

## Non Destructive Tests of Structural Bonds by Guided Ultrasonic Waves: Effect of a Surface Pretreatment or a Localized Defect

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**Abstract:** The aim of this study was to propose a non destructive method to test the mechanical quality of a bond aluminium/epoxy/aluminium. Thus, for each aluminium/epoxy interface, an intermediate interface modelling between a simple geometrical interface and an interphase (3D material medium), i.e. a 2D material medium. In order to describe the surface stresses, we introduce the surface free energy (the surface tension). This parameter appears in the jump conditions of stresses and displacements at each aluminium/epoxy interface. So, it influences the ultrasonic waves propagation and particularly the guided modes of the tri-layer structure. The results show that the surface free energy does not arise at normal angle of propagation, whereas near the Rayleigh angle, the behaviour of the  $A_0$  and  $S_0$  Lamb modes varies strongly according to its value. Such a modelling seems well adapted to take into account the pre-treatments of the aluminium plates or some localized interface defects.

**Key words:** Ultrasonic Waves, Adhesion, Interface Model, Surface Free Energy

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### INTRODUCTION

The use of adhesives is of great interest in industry because they allow to assemble different metal parts without having to bring them up at very high temperatures. Joining does not have the disadvantages of the welding, which modifies the mechanical characteristics of the materials, nor those of the riveting, which requires to bore the two parts before joining them together.

It is well-known that, in order to obtain a better adhesion, metallic plates must be treated before joining. The principal surface treatments are: mechanical treatments (sanding, abrasion, ultrasounds), chemical treatments (cleaning with solvent, alkaline cleaning, primer), physico-chemical treatments (plasma, the exposure to the ultraviolet rays).

There has been extensive research on the theoretical and experimental characterization of adhesively bonded structures [1-8], but there is no information about taking into account the pretreatments upon the guided waves. Hence, the interface conditions usually used are the perfect contact conditions, the slip conditions and the rheological conditions with or without inertia [9-16]. In the appendix we recall, for each one of them, their writing in term of stresses and displacements.

From the literature [17-19], it seems that the pretreatments increase the surface free energy of the metallic plates. Then, the aim of this study was to introduce this parameter into the modelling. So, each aluminium/epoxy interface is described by an intermediate interface modelling between a simple geometrical interface and an interphase (3D material

medium), i.e. a 2D material medium. In 1949, Shuttleworth proposed an extension of the surface tension of solids which depends on the surface free energy and the surface deformation. In 1985, Felder developed a thermodynamic theory which describes the surface interactions between two solids media. This allows to obtain the jump conditions of stresses and displacements at each aluminium/epoxy interface. These conditions, the so-called Felder's jump conditions, depend on the surface free energy. Consequently, the ultrasonic waves propagation and particularly the guided modes of the tri-layer structure are influenced. The results show that the surface free energy does not arise at normal angle of propagation, whereas near the Rayleigh angle, the behaviour of the  $A_0$  and  $S_0$  Lamb modes varies strongly according to its value. The reversal which appears is compared to the asymptotic behaviour of the Rayleigh wave at the aluminium/vacuum interface and for the Felder's jump conditions. By extension, all that involves a stress concentration near the bonded interfaces (micro-defects, cracks) must be able to be taken into account by this type of modelling.

### FUNDAMENTAL CONCEPTS

In this study, we will use the surface properties of the solids in contact and, for this reason, we will make some recalls.

**Laplace's Law (1806): Mechanical Equilibrium of a Liquid Surface:** Let us consider a point P, belonging to any liquid surface S represented in Fig. 1. Two

orthogonal planes containing the normal line to the surface at point P define two arcs AB and CD, whose the principal radii of curvature are  $R_1$  and  $R_2$ , respectively.

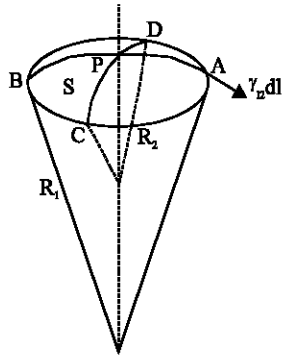


Fig. 1: Mechanical Equilibrium of an Arbitrary Curved Surface

The difference of pressure across the surface defined by  $\Delta P = P_i - P_e$  is written:

$$\Delta P = \gamma_{12} \left( \frac{1}{R_1} + \frac{1}{R_2} \right), \quad (1)$$

where,  $P_i$  and  $P_e$  are the interior and the external pressure and  $\gamma_{12}$  is the surface tension.

**Young's Formula (1805): Mechanical Equilibrium of a Liquid Drop on a Solid Surface:** A liquid drop, placed on the surface of a solid, can exhibit two different behaviours: (i) the drop may spread out completely over the solid or (ii) the drop conserves its form in an equilibrium configuration. In the second case, the tangent to the liquid surface forms with the plane of the solid an angle  $\theta$ , different from zero and called the "contact angle" (Fig. 2).

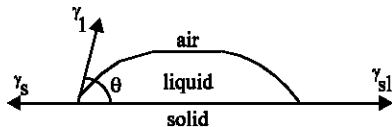


Fig. 2: A Liquid Drop on a Solid Surface

The relation between the contact angle and the intermolecular forces is written:

$$\gamma_s - \gamma_{sl} = \gamma_l \cos \theta \quad (2)$$

where,  $\gamma_s$ ,  $\gamma_l$  and  $\gamma_{sl}$  are the surface tensions for solid/air, liquid/air and solid/liquid interfaces respectively.

We remark that for a given liquid, we can increase the criterion of absorption by increasing the surface tension of the solid (pre-treatments).

**Shuttleworth's Formula (1949): Laplace's Law Extension to an Isotropic Solid Surface:** Many authors show that for a liquid, the surface tension and the surface free energy are numerically equal. On the contrary, in solids, a distinction is made between the surface free energy  $F$  and the surface tension  $\gamma$ . A thermodynamic relation between  $F$  and  $\gamma$  can be derived. So, let consider a square surface  $\Sigma$  of area  $A$  (Fig. 3).

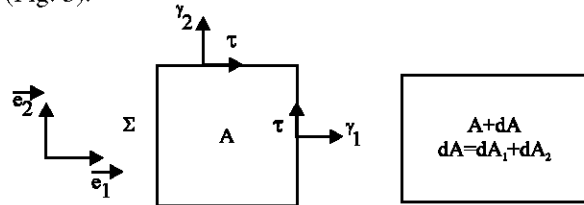


Fig. 3: Deformation and Components of the Surface Stress Tensor of a Square Surface  $\Sigma$  of Area  $A$

The work needed for varying its area, in a isothermal and reversible process, is  $d(FA) = \gamma dA$  and then,

$$\gamma = F + A \frac{dF}{dA}. \quad (3)$$

Like in a liquid medium, the atoms located at the surface of a solid are in a steady state because bonds are established between them. To describe these bonds, we introduce a symmetrical surface stress tensor  $\sigma^\Sigma$ . In

the direct orthonormal base  $(\vec{e}_1, \vec{e}_2)$ , it is defined by:

$$\sigma^\Sigma = \begin{pmatrix} \gamma_1 & \tau \\ \tau & \gamma_2 \end{pmatrix}, \quad (4)$$

where,  $\tau$  is the shear component and  $\gamma_1, \gamma_2$  are the normal components.

Then, the surface free energy variation is also equal to:

$$d(AF) = \gamma_1 dA_1 + \gamma_2 dA_2 \Rightarrow \gamma = \gamma_1 \frac{dA_1}{dA} + \gamma_2 \frac{dA_2}{dA}. \quad (5)$$

Since  $d(AF)$  does not depend upon the orientation of the square,  $dA_1 = dA_2 = dA/2$  and then  $\gamma = (\gamma_1 + \gamma_2)/2$ , i.e. half the sum of the two principal stresses, which is a mathematical invariant. The relation (3) shows that the surface tension  $\gamma$  is equal to the surface free energy plus a term depending on the relative deformation  $dA/A$  [20]. For a liquid, the surface free energy is not a function of the deformation and  $\gamma = F$ .

**Felder's Theory (1985): Thermodynamics of Surface Interactions between Two Isotropic Solid Media:** Let consider two solids noted 1 and 2. The material

domains  $\Omega_1$  and  $\Omega_2$  are in contact with each other through the zone  $\Omega_c = C_1 \cup \Sigma_c \cup C_2$  (Fig. 4). The zones  $C_1$  and  $C_2$  are composed of solid 1 and 2, respectively and  $\Sigma_c$  is the geometrical interface between the two media.

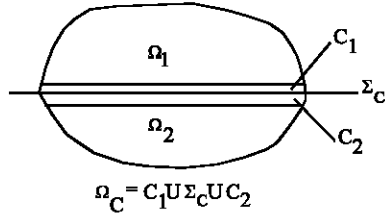


Fig. 4: Two Material Domains and their Contact Zone

We assume that (i) there is no flow of mass between  $C_1$  and  $C_2$  through  $\Sigma_c$  and (ii) the thickness of  $C_1$  and  $C_2$ ,  $h_1$  and  $h_2$ , tend towards zero. Consequently,  $\Omega_c$  can be replaced by a material interface  $\Sigma$ , i.e. a 2D continuum medium. Then, on  $\Sigma$ , any mechanical parameter  $\xi$  is defined by:

$$\xi^\Sigma = \sum_{i=1}^2 \xi_i h_i = \sum_{i=1}^2 \xi^{\Sigma_i}, \quad (6)$$

where,  $\xi_i$  is the value of  $\xi$  on  $\partial\Omega_i \cap \Sigma_c$ .

Following the way used to build the constitutive equations of continuous media, Felder apply the two principles of thermodynamics to this interface. The application of the first principle establishes the local mechanical equations of the interface and defines a generalized surface stress tensor. The application of the second principle, for a reversible thermodynamic process, gives the behaviour law of the interface  $\Sigma$ . During the calculations, Felder makes three assumptions: (a) the external surface efforts are null, (b) the mass of the interface could be neglected and consequently the surface free energy  $F^{\Sigma_i}$ ,  $i=1,2$  only depends on the deformation, (b) the strain of the interface are elastic and sufficiently small to assume that  $\partial u_z / \partial x = 0$  (but  $\partial^2 u_z / \partial x^2 \neq 0$ ). The mathematical details of these calculations are presented in Felder [21].

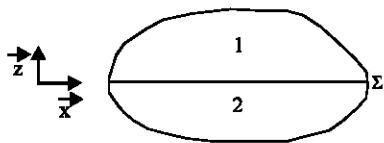


Fig. 5: The Problem Geometry

The conservation of the momentum at the interface  $\Sigma$  gives the stress and displacement jump conditions at the  $z=0$  interface:

$$\begin{cases} \sigma_{xz}^2 - \sigma_{xz}^1 = -\frac{\partial F^{\Sigma_1}}{\partial x} - \frac{\partial F^{\Sigma_2}}{\partial x}, \\ \sigma_{zz}^2 - \sigma_{zz}^1 = 2F^{\Sigma_1} \frac{\partial^2 u_z}{\partial x^2} + 2F^{\Sigma_2} \frac{\partial^2 u_z}{\partial x^2}, \end{cases} \quad (7)$$

where,  $u_z^1 = u_z^2 = u_z$  and  $u_x^1 = u_x^2$ .

If we take into account the assumptions (b) and (c), the free energy of the interface  $F^\Sigma$  is defined by:

$$F^\Sigma = \sum_{i=1}^2 F^{\Sigma_i}(\underline{\underline{\epsilon}}^{\Sigma_i})$$

where  $\underline{\underline{\epsilon}}^{\Sigma_i}$  is the 2D linearized strain tensor. Using the equation (6), the components of this tensor may be written:

$$(\underline{\underline{\epsilon}}^{\Sigma_i})_{lm} = \frac{h_i}{2} (u_{l,m}^i + u_{m,l}^i).$$

Then, the interface conditions (7) are given by:

$$\begin{cases} \sigma_{xz}^2 - \sigma_{xz}^1 = -\frac{\partial F^{\Sigma_1}}{\partial \underline{\underline{\epsilon}}^{\Sigma_1}} \frac{\partial \underline{\underline{\epsilon}}^{\Sigma_1}}{\partial x} - \frac{\partial F^{\Sigma_2}}{\partial \underline{\underline{\epsilon}}^{\Sigma_2}} \frac{\partial \underline{\underline{\epsilon}}^{\Sigma_2}}{\partial x}, \\ \sigma_{zz}^2 - \sigma_{zz}^1 = 2F^\Sigma \frac{\partial^2 u_z}{\partial x^2}, \\ u_z^2 = u_z^1 = u_z, \\ u_x^2 - u_x^1 = 0, \end{cases} \quad (8)$$

where,  $\frac{\partial F^{\Sigma_i}}{\partial x} = \frac{\partial F^{\Sigma_i}}{\partial \underline{\underline{\epsilon}}^{\Sigma_i}} \frac{\partial \underline{\underline{\epsilon}}^{\Sigma_i}}{\partial x}$ ,  $i=1, 2$ .

In the applications, the term  $\frac{\partial F^{\Sigma_i}}{\partial \underline{\underline{\epsilon}}^{\Sigma_i}}$  which corresponds to the term  $A \frac{dF}{dA}$  of the Shuttleworth's equation is always smaller than  $F^{\Sigma_i}$  so, it can be neglected. Then, the equations (8) are simplified:

$$\begin{cases} \sigma_{xz}^2 - \sigma_{xz}^1 = 0, \\ \sigma_{zz}^2 - \sigma_{zz}^1 = 2F^\Sigma \frac{\partial^2 u_z}{\partial x^2}, \\ u_z^2 = u_z^1 = u_z, \\ u_x^2 - u_x^1 = 0, \end{cases} \quad (9)$$

So, the shear stresses and the displacement vector at the interface are continue, whereas the jump of the normal stresses depend on the free energy of the interface. This

equation is the generalization of Laplace law (1),  $\frac{\partial^2 u_z}{\partial x^2}$  corresponding to the curvature radius of the interface.

**EFFECT OF THE SURFACE FREE ENERGY ON THE GUIDED MODES OF THE ALUMINIUM JOINT**

We consider a symmetric tri-layer structure composed of two isotropic and homogeneous aluminium plates ( $S_1$  and  $S_3$ ) joined by an adhesive ( $S_2$ ). For the metallic plates, we denote the thickness  $2h$ , the density  $\rho_1$ , the longitudinal waves velocity  $c_{L1}$  and the transversal waves velocity  $c_{T1}$ . The density of the adhesive layer is  $\rho_2$ , its thickness is  $d$  and the corresponding longitudinal and transversal waves velocities are  $c_{L2}$  and  $c_{T2}$  (Fig. 6). Each aluminium/adhesive interface ( $z=\pm d/2$ ) is described by Fel'der's theory.

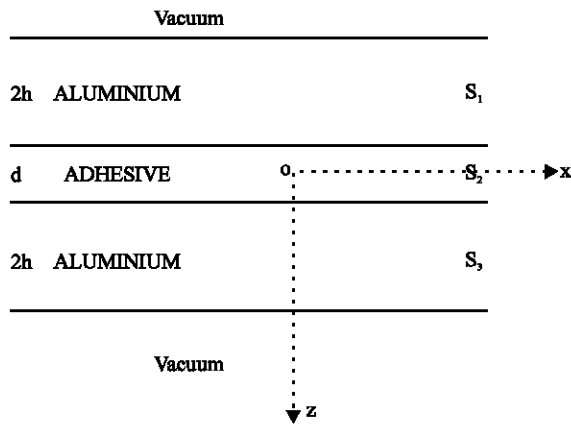


Fig. 6: Geometry of the Tri-layer

In plane deformations, the scalar and vectorial potentials are defined by:

$$\begin{cases} \phi_1 = [A_{1L} \cos(k_{Lz1}(z+d/2)) + B_{1L} \sin(k_{Lz1}(z+d/2))] e^{i(k_x x - \omega t)} \\ \psi_1 = [A_{1T} \cos(k_{Tz1}(z+d/2)) + B_{1T} \sin(k_{Tz1}(z+d/2))] e^{i(k_x x - \omega t)} \end{cases} \quad (10)$$

for the  $S_1$  -layer,

$$\begin{cases} \phi_2 = [A_{2L} \cos(k_{Lz2} z) + B_{2L} \sin(k_{Lz2} z)] e^{i(k_x x - \omega t)} \\ \psi_2 = [A_{2T} \cos(k_{Tz2} z) + B_{2T} \sin(k_{Tz2} z)] e^{i(k_x x - \omega t)} \end{cases} \quad (11)$$

for the  $S_2$  -layer, and

$$\begin{cases} \phi_3 = [A_{3L} \cos(k_{Lz1}(z-d/2)) + B_{3L} \sin(k_{Lz1}(z-d/2))] e^{i(k_x x - \omega t)} \\ \psi_3 = [A_{3T} \cos(k_{Tz1}(z-d/2)) + B_{3T} \sin(k_{Tz1}(z-d/2))] e^{i(k_x x - \omega t)} \end{cases} \quad (12)$$

for the  $S_3$  -layer.

We introduce the notations:  $k_{Lz_i}, k_{Tz_i}, i=1,2$  which are the  $z$ -components of the wave vectors for the longitudinal and transversal waves in the layers,  $k_x$  the common  $x$ -component of the wave vectors and  $\omega$  the angular frequency.

The dispersion equation of the guided modes into the structure is classically found by writing the boundary conditions:

- \* the free surface conditions at the interfaces  $z=\pm(2h+d/2)$ ,
- \* the conditions established previously (eq. 9) at the interfaces  $z=\pm d/2$ .

This leads to a 12x12 system; the roots of its determinant give the guided modes of the tri-layer. In fact, we remark that the 12x12 matrix can be written as  $A+FB$ , where  $A$  and  $B$  are two 12x12 matrix which do not depend on  $F$ . Then, the determinant of  $A+FB$  can be written as a 3 degree polynomial in  $F$ :

$$\det(A+FB) = D_0 + F D_1 + F^2 D_2 + F^3 D_3, \quad (13)$$

where,  $D_0 = \det(A)$ ,  $D_3 = \det(B)$ ,  $D_1$  and  $D_2$  are function of  $A$  and  $B$ . If  $F=0$ , we obtain the case of the tri-layer with perfect contact conditions [22]. Each term  $D_i, i=0,1,2,3$  depends on the variables  $\omega$  and  $k_x$ , which are replaced by the dimensionless quantities  $\bar{\omega} = 2k_e h, \bar{k}_x = k_x/k_e$ . Moreover, we introduce the parameter  $\bar{F} = F/(2h\mu_1)$  which compares the surface stresses and the shear bulk effects. This dimensionless parameter is smaller than the unity and the terms of the development (13) decrease.

In the Fig. 7-9, we present the dispersion curves obtained for an aluminium /epoxy /aluminium tri-layer with  $c_{L1}=6380\text{ m/s}$ ,  $c_{T1}=3100\text{ m/s}$ ,  $\rho_1=2800\text{ kg/m}^3$ ,  $c_{L2}=2662\text{ m/s}$ ,  $c_{T2}=1356\text{ m/s}$ ,  $\rho_2=1160\text{ kg/m}^3$ ,  $d/2h=10^{-1}$  and for different values of  $\bar{F}$ . The curves plotted in triangles correspond to  $\bar{F}=0$  (perfectly bonded tri-layer), whereas the curves plotted in circles correspond to  $\bar{F} \neq 0$ .

It is observed (Fig. 7-9) that the modes are sensitive to the variations of the surface free energy only for oblique angles of propagation ( $\bar{k}_x = k_x/k_e \neq 0$ ).

For  $\bar{k}_x=0$ , the term  $\frac{\partial^2 u_z}{\partial x^2}$  is equal to zero because no

effect of curvature can be observed for normal propagation. Then, the cutoff-frequencies of guided modes are the same as for  $\bar{F}=0$ .

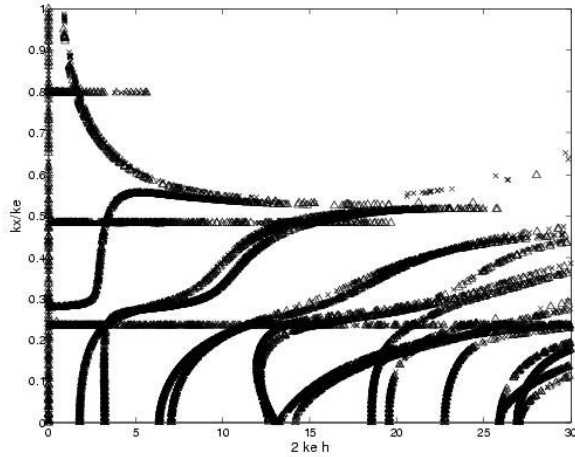


Fig. 7: Dispersion Curves Comparison ( $\bar{F}=10^{-2}$ )

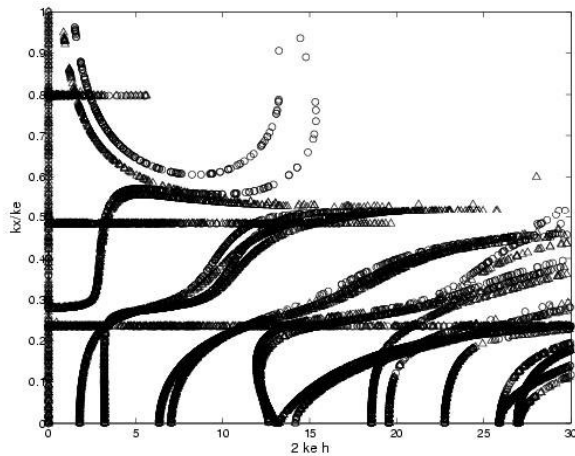


Fig. 8: Dispersion Curves Comparison ( $\bar{F}=5 \cdot 10^{-2}$ )

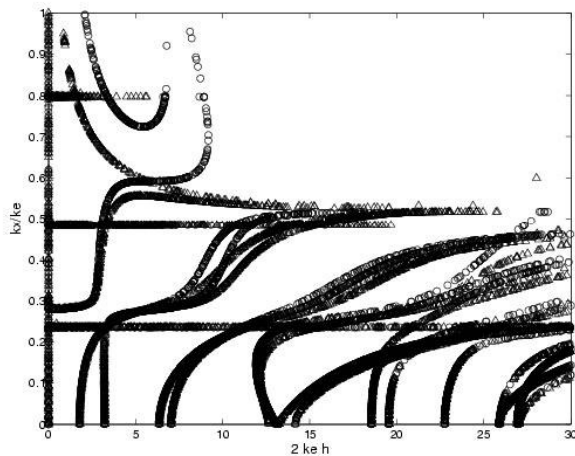


Fig. 9: Dispersion Curves Comparison ( $\bar{F}=10^{-1}$ )

For  $\bar{k}_x \neq 0$ , the effect of the surface free energy is more significant when  $\bar{F}$  increases.

The  $A_0$  and  $S_0$  Lamb modes are particularly sensitive. On Fig. 8, we observe that the asymptotic Rayleigh

wave limit (for an interface aluminium/vacuum and free surface conditions) does not appear for  $\bar{F} \neq 0$ . These modes undergo a “reversal”, the corresponding frequencies at the “reversal” point vary according to  $\bar{F}$ . In order to explain this behaviour, we studied the Rayleigh wave which appears at the aluminium/vacuum interface with Felder's jump conditions. In this case, the Rayleigh equation is given by Biryukov [23]:

$$4 \bar{k}_x^2 \bar{k}_{Lz1} \bar{k}_{Tz1} + (n_{T1}^2 - 2 \bar{k}_x^2)^2 + 2 \bar{\omega} \bar{F} \bar{k}_x^2 n_{T1}^2 \sqrt{\bar{k}_x^2 - n_{L1}^2} = 0, \quad (14)$$

where,

$$\bar{k}_{Lz1}^2 = n_{L1}^2 - \bar{k}_x^2, \bar{k}_{Tz1}^2 = n_{T1}^2 - \bar{k}_x^2, n_{L1} = c_e/c_{Lb}, n_{T1} = c_e/c_{T1}, \text{ and } c_e \text{ is the wave velocity in water.}$$

This equation is the sum of the Rayleigh equation for an aluminium/vacuum interface with  $\bar{F}=0$  and a term proportional to  $\bar{F}$  and  $\bar{\omega}$ . Thus, when we take into account the surface free energy, the Rayleigh equation becomes a dispersive equation. This is explained by the introduction of a characteristic length of reference (one of the thicknesses  $h_i$  introduced in Felder's theory §2.4). The numerical solutions of the equation (14) for different values of  $\bar{F}$  are given on Fig. 10. We observe that the Rayleigh wave undergoes a “reversal” like the  $A_0$  and  $S_0$  modes.

If we report the curves (a), (b), (c) on the Fig. 7, 8 and 9, respectively, we observe a good agreement between the asymptotic behaviour of the  $A_0$  and  $S_0$  modes and the Rayleigh wave (for the aluminium/vacuum interface with Felder's jump conditions).

The results presented above show that it is theoretically possible to evaluate the surface tension  $\bar{F}$  - let us recall however that its value is representative of the bond quality - but what does happen applications ?

Considering an aluminium/epoxy/aluminium tri-layer where the thicknesses of the two materials are  $2h=3mm$  and  $d=300\mu m$ , i.e.  $d/2h=10^{-1}$ . After a standard pre-treatment, the average value of the thickness of the resulting zone is on the order of  $25\mu m$ , which corresponds to  $F=7 \cdot 10^6 N/m$ , i.e.  $\bar{F} \approx 10^{-2}$ . So, the dispersion curves of guided modes correspond to the Figure 7. The influence of the surface tension on the asymptotic limit of the  $A_0$  and  $S_0$  modes becomes sensitive near  $\bar{\omega}=23$ , i.e. the wavelengths in the structure vary between  $60\mu m$  and  $200\mu m$  for an average frequency about  $2 MHz$ . The value of the wavelengths agrees with the modelling (both aluminium and epoxy layers are 3D media and the pretreated zone is a 2D medium).

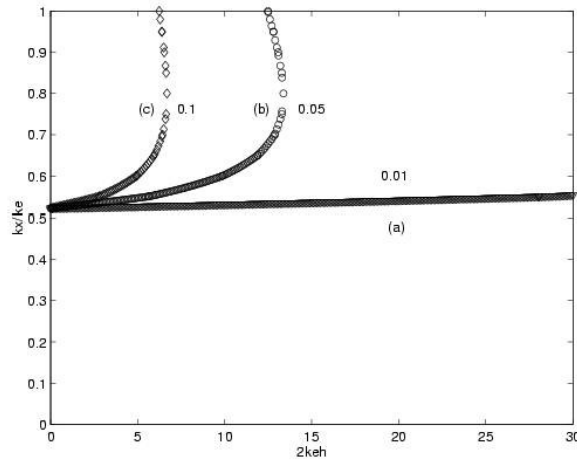


Fig. 10: Rayleigh Wave for an Aluminium/vacuum Interface with Felder's Jump Conditions for Different Values of  $\bar{F}$

### CONCLUSION

In this paper we studied, by the mean of ultrasonic waves, an aluminium/adhesive/aluminium structure; the adhesion zone being described by a 2D material interface. This yields to introduce the "surface free energy" parameter. This parameter appears in the jump conditions of the problem, the so-called Felder's jump conditions. Its influence on the acoustic guided modes of the aluminium joint is studied.

The dispersion equation is the sum of two terms: the first corresponds to the dispersion equation for the conditions of perfect adhesion, the second is proportional to the free energy of interface and vanishes at normal incidence. The numerical study of the dispersion curves allows to evaluate the influence of the surface free energy compared to the case of reference (aluminium/adhesive/aluminium with contact perfect conditions). The  $A_0$  and  $S_0$  Lamb modes undergo a "reversal" which can be explained by the high-frequency behaviour of the Rayleigh wave (at the aluminium/vacuum interface with Felder's jump conditions).

The Felder's jump conditions may be used to model a stress concentration near the bonded interfaces. Such a stress concentration can occur when the aluminium plates are pre-treated or when some defects, as cracks, are localized at the interfaces.

**Appendix:** Conditions of interface usually used in the literature.

1. Perfect contact conditions:

$$\begin{aligned} \llbracket u_x \rrbracket &= 0, \llbracket u_z \rrbracket = 0, \\ \llbracket \sigma_{xz} \rrbracket &= 0, \llbracket \sigma_{zz} \rrbracket = 0. \end{aligned}$$

2. Slip conditions:

$$\begin{aligned} \llbracket u_x \rrbracket &= 0, \\ \sigma_{xz}^2 = \sigma_{xz}^1 &= 0, \llbracket \sigma_{zz} \rrbracket = 0. \end{aligned}$$

3. Rheological conditions without inertia:

$$\begin{aligned} \sigma_{xz}^2 = \sigma_{xz}^1 &= K_T \llbracket u_x \rrbracket \\ \sigma_{zz}^2 = \sigma_{zz}^1 &= K_L \llbracket u_z \rrbracket \end{aligned}$$

4. Rheological conditions with inertia:

$$\begin{aligned} \sigma_{xz}^2 + \sigma_{xz}^1 &= 2K_T \llbracket u_x \rrbracket, \sigma_{zz}^2 + \sigma_{zz}^1 = 2K_L \llbracket u_z \rrbracket, \\ \llbracket \sigma_{xz} \rrbracket &= \frac{m}{2} \frac{d^2}{dt^2} (u_x^1 + u_x^2), \llbracket \sigma_{zz} \rrbracket = \frac{m}{2} \frac{d^2}{dt^2} (u_z^1 + u_z^2) \end{aligned}$$

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