qualitative investigation of many physical properties and in particular for our study of the transverse and longitudinal polarized phonons.[6]

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

$$\sum_{l'}H_{0i}(ll';\omega^2)\mathbf{u}(l')=\mathbf{0} , \tag{3}$$

where

$$H_{0i}(ll';\omega^2)=\left[\omega^2-6\frac{\beta_i}{M_i}\right]\delta_{ll'}+\frac{\beta_i}{M_i}\sum_p\delta_{l,l'+p} . \tag{4}$$

The corresponding threefold-degenerate bulk phonon dispersion relation is

$$\omega^2=2\frac{\beta_i}{M_i}[3-\cos(k_1a_0)-\cos(k_2a_0)-\cos(k_3a_0)] , \tag{5}$$

where $\mathbf{k}=(k_1, k_2, k_3)$ is the propagation vector.

B. Bulk response function

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

$$\overleftrightarrow{H}_{0i}\overleftrightarrow{G}_{0i}=\overleftrightarrow{I} , \tag{6}$$

where $\overleftrightarrow{I}$ stands for the unit matrix.

Taking advantage of the periodicity of the system in directions parallel to the (001) planes, we introduce the following two-dimensional vectors,

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

$$\mathbf{k}_{\parallel}(l)=k_1\hat{\mathbf{x}}_1+k_2\hat{\mathbf{x}}_2 , \tag{7b}$$

and a Fourier transformation of the response function

$$G_{0i}(ll';\omega^2)=\frac{1}{N^2}\sum_{\mathbf{k}_\parallel} G_{0i}(ll';\mathbf{k}_\parallel\omega^2)\times\exp\{i\mathbf{k}_\parallel\cdot[\mathbf{x}_\parallel(l)-\mathbf{x}_\parallel(l')]\}\ , \tag{8}$$

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

The corresponding bulk response function is[6]

$$G_{0i}(l_3l_3';\mathbf{k}_\parallel\omega^2)=\frac{M_i}{\beta_i}\frac{t_i^{|l_3-l_3'|+1}}{t_i^2-1}\ , \tag{9}$$

with

$$t_i=\begin{cases}\xi_i-(\xi_i^2-1)^{1/2}, & \xi_i>1\\ \xi_i+i(1-\xi_i^2)^{1/2}, & -1<\xi_i<1\\ \xi_i+(\xi_i^2-1)^{1/2}, & \xi_i<-1,\end{cases} \tag{10}$$

and

$$\xi_i=3-\cos(k_1a_0)-\cos(k_2a_0)-\frac{M_i}{2\beta_i}(\omega^2+i\varepsilon)\ , \tag{11}$$

where ε is an infinitesimal positive number.

C. The surface response operators for one slab

Let us now create 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 $l_3=L_i+1$ planes. The corresponding cleavage operator $\overleftrightarrow{V}_{0i}$, which when added to the $\overleftrightarrow{H}_{0i}$ gives the dynamical matrix $\overleftrightarrow{h}_{0i}$ of the slab and of the two semi-infinite crystals, is

$$V_{0i}(l_3l_3')=\frac{\beta_i}{M_i}(\delta_{l_30}\delta_{l_3'0}+\delta_{l_31}\delta_{l_3'1}-\delta_{l_30}\delta_{l_3'1}-\delta_{l_31}\delta_{l_3'0})+\frac{\beta_i}{M_i}(\delta_{l_3L_i}\delta_{l_3'L_i}+\delta_{l_3,L_i+1}\delta_{l'_3,L_i+1}-\delta_{l_3L_i}\delta_{l_3',L_i+1}-\delta_{l_3,L_i+1}\delta_{l_3'L_i})\ . \tag{12}$$

The surface response operator $\overleftrightarrow{A}'_{si}$ associated to this slab is formed out of the elements of

$$\overleftrightarrow{A}'_{0i}=\overleftrightarrow{G}_{0i}\overleftrightarrow{V}_{0i}\ , \tag{13}$$

belonging strictly to the slab, namely

$$A'_{si}(l_3l_3')=-\frac{1}{t_i+1}(\delta_{l_3'1}t_i^{l_3}+\delta_{l_3'L_i}t_i^{L_i-l_3+1})\qquad 1\le l_3,l_3'\le L_i\ . \tag{14}$$

The above expressions (9)–(14) in another form were used before[7] for the study of sandwich phonons. They enable us to treat in the next section the N-layer superlattice phonons.

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

A. Definition of the N-layer superlattice

Consider N different homogeneous slabs, each bounded by ideally truncated free surfaces. Let these N ideally cleaved slabs $(i=1,2,\ldots,N)$ be coupled together by nearest-neighbor interactions $\beta_{ii'}$, between adjacent surface atoms; β_{12} couples surface atoms of the L_1 atomic plane of the slab $i=1$ to those of the $l_3=1$ atomic plane of the slab $i=2$ and so on. The unit cell formed by these N slabs will be labeled by an integer n.

In the same manner, one can couple periodically an infinite number $(-\infty<N<+\infty)$ of the above N layered slabs in order to obtain a new bulk material, the N-layer superlattice.

In what follows, the following more condensed notation,

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

$$m'\equiv n'N+i'\equiv(n',i')\ .$$

will prove convenient for labeling a given slab i (or i') in the unit cell n (or n').

The interface atomic interactions are globally represented by the coupling operator $\overleftrightarrow{V}_I$:

$$\begin{aligned}V_I(ml_m;m'l'_{m'})=\;&\delta_{mm'}(\delta_{l_m,l_{N_m}}\delta_{l'_{m'},l'_{N'_m}}\beta_{m,m+1}\\&+\delta_{l_m,1}\delta_{l'_{m'},1}\beta_{m-1,m})\\&-\delta_{m+1,m'}\delta_{l_m,l_{N_m}}\delta_{l'_{m'},1}\beta_{m,m+1}\\&-\delta_{m,m'-1}\delta_{l_m,1}\delta_{l'_{m'},l_{N'_{m'}}}\beta_{m,m+1}\ .\end{aligned} \tag{15}$$

B. Reference response function $\overleftrightarrow{G}$ of the N-layer superlattice

Define[3] for the N-layered superlattice a reference response function $\overleftrightarrow{G}$ as a block-diagonal matrix formed out of only the elements of the bulk response functions $\overleftrightarrow{G}_{0i}$ contained within the space of the definition of each slab, namely, with the notation given above:

$$G(ml_3,m'l_3')=\delta_{mm'}G_m(l_3l_3'),\quad 1\le l_3,l_3'\le L_m \tag{16}$$

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

C. Response function $\overleftrightarrow{g}$ of the N-layer superlattice

The response function $\overleftrightarrow{g}$ or the N-layer superlattice can be calculated directly from the reference response function $\overleftrightarrow{G}$ defined above, through the universal relation[3]

$$(\overleftrightarrow{I}+\overleftrightarrow{A}')\overleftrightarrow{g}=\overleftrightarrow{G}\ , \tag{17}$$

where the interface response operator $\overleftrightarrow{A}'$ is defined[3] by

$$\overleftrightarrow{A}'=\overleftrightarrow{A}'_s+\overleftrightarrow{G}\overleftrightarrow{V}_I\ . \tag{18}$$

$\overleftrightarrow{A}'_s$ is the block-diagonal surface response operator formed out of the $A'_s(ml_3;\, m'l'_3)$ given by Eq. (14),

$$A'_s(ml_3;m'l'_3)=\delta_{mm'}A'_{sm}(l_3l'_3)\ , \tag{19}$$

and $\overleftrightarrow{V}_I$ is the coupling operator defined above.

The general solution of Eq. (17) for any model of an N-layer superlattice was given before.[8] For the present model, the elements $g(ml_3;m'l'_3)$ are scalars and can be calculated in closed form with the help of the following (2×2) matrices:

$$\overleftrightarrow{K}(m)=\begin{vmatrix} A'(m1;mL_m) & A'(m1;m+1,1) \\ 1+A'(mL_m;mL_m) & A'(mL_m;m+1,1)\end{vmatrix}\ , \tag{20}$$

$$\overleftrightarrow{H}(m)=\begin{vmatrix} A'(m1;m-1,L_{m-1}) & 1+A'(m1;m1) \\ A'(mL_m;m-1,L_{m-1}) & A'(mL_m;m1)\end{vmatrix}\ , \tag{21}$$

$$\overleftrightarrow{P}(m)=-\overleftrightarrow{K}^{-1}(m)\overleftrightarrow{H}(m)\ , \tag{22}$$

and

$$\overleftrightarrow{R}(N)=\overleftrightarrow{P}(N)\overleftrightarrow{P}(N-1)\cdots\overleftrightarrow{P}(1)\ . \tag{23}$$

The explicit expressions for $\overleftrightarrow{K}(m)$ and $\overleftrightarrow{H}(m)$ are

$$\overleftrightarrow{K}(m)=\begin{vmatrix} \frac{t_m^{L_m}}{t_m+1}+\frac{\beta_{m,m+1}}{\beta_m}\frac{t_m}{t_m^2-1} & -\frac{\beta_{m,m+1}}{\beta_m}\frac{t_m^{L_m}}{t_m^2-1} \\ \frac{1}{t_m+1}+\frac{\beta_{m,m+1}}{\beta_m}\frac{t_m}{t_m^2-1} & -\frac{\beta_{m,m+1}}{\beta_m}\frac{t_m}{t_m^2-1}\end{vmatrix}\ , \tag{24}$$

and

$$\overleftrightarrow{H}(m)=\begin{vmatrix} -\frac{\beta_{m-1,m}}{\beta_m}\frac{t_m}{t_m^2-1} & \frac{1}{t_m+1}+\frac{\beta_{m-1,m}}{\beta_m}\frac{t_m}{t_m^2-1} \\ -\frac{\beta_{m-1,m}}{\beta_m}\frac{t_m^{L_m}}{t_m^2-1} & \frac{t_m^{L_m}}{t_m+1}+\frac{\beta_{m-1,m}}{\beta_m}\frac{t_m^{L_m}}{t_m^2-1}\end{vmatrix}\ . \tag{25}$$

Note also that

$$\det\overleftrightarrow{K}(m)=-\frac{\beta_{m,m+1}}{\beta_m}\frac{t_m^{L_m}}{t_m^2-1}\ . \tag{26}$$

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

$$t_m=e^{q_m} \tag{27}$$

and

$$A_m=\frac{1}{\beta_m}\frac{\sinh[(L_m-1)q_m]}{\sinh(q_m)}\ , \tag{28}$$

$$B_m=\frac{\cosh[(L_m-\frac{1}{2})q_m]}{\cosh(q_m/2)}\ , \tag{29}$$

and

$$C_m=2\beta_m\tanh(q_m/2)\sinh(q_mL_m)\ . \tag{30}$$

Then $\overleftrightarrow{P}(m)$ is for the present phonon model given by

$$\overleftrightarrow{P}(m)=\begin{vmatrix} -\beta_{m-1,m}A_m & \beta_{m-1,m}A_m+B_m \\ -\beta_{m-1,m}A_m-\frac{\beta_{m-1,m}}{\beta_{m,m+1}}B_m & \beta_{m-1,m}A_m+\left[1+\frac{\beta_{m-1,m}}{\beta_{m,m+1}}\right]B_m+\frac{1}{\beta_{m,m+1}}C_m\end{vmatrix}\ . \tag{31}$$

Use can be made of these results (24)–(31) for the calculation of the elements of the response function $\overleftrightarrow{g}$ for a two-layer superlattice[9].

D. The N-layered superlattice phonons

The expression giving the N-layer superlattice phonons appears in the denominator of $g(nil_3;n'i'l'_3)$. It was also shown[8] that one can obtain N-layered superlattice modes directly from the trace of the (2×2) matrix $\overleftrightarrow{R}(N)$, namely

$$\cos(k'_3a_u)=\mathrm{Tr}\overleftrightarrow{R}(N)\ , \tag{32}$$

where

$$a_u=a_0\sum_{i=1}^{N}L_i \tag{33}$$

is the width of the unit cell of the superlattice and k'_3 the component of propagation vector $\mathbf{k}$ perpendicular to the interfaces.

So for any value of the integer $N\geq 1$, the result given by Eq. (32), together with the Eqs. (23) and (27)–(31), enables us to find a closed form for the expression for N-layered superlattice phonons. In what follows, we give the results for $N=2$ and $N=3$.

The expression for a two-layer superlattice phonon is

$$2\cos(k'_3a_u)=2B_1B_2+\frac{C_1C_2}{\beta_{12}}+\epsilon_{ij}\left[\frac{1}{2}A_iC_j+\frac{1}{\beta_{12}}C_iB_j\right]\ , \tag{34}$$

where the Einstein summation rule for tensors is used and the Levi-Civita symbol is

$$\epsilon_{ij}=\begin{cases}1 & \text{if } i,j=1 \text{ or } 2,\ i\neq j\\ 0 & \text{otherwise}\,.\end{cases} \tag{35}$$

This result was given before[1] in a less condensed form.

The new result for three-layer superlattice phonons is

$$2\cos(k_3'a_u)=2B_1B_2B_3+\frac{C_1C_2C_3}{\beta_{12}\beta_{23}\beta_{31}}+|\epsilon_{ijk}|\left[B_iA_jC_k+\tfrac{1}{2}B_iB_jC_k\left[\frac{1}{\beta_{12}}+\frac{1}{\beta_{23}}+\frac{1}{\beta_{31}}\right]+\frac{1}{2\beta_{jk}}A_iC_jC_k+\frac{1}{2\beta_{jk}}B_iC_jC_k\left[\frac{1}{\beta_{ki}}+\frac{1}{\beta_{ij}}\right]\right], \quad (36)$$

bearing in mind again the Einstein summation rule for tensors and the Levi-Civita symbol,

$$\epsilon_{ijk}=\begin{cases}1 \text{ if } i,j,k \text{ is an even permutation of } i,j,k=1,2,3\\ -1 \text{ if } i,j,k \text{ is an odd permuation of } i,j,k=1,2,3\\ 0 \text{ otherwise}.\end{cases} \quad (37)$$

This closed-form result (36) gives implicitly the dispersion relations for three-layer superlattice phonons (frequency versus wave vector). In Eq. (36), the wave-vector component k_3' normal to the layers is clearly visible; the wave-vector components k_1,k_2 parallel to the layers and the frequency ω appear in the quantities defined by Eqs. (10), (11), and (27)–(30). If there are many atomic layers in each superlattice slab, there will be a large number of solutions ω inside the reduced Brillouin zone. Equation (36) can be solved easily numerically in the same manner as for a two-layer superlattice.[1] The results of an explicit calculation of three-layer superlattice phonons will be given in a forthcoming paper.[9] An extension to a four-layer semiconductor superlattice,[9] and a comparison with forthcoming experimental results,[10] are also in progress.

IV. DISCUSSION

A general theory of phonons in N-layered superlattices was presented for the first time in this paper, in the frame of a simple phonon model. Although the lattice model is somewhat simple, it is sufficient to illustrate the salient features.

The present formalism can be made to deal fairly straightforwardly[8] with more sophisticated models, in order to deal with acoustic as well as optic modes in ionic N-layered superlattices. In such more-realistic models, there will also be, in general, a coupling between transversely and longitudinally polarized modes.

A complete discussion of all possible extensions and improvements of the present paper would be very lengthy, especially when one recalls that the interface response theory applies to any composite system, without any limitations in the shape of the interfaces and the number of components. If the present paper stimulates more realistic and interesting future theoretical and experimental studies of composite systems, that will be a source of considerably satisfaction to its authors.

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