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## Resonant guided elastic waves in an adsorbed slab: theoretical analysis of the density of states

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**Abstract.** An isolated elastic slab presents an infinite number of guided vibrational modes. Upon its adsorption on a semi-infinite substrate some of them become resonant with the bulk modes of the substrate. Such resonances were initially studied by Brillouin and by inelastic helium-atom scattering. We present here an exact method for obtaining the total vibrational density of states of the adsorbed slab. This method is then applied to isotropic elastic media and gives a semi-analytical expression for the vibrational density of states. Detailed analysis for an Al slab on a W substrate and vice versa shows that the resonant modes appear in general as well defined peaks in the total density of states. The position of these peaks enables us to study the speed of the resonant modes as a function of the thickness of the slab or of the propagation vector parallel to the surface.

### 1. Introduction

Vibrational modes of an isotropic adsorbate slab on an isotropic substrate include Love [1] and Sezawa and Kanai [2] waves. These modes [3] are respectively of shear horizontal and sagittal polarization, which means a polarization perpendicular or parallel to the sagittal plane defined by the normal to the surface and the wave vector  $k_{\parallel}$  parallel to the surface. When  $c_{t1} < c_{t2}$ , where  $c_{t1}$  and  $c_{t2}$  are the transverse velocities of sound in the adsorbate and substrate respectively, these waves emerge from the bottom of the substrate bulk bands with increasing  $k_{\parallel}$ ; their dispersion is a function of the quantity  $2k_{\parallel}a$  where  $2a$  is the thickness of the slab. Their extension into the substrate bulk bands corresponds to resonant or leaky waves which can have the character of longitudinal guided waves in the slab when their velocity lies below  $c_{l2}$  (the substrate longitudinal velocity of sound). For  $c_{t1} > c_{t2}$  all the above waves become resonant with the bulk bands of the substrate. Some of the above leaky waves, having a longitudinal character and lying below the substrate longitudinal bulk band, were first observed by Brillouin scattering in ZnSe films on GaAs [4] and in a polymeric Langmuir–Blodgett film on molybdenum [5]. These experimental results were found to be in agreement with a Brillouin scattering cross section theory [6]. Such resonant longitudinal waves were also observed by inelastic helium-atom scattering in sodium epitaxial multilayers on Cu(001); their frequencies were found to be in agreement with a simple model for a Na slab adsorbed on a rigid substrate [7].

However the total vibrational density of states associated with the resonant modes of an adsorbed slab has been studied [8] previously only for a simple atomic model having no coupling between the transverse and longitudinal waves. In the present paper we give a general expression for the calculation of the total density of states associated with an

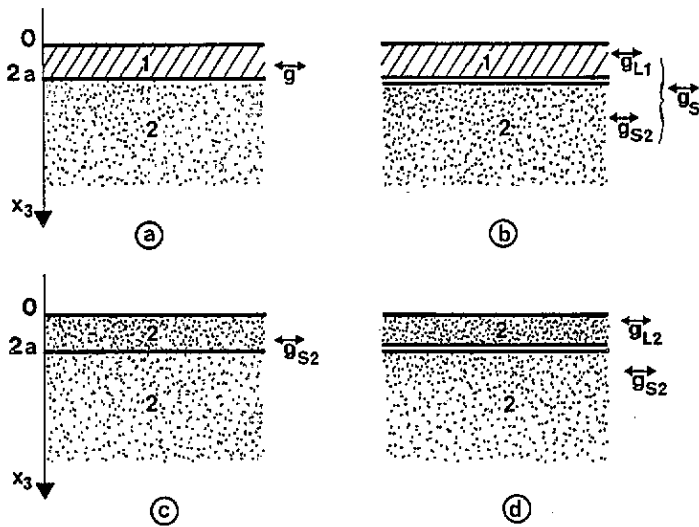


Figure 1. The adsorbed slab (a) as constructed from the reference system of (b). The corresponding Green's functions are given on the right-hand side of these figures. The semi-infinite substrate (c) and its reference system (d) as used in the demonstration given in the text, together with the corresponding Green's functions.

adsorbed slab (section 2). This expression is then used in the frame of the elasticity theory of isotropic media and provides semi-analytical expressions for the densities of states associated with the modes polarized perpendicular to the sagittal plane as well as with those polarized within the sagittal plane (section 3). Finally applications to Al slabs on a W substrate and vice versa, as well as to ZnSe slabs on a GaAs substrate, illustrate these general results (section 4).

## 2. Adsorbed slab density of states

Consider a slab of a material  $i = 1$  adsorbed on a semi-infinite substrate of a different material  $i = 2$  (figure 1(a)). The vibrational properties of such a system can be modelled either by a dynamical matrix within a lattice dynamics approach or by a differential form within elasticity theory. (For simplicity figure 1 was drawn only for the latter case.) In both cases, one can associate with this system a Green's function  $\mathbf{g}(\omega)$  where  $\omega$  is the frequency. This response function  $\mathbf{g}(\omega)$  can be constructed out of a reference Green's function  $\mathbf{g}_s(\omega)$ ; the latter can be taken to be formed from two disconnected parts, namely the Green's functions  $\mathbf{g}_{L1}(\omega^2)$  for a free slab of material 1 and  $\mathbf{g}_{S2}(\omega^2)$  for the semi-infinite substrate made of material 2 (see figure 1(b)). Several general relations exist between  $\mathbf{g}(\omega^2)$  and  $\mathbf{g}_s(\omega^2)$ . We use here the one given initially [9] for an interface between two continuous media. As this relation is in fact valid for any composite material in a discrete as well as in a continuous approach, we write it in the following matrix form:

$$\mathbf{g}(DD) = \mathbf{g}_s(DD) - \mathbf{g}_s(DM)[\mathbf{g}_s(MM)]^{-1}\mathbf{g}_s(MD) + \mathbf{g}(DM)[\mathbf{g}(MM)]^{-1}\mathbf{g}(MD) \quad (1)$$

where  $D$  stands for the total real space of the system and  $M$  for the interface space between the slab and the substrate. Within an elastic model,  $M$  is just limited to the plane  $x_3 = 2a$ , shown in figure 1.

The total density of states  $n(\omega)$  for the adsorbed slab can be obtained from the trace of the imaginary part of  $\mathbf{g}(\omega)$ . It can be related to the density of state  $n_s(\omega)$  of the reference system by

$$n(\omega) - n_s(\omega) = (1/\pi)\text{Im}(d/d\omega) \ln \det[\mathbf{g}(MM)/\mathbf{g}_s(MM)]. \quad (2)$$

A demonstration of this relation can be obtained following those given in [9] (see also, for example, [10]). However the result (2) is simpler here because we use for the reference Green's function the complete matrix  $\mathbf{g}_s$  rather than the truncated bulk Green's functions.

So in order to calculate the difference between the densities of states of the adsorbed slab and of the reference system (i.e. a free slab of material 1 and the semi-infinite substrate of material 2), we need only to know the interface elements  $\mathbf{g}(MM)$ ,  $\mathbf{g}_{L1}(MM)$  and  $\mathbf{g}_{s2}(MM)$ . We may search for the difference between the densities of states of the adsorbed slab and the substrate alone; then we have to subtract out in the numerical computations the density of states of the free slab, formed out of delta peaks situated at the slab eigenfrequencies  $\omega^2(k_{\parallel})$  for a given value for the propagation vector  $k_{\parallel}$  parallel to the surface. The drawback in such a procedure is that we compare two systems that do not have the same degree of freedom (see figures 1(a) and 1(b)). In order to improve on this, it is helpful to compare the density of states of the adsorbed slab with that of a semi-infinite substrate having its free surface at the plane  $x_3 = 0$  rather than at  $x_3 = 2a$  (figure 1(c)). This difference can be obtained by using equation (2) twice, namely once to obtain the variation of the density of states between the systems displayed in figures 1(a) and 1(b) and a second time to compare the systems shown in figures 1(c) and 1(d). Then

$$\Delta n(\omega) = n_a(\omega) - n_c(\omega) = \frac{1}{\pi} \text{Im} \frac{d}{d\omega} \ln \det \left[ \frac{\mathbf{g}(2a, 2a)\mathbf{g}_{L2}(2a, 2a)}{\mathbf{g}_{s2}(2a, 2a)\mathbf{g}_{L1}(2a, 2a)} \right] \\ + \text{density of states of slab L1} - \text{density of states of slab L2} \quad (3)$$

where L1 and L2 refer to slabs of thickness  $2a$  made respectively of materials 1 and 2.

With a slight modification of the notations for the interface space, this relation remains valid for a lattice dynamical model. Note that the densities of states of slabs L1 and L2 provide only delta peaks situated at the eigenfrequencies  $\omega(k_{\parallel})$  of the free surface slabs L1 and L2.

In the next section, we apply this general result to an elastic model for which all the Green's function elements appearing in equation (3) can be obtained in closed form.

### 3. An elastic model of the adsorbed slab

We choose now to describe the media forming the slabs and the substrate as isotropic elastic media. The parameters involved for each material are its mass density  $\rho$  and its elastic constraints  $C_{11}$  and  $C_{44}$ . The squares of the bulk longitudinal and shear plane wave velocities are respectively

$$c_l^2 = C_{11}/\rho \quad \text{and} \quad c_t^2 = C_{44}/\rho. \quad (4)$$

Such a material is also isotropic within the  $(x_1, x_2)$  plane. This enables one, for a given value for the propagation vector  $k_{\parallel}$  parallel to the surface, to decouple the shear horizontal vibrations from those polarized within the sagittal plane. The corresponding

Green's functions were derived in closed form for a semi-infinite solid [11], for an isolated slab [12–13], and for the shear horizontal component of an adsorbed slab [13].

The adsorbed slab Green's function corresponding to the modes polarized within the sagittal plane has not been calculated previously. However, in the derivation of the density of states, we only need its truncated part in the interface space, i.e.  $\mathbf{g}(2a, 2a)$ , which can be easily obtained from the following relation [10]:

$$[\mathbf{g}(2a, 2a)]^{-1} = [\mathbf{g}_{L1}(2a, 2a)]^{-1} + [\mathbf{g}_{s2}(0, 0)]^{-1}. \quad (5)$$

Using the Green's function interface elements given before [10–13], one obtains the expressions necessary in equation (3) for the density of states calculation.

In what follows we give explicit expressions for  $\mathbf{g}_s(0, 0)$  and  $\mathbf{g}_s(2a, 2a)$  for a semi-infinite substrate and for  $\mathbf{g}_L(2a, 2a)$  for a free slab. Let us recall that for all the above Green's functions the components  $g_{22}$  are decoupled from the components  $g_{11}$ ,  $g_{13}$ ,  $g_{31}$ ,  $g_{33}$ , i.e.  $g_{12} = g_{21} = g_{23} = g_{32} = 0$ . The contribution of the shear horizontal vibrations to the density of states only comes from the 22 components whereas components 11, 13, 31, 33 give the contribution to the density of states of sagittal modes.

We first define the following quantities:

$$\alpha_t^2 = k_{\parallel}^2 - \omega^2/c_t^2 \quad (6)$$

$$\alpha_l^2 = k_{\parallel}^2 - \omega^2/c_l^2 \quad (7)$$

$$\epsilon = \alpha_t \alpha_l / k_{\parallel}^2 \quad (8)$$

$$C^t = \cosh(\alpha_t a) \quad C^l = \cosh(\alpha_l a) \quad (9)$$

$$S^t = \sinh(\alpha_t a) \quad S^l = \sinh(\alpha_l a). \quad (10)$$

### 3.1. The 22 components of the Green's function elements

$$g_s(0, 0) = -(\rho \alpha_t c_t^2)^{-1} \quad (11)$$

$$g_s(2a, 2a) = -(1/2\rho \alpha_t c_t^2)(1 + e^{-4\alpha_t a}) \quad (12)$$

$$g_L(2a, 2a) = -\cosh(2\alpha_l a) / \rho \alpha_l c_l^2 \sinh(2\alpha_l a). \quad (13)$$

### 3.2. The 11, 13, 31 and 33 components of the Green's function elements

$$g_s(0, 0) = \frac{1}{\rho c_t^2 [(k_{\parallel}^2 + \alpha_t^2)^2 - 4\alpha_l \alpha_t k_{\parallel}^2]} \begin{bmatrix} \alpha_t \omega^2 / c_t^2 & ik_{\parallel} [\alpha_t^2 + k_{\parallel}^2 - 2\alpha_l \alpha_t] \\ -ik_{\parallel} [\alpha_t^2 + k_{\parallel}^2 - 2\alpha_l \alpha_t] & \alpha_l \omega^2 / c_l^2 \end{bmatrix} \quad (14)$$

$$g_L(2a, 2a) = \begin{bmatrix} m & in \\ -in & p \end{bmatrix} \quad (15)$$

where

$$m = F(\alpha_t \omega^2 / k_{\parallel} c_t^2) [(k_{\parallel}^2 + \alpha_t^2)^2 S^1 C^1 (C^{t^2} + S^{t^2}) - 4\alpha_1 \alpha_t k_{\parallel}^2 S^t C^t (C^{t^2} + S^{t^2})] \quad (16)$$

$$n = 2F\{\alpha_1 \alpha_t (k_{\parallel}^2 + \alpha_t^2) (3k_{\parallel}^2 + \alpha_t^2) (S^{t^2} C^{t^2} + C^{t^2} S^{t^2}) - C^1 S^1 S^t C^t [(k_{\parallel}^2 + \alpha_t^2)^3 + 8\epsilon^2 k_{\parallel}^6]\} \quad (17)$$

$$p = F \frac{\alpha_1 \omega^2}{k_{\parallel} c_t^2} [(k_{\parallel}^2 + \alpha_t^2)^2 S^t C^t (S^{t^2} + C^{t^2}) - 4\alpha_1 \alpha_t k_{\parallel}^2 C^1 S^1 (S^{t^2} + C^{t^2})] \quad (18)$$

$$F^{-1} = (2\rho c_t^2 / k_{\parallel}) [(k_{\parallel}^2 + \alpha_t^2)^2 C^1 S^t - 4\alpha_1 \alpha_t k_{\parallel}^2 C^t S^1] [(k_{\parallel}^2 + \alpha_t^2)^2 S^1 C^t - 4\alpha_1 \alpha_t k_{\parallel}^2 S^t C^1] \quad (19)$$

$$g_s(2a, 2a) = \frac{k_{\parallel}}{2\rho\omega^2 r_+} \begin{bmatrix} u & iv \\ -iv & w \end{bmatrix} \quad (20)$$

where

$$u = -(k_{\parallel} / \alpha_t) [(1 - \epsilon)r_+ + 2e^{-2a(\alpha_1 + \alpha_t)} + r_-(e^{-4\alpha_1 a} + \epsilon e^{-4\alpha_t a})] \quad (21)$$

$$v = e^{-2a(\alpha_1 + \alpha_t)} (1 + 1/\epsilon) + r_-(e^{-4\alpha_1 a} + e^{-4\alpha_t a}) \quad (22)$$

$$w = -(k_{\parallel} / \alpha_t) [(1 - \epsilon)r_+ + 2e^{-2a(\alpha_1 + \alpha_t)} + r_-(e^{-4\alpha_1 a} + \epsilon e^{-4\alpha_t a})] \quad (23)$$

$$r_{\pm} = [-4\alpha_1 \alpha_t k_{\parallel}^2 \pm (k_{\parallel}^2 + \alpha_t^2)^2] / 4\alpha_1 \alpha_t (k_{\parallel}^2 + \alpha_t^2). \quad (24)$$

#### 4. Applications and discussion of results

This section contains a few illustrations of the densities of states and dispersion curves for a W slab of thickness  $2a$  deposited on an Al substrate and vice versa; the parameters for these materials are listed in table 1. We shall discuss the general behaviour of these properties as well as their peculiarities for the examples under consideration. In particular, it should be noticed that due to the large difference between the elastic constants for W and Al, most of the dispersion curves in the case W/Al (respectively Al/W) are very close to those of a free slab of W (respectively a slab of Al having one surface free of stress and the other rigidly bound).

Table 1. Transverse and longitudinal velocities and mass densities for W, Al, ZnSe and GaAs.

	$c_t$ (m s <sup>-1</sup> )	$c_l$ (m s <sup>-1</sup> )	$\rho$ (kg m <sup>-3</sup> )
W	2860	5231	19 300
Al	3110	6422	2700
ZnSe	2725	4065	5264
GaAs	3342	4710	5316.9

Before discussing the results in a range of longitudinal velocities of sound, as investigated in recent work [4-5], let us first present in figure 2 the dispersion of localized and resonant modes of shear horizontal polarization for a W slab on an Al substrate; these modes are obtained from the peaks of the density of states  $n(\omega, k_{\parallel})$ , as illustrated in figure 3 for a given value for the dimensionless wave vector  $2k_{\parallel}a$ . In figure 2, the curves below the transverse sound line of the substrate represent localized modes decaying exponentially into the substrate and appearing as delta peaks in the density of states. In the limit of  $k_{\parallel}a \rightarrow \infty$ , they asymptotically tend to the limit of  $c_t(W)$ . On the other hand, their extension into the substrate bands represents resonant states (or leaky waves) whose lifetimes are related to the finite widths of the peaks in the density of states. Figure 3 shows that, for a given value for  $k_{\parallel}a$ , these peaks become wider and their intensities decrease when the frequency increases.

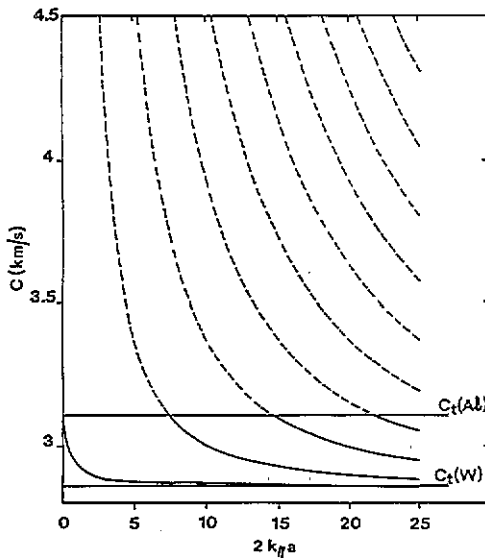


Figure 2. The dispersion of localized and resonant transverse modes for a W slab on an Al substrate. Full and broken curves respectively represent localized and resonant modes.  $c$  is the velocity.

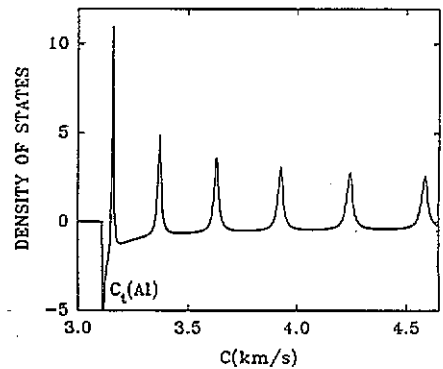


Figure 3. The variation of the density of states of transverse modes (in units of  $2a/c_t(\text{Al})$ ) between an adsorbed slab of W on an Al substrate and the same amount of a semi-infinite crystal of Al. The figure is sketched for  $2k_{\parallel}a = 20$ .

As pointed out above, the dispersion curves in figure 2 are very close to those obtained for a free slab of W; in the same way the results for an Al slab on W (not shown here) are similar to those for an isolated Al slab with one surface free of stress and the other rigidly bound. These are two particularly extreme situations which are not realized in general for arbitrary parameters of the constituents. However they may be satisfactorily reproduced for the parts of the dispersion curves which are far from the substrate sound line, this means at frequencies which are significantly lower or higher than the bottom of the substrate bands. As a particular example let us notice that for  $k_{\parallel}a \equiv 0$ , the positions of the resonant states are exactly given by  $\sin(2\omega a/c_t) = 0$  or  $\cos(2\omega a/c_t) = 0$ , i.e. the dispersion curves of an isolated slab in the two above-mentioned limits, depending on whether  $c_t < c_b$  or  $c_b < c_t$ . This demonstration at  $k_{\parallel} = 0$  is also valid for pure longitudinal waves

propagating perpendicular to the surface, provided the transverse velocities are replaced by the longitudinal ones. Such an analysis was successfully used [7] to explain the positions of the resonant longitudinal waves in Na/Cu(001) multilayers observed by helium scattering.

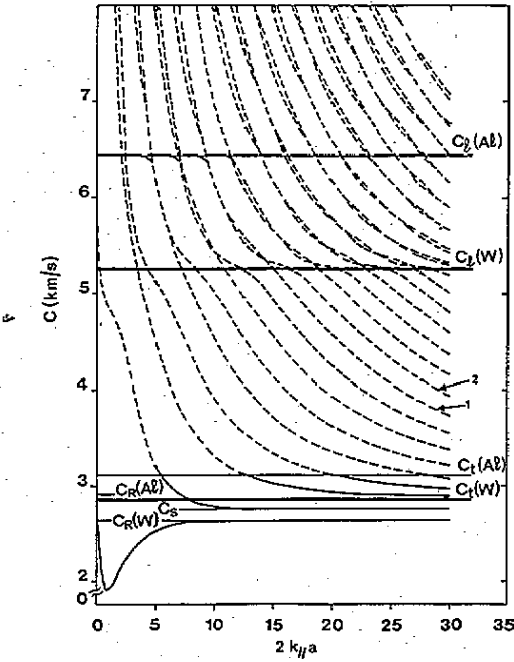


Figure 4. Dispersion curves for localized and resonant sagittal waves, for a W slab deposited on an Al substrate. The localized modes (full curves) below  $c_1(\text{Al})$  extend as resonances (broken curves) into the bulk bands of the substrate. The asymptotic limits of the lowest two branches are respectively  $c_R(\text{W})$  (the Rayleigh wave velocity of W) and  $c_s$  (the velocity of the Stoneley wave at the Al-W interface).

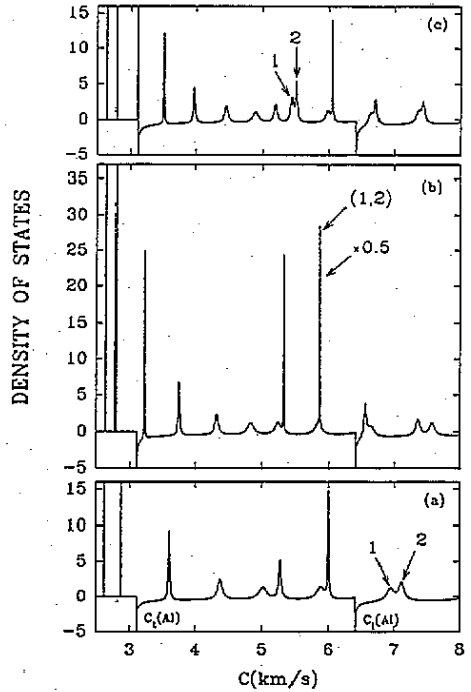


Figure 5. The variation of the density of states of sagittal waves (in units of  $2a/c_1(\text{Al})$ ) between an adsorbed slab of W on an Al substrate and the same amount of a semi-infinite crystal of Al. The Sezawa and Kanai modes localized within the slab of W give rise to delta peaks represented below  $c_1(\text{Al})$ . The figures are sketched for  $2k_{\parallel}a = 8$  (a), 11 (b) and 13 (c). The evolution of two resonances, labelled 1 and 2 in figure 4, is emphasized.

Figure 4 presents the dispersion curves for sagittal waves in the case W/Al; the behaviour of the density of states is illustrated in figure 5 for a few values for  $k_{\parallel}$ . In the limit  $k_{\parallel}a \rightarrow \infty$  in figure 4, the lowest two branches respectively move to the velocities of the Rayleigh wave on a W crystal of the Stoneley wave at the Al-W interface; the next branches move to the W transverse sound line. An analysis of the partial densities of states shows a gradual change of the predominant character of these waves from transverse to longitudinal when the frequency (or the velocity  $c$ ) increases. In this figure, one can also notice an important interaction in the vicinity of the W longitudinal sound line.

A peculiar behaviour in figure 4 is the arrangement of resonant states as a set of doublets in the range of longitudinal velocities and above. This is also the result observed for a free slab of W, where the two curves in a doublet respectively have symmetric and asymmetric character. Due to their finite width, the resonant states in figure 4 may mix together, displaying the apparent crossings of the two curves in a doublet (see also figure 5, showing



the evolution of the density of states with the wave vector  $k_{\parallel}$ ). These behaviours are not necessarily encountered with arbitrary parameters for the materials.

For a given wave vector  $k_{\parallel}$  one can observe in the density of states, sketched in figure 5 that the intensities of the peaks associated with the resonant states are first decreasing before increasing very significantly in the range of longitudinal velocities. In the latter region, the most prominent resonances have longitudinal character when their velocity  $c$  falls between  $c_1(W)$  and  $c_1(Al)$ ; their major component is attenuated inside the substrate and therefore these resonances correspond to guided longitudinal waves, as first pointed out in [4-5]; however well defined resonances also exist for velocities just above  $c_1(Al)$  where the waves have propagative behaviour in the substrate (see figure 5). A better insight into the variations of the resonance intensity all along the dispersion curve is given in figure 6; the maximum intensity, accompanied by a narrowing of the peaks, is found when the two resonant states in a doublet mix together; the height of the peaks in the density of states may therefore present a noticeable variation when changing  $k_{\parallel}$  (or  $c$ ) by a small amount.

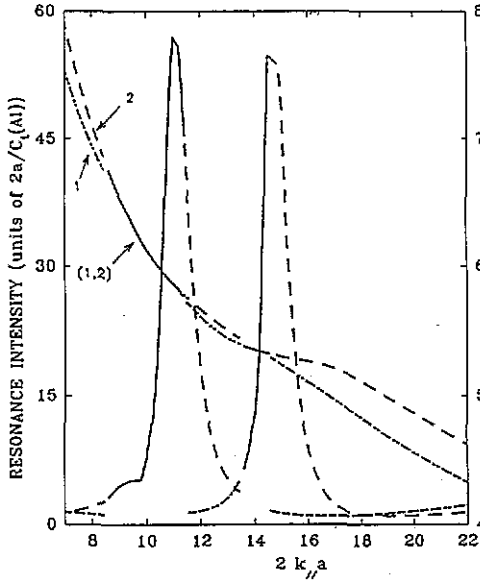


Figure 6. The intensities of the resonances, labelled 1 and 2 in figures 4 and 5, along the dispersion curves. The intensity becomes very important when the two resonances mix together. To emphasize this behaviour, the dispersion curves are also reproduced in this figure.

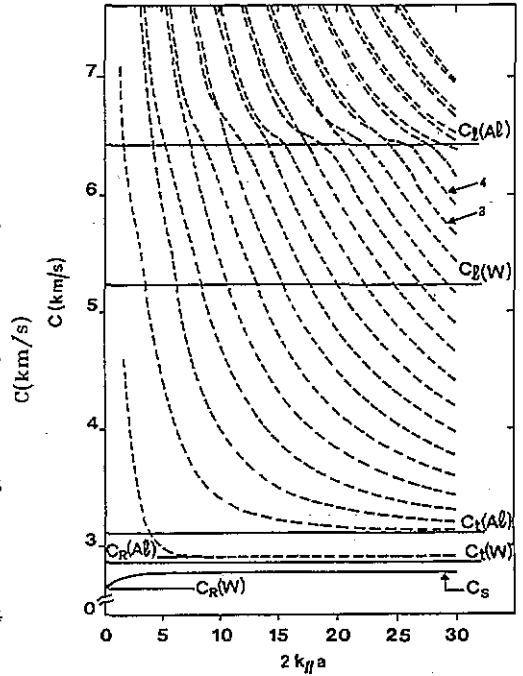


Figure 7. The same as in figure 4 for the case of an Al slab deposited on a W substrate.

In figure 7 we present the dispersion curves for the Al/W system; these curves, except for the lowest two branches, are very close to those for an isolated Al slab with one surface free of stress and the other rigidly bound. The doublet character of the branches happens for  $c > c_1(Al)$ . The narrowest longitudinal resonances occur also in this range (see figure 8), especially just above  $c_1(Al)$ , even though their intensities are in general smaller than those corresponding to the W/Al case.

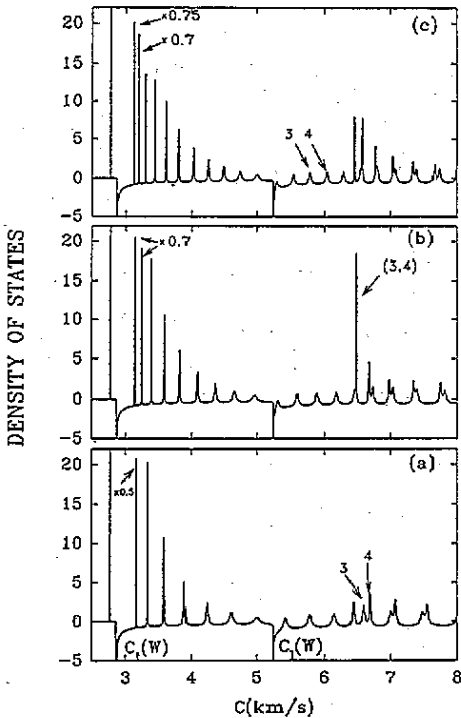


Figure 8. The same as in figure 5 for the case Al/W. The figures are sketched for  $2k_1a = 19$  (a), 24.3 (b) and 29 (c).

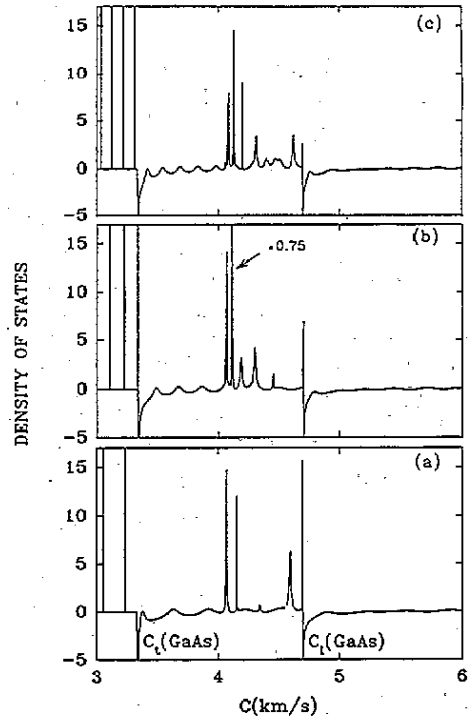


Figure 9. The same as in figures 5 and 8 for the case ZnSe/GaAs. The figures are sketched for  $2k_1a = 20$  (a), 30 (b) and 40 (c).

Finally, as a matter of comparison, we have also studied here the case of a ZnSe slab deposited on a GaAs substrate. This system has been investigated previously [4-5], both experimentally and theoretically, and its dispersion curves are given in [5]. In figure 9, displaying the density of states of sagittal waves in this case, one can notice that, in contrast to figures 5 and 8, intense resonances only occur in the velocity range limited by the longitudinal velocities of the two constituents. Also the doublet character of the dispersion curves discussed before is not encountered in this case.

## 5. Conclusions

In this paper we have presented a simple and general expression to calculate the total density of states of an adsorbed slab. Of course, the Green's function approach used in this analysis also enables us to obtain the local densities of states, even though we do not emphasize this aspect here. We applied this general method to the calculation of the vibrational density of states in isotropic elastic media. Intense resonances may be found both as extensions of Love and Sezawa and Kanai waves into the bulk bands of the substrate as well as in the range of longitudinal velocities of sound (especially in the case of guided longitudinal waves first predicted in [4-5]); however the relative importance of the peaks in the density of states is very dependent on the elastic parameters of the constituents.

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