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Eigenvectors of composite systems: II. Phonon eigenvectors in some layered materials

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Abstract. The general theory of deformations and eigenvectors of composite systems presented in the preceding paper (I) is illustrated here by application to the phonon eigenvector of layered composite systems. Explicit expressions for these eigenvectors are derived for a simple model of several layered composite systems: semi-infinite solid, one slab, a double-layer slab and one adsorbed slab on a semi-infinite crystal.

1. Introduction

The study of phonons in composite materials is only beginning. However, if one considers a slab and a semi-infinite crystal as simple composite materials, one can foresee a little how the theoretical study of more complex composite materials can be tackled in the near future. The theoretical investigation of surface phonons (see, e.g., [1]) is done mostly by direct numerical diagonalisation of the slab dynamical matrix [2], by matching the eigenvectors at the surface [3] and by using response functions [4]. The first two methods also provide the frequencies of the surface phonons as well as their eigenvectors. The method using response functions provided in the past the frequencies of the surface phonons and not their eigenvectors [1, 4]. Knowledge of the eigenvectors is necessary if one wants to study the localisation of these vibrational modes.

In this paper, we show how all the phonon eigenvectors can be calculated within the response function approach, for a surface of a semi-infinite crystal, a single slab, a double-layer slab and one adsorbed slab on a semi-infinite crystal.

In § 2, we shall first recall the simple phonon model for which all these studies are done. The simplicity of this model enables us to make all calculations in closed form, so that any motivated reader can easily recalculate them and be able then to solve the eigenvalue problem for more sophisticated models, along the same lines.

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2. The phonon model

The bulk phonon model used in this paper is the well known Montroll–Potts [5] model used for many studies of surface phonons [1] and also more recently for the lattice dynamics of systems with two interfaces [6]. We shall therefore recall only its main features here.

This model assumes an infinite crystal of type i to be a simple cubic lattice of atoms of mass m_i. Let $u(n)$ denote the threefold-degenerate component of the displacement of the atoms at lattice site $\boldsymbol{x}(n) = a_0(n_1\hat{\boldsymbol{x}}_1 + n_2\hat{\boldsymbol{x}}_2 + n_3\hat{\boldsymbol{x}}_3)$, where a_0 is the lattice parameter and $\hat{\boldsymbol{x}}_1$, $\hat{\boldsymbol{x}}_2$ and $\hat{\boldsymbol{x}}_3$ are unit vectors. The model assumes atomic interactions β_i with the six first nearest neighbours of each atom. The bulk phonon dispersion relation is

$$\omega^2 = 2(\beta_i/m_i)[3 - \cos(k_1a_0) - \cos(k_2a_0) - \cos(k_3a_0)] \tag{1}$$

where $\boldsymbol{k}$ is the propagation vector.

The corresponding bulk response function is [1, 6] within the $\boldsymbol{k}_\parallel \equiv (k_1, k_2)$ representation

$$G_{oi}(nn'; \boldsymbol{k}_\parallel \omega^2) = (m_i/\beta_i)[t_i^{|n_3-n_3'|+1}/(t_i^2 - 1)] \tag{2}$$

with

$$t_i = \begin{cases} \xi_i - (\xi_i^2 - 1)^{1/2} & \xi_i > 1 \\ \xi_i + \mathrm{i}(1 - \xi_i^2)^{1/2} & -1 < \xi_i < 1 \\ \xi_i + (\xi_i^2 - 1)^{1/2} & \xi_i < -1 \end{cases} \tag{3a}$$

and

$$\xi_i = 3 - \cos(k_1a_0) - \cos(k_2a_0) - (m_i/2\beta_i)(\omega^2 + \mathrm{i}\varepsilon). \tag{3b}$$

For what follows, it is also useful to define a new variable q_i by

$$t_i = \exp(q_i) \tag{4}$$

and to note that $t_i^{n_3}$ represents a progressive plane wave inside the bulk band ($-1 < \xi_i < 1$) and an exponentially decaying wave outside the bulk band ($\xi_i > 1$ or $\xi_i < -1$). This entity $t_i^{n_3}$ is for the bulk lattice the phonon eigenvector corresponding to the eigenvalue ω^2 given by equation (1).

Let us now use this bulk phonon model to calculate eigenvectors in a few layered composite materials. The index i will enable us to distinguish between the different submaterials out of which each composite material is built.

3. Phonon eigenvectors for some layered composite materials

We use now the general theory presented in the preceding paper [7] to calculate a few phonon eigenvectors. More precisely, use will be made of equations (13*a*) or (16*a*) of [7] for the phonon model described above. Knowledge of the interface response operator **A** and of the bulk reference eigenvector is sufficient to calculate the eigenvector of the composite system. We shall therefore give only these entities for each system considered. Note that the interface response operator **A** defined by equation (9*a*) of [7] can be easily obtained for the present model from equations (9), (14)–(16) and (18) of [6]. The corresponding algebra is straightforward. We shall not expand on it and shall thus present the results directly.

3.1. The semi-infinite crystal

Consider a semi-infinite solid $i = 2$ ($n_3 \leqslant 0$) with a (001) free surface at $n_3 = 0$. Within this model and without modification of the surface force constants, there is no localised surface mode. However, the bulk eigenvectors experience a phase shift due to reflection at the surface. From the unnormalised bulk eigenvector given by

$$U(n_3) = t_2^{n_3} \tag{5}$$

corresponding to the eigenvalues given by equation (1) and the surface response operator $\mathbf{A}_{s1}$ given by equation (15) of [6] as

$$A_{s2}(on_3') = -t_2^{1-n_3'}/(t_2+1) \tag{6}$$

it is straightforward, using equation (13*a*) of [7] to obtain the unnormalised eigenvector for this semi-infinite solid to be

$$u(n_3) = \cosh[q_2(n_3 - \tfrac{1}{2})]. \tag{7}$$

Note that, inside the bulk band, q_2 is purely imaginary.

3.2. The slab

Consider a slab $i = 1$ ($1 \leqslant n_3 \leqslant L$) with (001) free surfaces at $n_3 = 1$ and $n_3 = L$. Using the corresponding surface response operator

$$A_{s1}(n_3 n_3') = [-1/(t_1+1)](\delta_{n_3 1} t_1^{n_3'} + \delta_{n_3 L} t_1^{L-n_3'+1}) \tag{8}$$

given by equations (14) of [6], the surface values of the bulk eigenvector $U(n_3) = t_1^{n_3}$ and equation (16*a*) of [7], it is also very easy to obtain the slab unnormalised eigenvector

$$u(n_3) = \cosh[q_1(n_3 - \tfrac{1}{2})] \tag{9a}$$

corresponding to the slab eigenvalues given by

$$\sinh(q_1 L) = 0. \tag{9b}$$

Note that the above expression is obtained from equation (15) of [7]. The eigenvectors of a slab within this model have been given before [8]. We recall them here as an easy check for our formalism.

3.3. The double-layer slab

Consider a double-layer slab formed out of the above slab $i = 1$ ($1 \leqslant n_3 \leqslant L$) bonded to another slab $i = 2$ ($L+1 \leqslant n_3 \leqslant N$) by first-nearest-neighbour interactions β_I between interface atoms. For this problem the interface space M is formed out of the four sites $n_3 = 1$, L, $L+1$ and N. Using equations (9), (15), (16) and (18) of [6], it is also straightforward to obtain here the interface response operator $\mathbf{A}$ defined by equation (9*a*) of [7]. Let us give here its non-zero matrix elements; we use the notation $\mathbf{A}(MD)$—this notation means that the $A(n_3 n_3')$ are such that $n_3 \in M$ and $n_3' \in D$, D being the whole space $1 \leqslant n_3 \leqslant N$.

For $1 \leqslant n_3' \leqslant L$,

$$\mathbf{A}(MD) = \begin{bmatrix} -t_1^{n_3'}/(t_1+1) \\ -\{1/(t_1+1) + (\beta_I/\beta_1)[1/(t_1^2-1)]\}t_1^{L-n_3'+1} \\ (\beta_I/\beta_1)(m_1/m_2)[t_1^{L-n_3'+1}/(t_1^2-1)] \\ 0 \end{bmatrix} \tag{10a}$$

and, for $L+1 \leqslant n_3' \leqslant N$,

$$\mathbf{A}(MD) = \begin{bmatrix} 0 \\ (\beta_I/\beta_2)(m_2/m_1)[t_2^{n_3'-L}/(t_2^2-1)] \\ -\{1/(t_2+1) + (\beta_I/\beta_1)[1/(t_2^2-1)]\}t_2^{n_3'-L} \\ -t_2^{N-n_3'+1}/(t_2+1) \end{bmatrix}. \tag{10b}$$

It is then easy to form the 4 × 4 matrix $\mathbf{\Delta}(MM) = \mathbf{I}(MM) + \mathbf{A}(MM)$. From the zero of the determinant of $\mathbf{\Delta}(MM)$, one obtains the equation giving all the eigenvalues of this double-layer slab, namely

$$\cosh[q_2(N-L+\tfrac{1}{2})]/\cosh[q_2(N-L-\tfrac{1}{2})] + \beta_I/\beta_2 - 1$$
$$= +(\beta_I^2/\beta_1\beta_2)\{\cosh[q_1(L+\tfrac{1}{2})]/\cosh[q_1(L-\tfrac{1}{2})] + \beta_I/\beta_1 - 1\}^{-1}. \tag{11}$$

Using then as the reference eigenvector

$$\langle U(M)| = [t_1, t_1^L, 0, 0] \tag{12a}$$

or

$$\langle U(M)| = [0, 0, t_2^{L+1}, t_2^N] \tag{12b}$$

and equation (16*a*) of [7], one obtains the unnormalised eigenvector of the double-layer slab.

$$u(n_3) = \begin{cases} \dfrac{-(m_1/\beta_1)\cosh[q_1(n_3-\tfrac{1}{2})]}{\sinh(q_1 L)\sinh(\tfrac{1}{2}q_1)} & 1 \leqslant n_3 \leqslant L \\[2ex] \dfrac{(m_2/\beta_2)\cosh(q_2(n_3-N-\tfrac{1}{2})]}{\sinh[q_2(N-L)]\sinh(\tfrac{1}{2}q_2)} & L+1 \leqslant n_3 \leqslant N. \end{cases} \tag{13}$$

Vibrations of a double-layer slab have been studied before [9] for the same model. However, only numerical results for the eigenvalues were presented. The closed-form results (11) and (13) presented for the first time here can also be obtained by using as reference the two uncoupled slabs. This approach was used [10] for electrons and magnons in double-layer and triple-layer slabs. The recurrent interface rescaling approach to the eigenvalue problem of finite layered composite systems [11] also gives, when applied to phonons, the same results.

3.4. *The adsorbed slab on a semi-infinite substrate*

Consider now a slab $i = 1$ $(1 \leqslant n_3 \leqslant L)$ adsorbed on a semi-infinite crystal $i = 2$ $(n_3 \leqslant 0)$. Here also the two different crystals are bound together by first-nearest-neightour interactions β_I between interface atoms. The interface space M is formed here of the three sites $n_3 = 0$, 1 and L. As above, it is easy to obtain the interface response operator $\mathbf{A}$.

For $n'_3 \leqslant 0$,

$$\mathbf{A}(MD) = \begin{bmatrix} -\{1/(t_2+1) + (\beta_I/\beta_2)[1/(t_2^2-1)]\}t_2^{1-n'_3} \\ (m_2/m_1)(\beta_I/\beta_2)[t_2^{1-n'_3}/(t_2^2-1)] \\ 0 \end{bmatrix} \tag{14a}$$

and, for $1 \leqslant n'_3 \leqslant L$,

$$\mathbf{A}(MD) = \begin{bmatrix} (m_1/m_2)(\beta_I/\beta_1)[t_1^{n'_3}/(t_1^2-1)] \\ -\{1/(t_1+1) + (\beta_I/\beta_1)[1/(t_1^2-1)]\}t_1^{n'_3} \\ -t_1^{L-n'_3+1}/(t_1+1) \end{bmatrix}. \tag{14b}$$

From the zero of the determinant of the (3×3) matrix $\mathbf{\Delta}(MM) = \mathbf{I}(MM) + \mathbf{A}(MM)$, one obtains the equation giving the phonons localised within the slab and decaying exponentially within the semi-infinite solid, namely†

$$2\{1 - (\beta_I/\beta_2)[t_2/(t_2-1)]\}\sinh(q_1 L) + (\beta_I/\beta_1)\cosh[q_1(L-\tfrac{1}{2})]/\sinh(\tfrac{1}{2}q_1) = 0 \tag{15}$$

with $q_1 \neq \mathrm{i}\pi$.

The eigenvectors corresponding to these localised phonons are then obtained from equation (16*a*) of [7], using as reference eigenvector

$$\langle U(M)| = [1, 0, 0] \tag{16a}$$

or

$$\langle U(M)| = [0, t_1, t_1^L] \tag{16b}$$

or

$$\langle U(M)| = [0, t_1^{-1}, t_1^{-L}]. \tag{16c}$$

As explained in [7], all these different reference eigenvectors of the reference system formed out of the bulk truncated and independent slabs provide the same answer for the unnormalised eigenvectors corresponding to the localised modes given by equation (15), namely

$$u(n_3) = \begin{cases} t_2^{1-n_3} & n_3 \leqslant 0 \\ 2(m_1/m_2)(\beta_2/\beta_1)[(t_2-1)/(t_1-1)] \\ \times [t_1^{L+1/2}/(1-t_1^{2L})]\cosh[q_1(n'-L-\tfrac{1}{2})] & 1 \leqslant n_3 \leqslant L. \end{cases}$$

We also calculated the eigenvectors corresponding to eigenvalues lying inside the bulk band of the semi-infinite crystal. In that case, one has, for a given eigenvalue ω^2, several possible eigenvectors corresponding, respectively, to

(i) a plane wave $U(n_3) = t_2^{n_3}$ coming from $n_3 = -\infty$ and giving rise to a reflected wave and a transmitted wave in the slab,

(ii) a plane wave $U(n_3) = t_1^{n_3}$ induced in the slab, scattered first by the free surface of the slab and then by the interface with the semi-infinite solid,

† Equation (15) has already been given as equation (41) in [6], but with a printer's error in the sign before the second term.

(iii) a plane wave $U(n_3) = t_1^{-n_3}$ induced in the slab, scattered first by the interface with the semi-infinite solid and then by the free surface of the slab and

(iv) any linear superposition of the eigenvectors obtained for the three preceding cases.

We feel that it is not necessary to give here the analytic expressions of these last eigenvectors, as any motivated reader will be able to obtain them easily.

4. Conclusion

This paper is mostly an illustration of the general and abstract theory given in the preceding paper [7]. Nevertheless, new results are presented here for phonon eigenvectors in a double-layer slab and in a slab adsorbed on a semi-infinite crystal. The simple phonon model used in these studies enabled us to perform all the calculations in closed form. This provides also a pedagogical value to this work and will enable us to understand qualitatively the initial experimental studies of phonons in adsorbed slabs. However, this work can be recalculated along the same lines for most sophisticated models, although of course with the help of a computer.

Finally let us recall that the phonon eigenvectors calculated here are isomorphic to the magnon eigenvectors within the Heisenberg model [8], used for the interpretation of spin-wave resonance spectra. Such experiments are also under way [12] for sandwich structures of an iron layer adsorbed on nickel.

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