Eur. Phys. J. AP 5, 221–226 (1999)

THE EUROPEAN PHYSICAL JOURNAL
APPLIED PHYSICS

Theoretical approach of the characterization of gradients in elastic properties by acoustic microscopy

H. Nounah[1], B. Cros[2,a], and J. Attal[2]

[1] LIM, Faculté des Sciences, Université Ibnou Zohr, B.P. 28/S, Agadir, Morocco
[2] LAIN, Université Montpellier II, 34095 Montpellier Cedex 05, France

Received: 25 September 1997 / Revised: 20 July 1998 / Accepted: 7 December 1998

Abstract. This study explains processes for the characterization by acoustic microscopy of materials presenting, along a thickness d close to the surface, a gradient in elastic properties in direction perpendicular to the surface. The method, modelling the non-homogeneous area by a finite number of layers of the same thickness, is sensitive to the factor fd (f is the working frequency) and to the nature of the gradient profile. The study of the dispersion of the Rayleigh velocity as a function of frequency shows that the shape of the curve mainly depends on the gradient profile and on the structure (surface material and bulk material). When the numerical derivative of the dispersion curves presents inflexion points, they can be linked to the depth and to the nature of the profile. Modelling of the materials displaying a gradient shows that, in some cases, the plot of the curvature of the dispersion curves as a function of frequency is a useful means of characterization.

Résumé. Cette étude propose une démarche de caractérisation par microscopie acoustique de matériaux présentant, sur une épaisseur d au voisinage de la surface, un gradient de propriétés élastiques dans la direction perpendiculaire à la surface. La méthode simulant la région inhomogène par un nombre fini de couches d'égale épaisseur est sensible au facteur fd (f est la fréquence) et à la nature du profil du gradient. L'étude de la dispersion de la vitesse de Rayleigh en fonction de la fréquence montre que son évolution dépend essentiellement du profil du gradient et de la structure (matériaux constituant la surface et le cœur de l'échantillon). Lorsque la dérivée numérique des courbes de dispersion présente des points d'inflexion, ces points permettent de déterminer l'épaisseur et la nature du profil. La modélisation de matériaux à gradient montre que, dans certains cas, la représentation de la courbure des courbes de dispersion des vitesses en fonction de la fréquence est un moyen efficace de caractérisation.

PACS. 43.35.Ns Acoustic properties of thin films – 81.70.Cv Nondestructive testing: ultrasonic testing, photoacoustic testing

1 Introduction

Acoustic microscopy is well-adapted for the characterization of elastic properties of thin films. The method consists in exploiting the acoustic signature [1] and calculating the velocity of the different modes which propagate in the layer (Lamb modes). When thickness and density of the layer are known, the elastic constant can be deduced [2,3]. Different cases should be considered, depending on whether the layer is isolated and immersed or deposited on a substrate. In the case of a stratified system (layer on substrate), the study of the second mode or Sezawa mode is an effective way to characterize adhesion and to solve the reverse problem for isotropic [4,5] or anisotropic [6] materials. The aim of this work is to extend the potentialities of characterization of acoustic microscopy to materials presenting a gradient of properties whose neither the gradient extent, nor the nature of the profile is known. In fact, a gradient in acoustic properties expresses a gradient in elastic properties or in chemical properties (gradients in composition). Applications concern the field of surface processing, for instance mechanical processing (lamination), thermal processing, thermochemical processing and diffusion processes. The role of these treatments is to impart to the surface of materials properties (wear resistance, thermal or chemical behaviour, aesthetic look) different from the properties of the bulk material (shock resistance, high elasticity). An interphase close to the surface (area with gradient in properties) is the result of the process.

In a previous work [7], we have developed a model to characterize, by acoustic microscopy, materials presenting a gradient in elastic properties in direction perpendicular to the surface. The method is based on the slicing of the gradient area in a finite number of layers with the same thickness (Fig. 1) in order to provide an accurate theoretical represention of the physical reality of the

[a] e-mail: cros@lain.univ-montp2

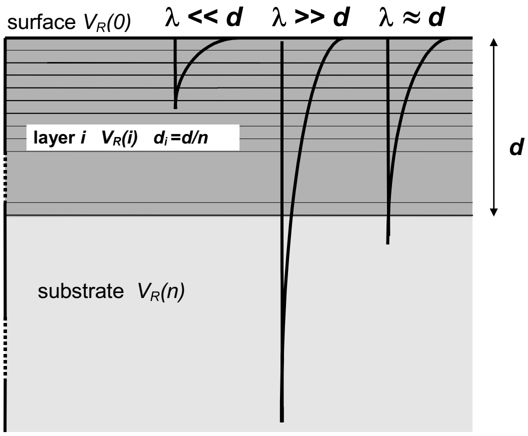

Fig. 1. Modelling of the non-homogeneous area of a material by series of layers. d is the depth of this area. The length of the arrays represent the depth of penetration of the surface waves according to their wavelength (the energy of the wave exponentially decreases in relation with the depth).

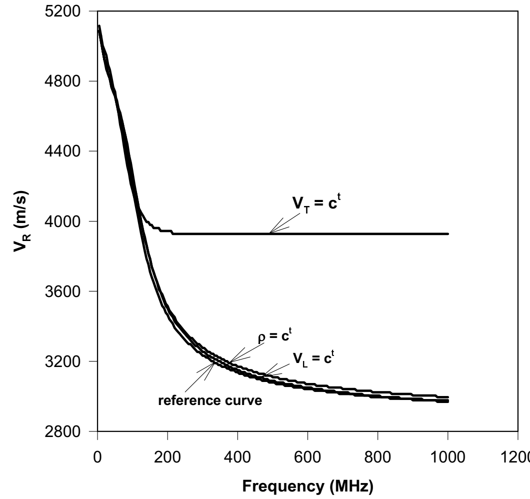

Fig. 2. Sensitivity of the velocity of the Rayleigh mode to the acoustic parameters V_L, V_T and ρ (exponential profile, $d = 20\ \mu$m).

non-homogeneous material. As the discontinuities created at each interface by the modelling lower the accuracy of the calculations, we have determined the minimal slicing to ensure both reliable results and easy calculations. A theoretical study of the dispersion of the surface waves velocity as a function of frequency or, more generally, of the factor *fd*, proves that the evolution of velocities mainly depends on the profile of variation of the acoustic parameters. The aim of the present work is to apply the theoretical model to the characterization of materials with gradient. The method consists in calculating the depth of the gradient area and the nature of the variation profile of the elastic constant from the study of the theoretical dispersion curves. This approach constitutes the reverse problem. The study is limited to the case of materials presenting a plane surface and a progressive gradient without discontinuity. In particular, the model material is assumed to be homogeneous in directions parallel to the surface. This method should be relevant in real cases and, therefore, should link the gradient in chemical or mechanical properties to a gradient in acoustic properties.

2 Approach of the reverse problem

The solution of the reverse problem from the dispersion curves has been used by other authors to calculate the elastic charasteristics and the thickness of plate materials [8]. In the direct problem, the existence of a velocity for a set frequency can be definitely determined provided that the acoustic parameters are known. To treat the reverse problem, we should invert the procedure in order to calculate, from the dispersion curves, the features of the gradient. The curves are plotted in the case of a decrease of the velocity in the gradient area from the bulk material toward the surface. The studied mode is the first Lamb mode which corresponds to the Rayleigh mode. The analysis of the curves (*cf.* Sect. 2.2) highlights two important points which are, according to the studied structure, the inflexion point or the maximum of the curvature. In this approach, we first looked the acoustic parameters $V_L(z)$, $V_T(z)$ and $\rho(z)$ (respectively the longitudinal velocity, the shear velocity and the density) of the gradient area to which acoustic microscopy is sensitive, in order to develop a method linking these parameters to the features of the gradient, mainly thickness and profile. The acoustic characteristics of the surface material and of the bulk homogeneous material ("extreme" materials) are assumed to be known. Should information on the materials bounding the gradient area be lacky, an additional experimental procedure is proposed.

2.1 Sensitivity of the velocity of the surface mode to the acoustic parameters V_L, V_T and ρ

The method of characterization of the gradient area consists in analysing the dispersion of the surface mode velocity (Rayleigh mode) V_R as a function of frequency. The elastic properties of this area will be inferred from the variation of V_R. In a preliminary study, we shall determine the sensitivity to the variations of the acoustic parameters V_L, V_T and ρ of the Rayleigh mode velocity. To study the influence, on the V_R value, of the variation of each of the acoustic parameters, we assume one of them to be constant, V_L, V_T and ρ successively, while the two others vary according to a profile of chosen type (exponential). The variation of V_R, in each case, is compared with a reference curve (Fig. 2) obtained by varying simultaneously the 3 parameters according to the same exponential type gradient. When the velocity V_T is set, for frequencies higher than 250 MHz, the corresponding curve draws aside from the two others and sharply tends towards an asymptotic value, while the two other curves remain close to the reference curve. Therefore, the value of velocity of the surface wave closely depends on the V_T value, whereas

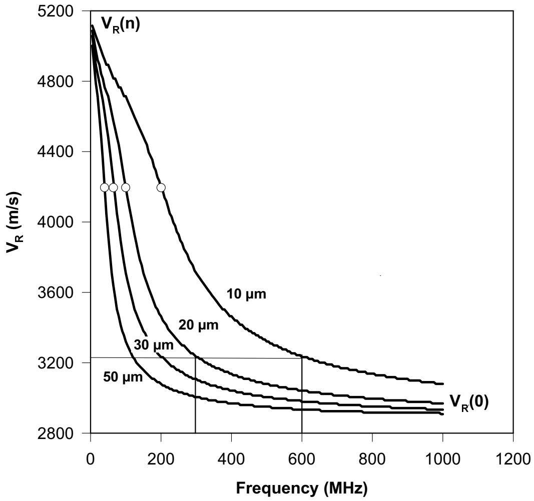

Fig. 3. Dispersion of the Rayleigh mode velocity as a function of frequency for various values of d, the thickness of the non-homogeneous area ($V_{\rm R}(0) = 2900$ m s^{-1}; $V_{\rm R}(n) = 5100$ m s^{-1}; exponential profile). The horizontal line represents a constant fd product. The inflexion points are highlighted by white circles.

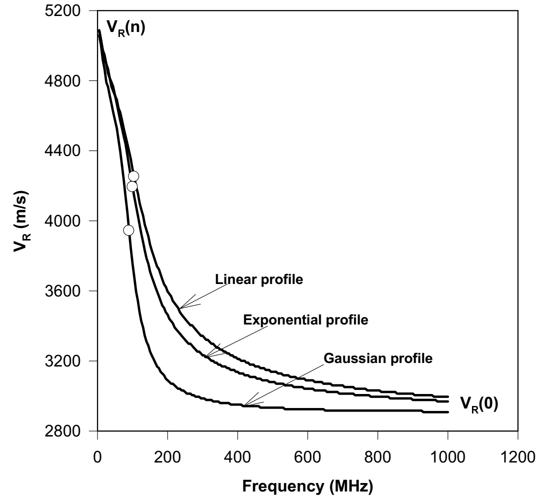

Fig. 4. Dispersion of the Rayleigh mode velocity as a function of frequency for various profiles (d = 20 μm). The inflexion points are represented by white circles.

$V_{\rm L}$ and ρ do not influence $V_{\rm R}$ much. The result is that a method sensitive to characterize the gradient should be based on the study of the dispersion of $V_{\rm T}$. These conclusions are in agreement with the results of the study of the monolayer/substrate system [5].

2.2 Study of the dispersion curves

The study of the modelling of a gradient material has led to draw up the dispersion curves of the surface mode velocity $V_{\rm R}$ as a function of frequency [7]. These curves are characterized by the existence of 3 ranges. When the thickness d of the gradient area (Fig. 1) is small with regard to λ (wavelength of the Rayleigh wave of the substrate), $V_{\rm R}$ stays almost constant (in these conditions, $V_{\rm R}$ remains non dispersive). When d is in the same order of magnitude as λ, $V_{\rm R}$ is very dispersive and presents the best sensitivity to frequency. If λ is small with regard to d, $V_{\rm R}$ is again non dispersive and, when d/λ increases, $V_{\rm R}$ becomes asymptotic to a stationary value, the velocity of the shear mode of the material close to the surface. $V_{\rm R}$ is then independent of frequency as in a bulk material.

2.2.1 Role of thickness

The study of the dispersion of the velocity of the surface mode as a function of frequency for various thicknesses d of a same kind of material (same "extreme" materials and same nature of profile) shows the role of the parameter d (Fig. 3). The velocity $V_{\rm R}$ varies from the substrate velocity $V_{\rm R}(n)$ (arbitrarily chosen to be equal to 5100 m s^{-1}) to the velocity $V_{\rm R}(0)$ of the surface material (arbitrarily chosen equal to 2900 m s^{-1}). The value $V_{\rm R}$ obtained with f = 600 MHz and d = 10 μm is identical to the value obtained with f = 300 MHz and d = 20 m. Indeed, in the calculation of the reflecting power [9], both parameters, frequency and thickness, are always linked and behave like a single variable. For a constant *fd* product, the velocity is constant (*cf.* Fig. 3). This result will be used to determine the kind of gradient from the plot of the variation of $V_{\rm R}$ as a function of the product *fd*. Moreover, the dispersion curves present a change of concavity, namely inflexion points. These points are significant elements for the solution of the reverse problem because they are characteristic of the gradient.

2.2.2 Role of the profile

The role of the profile is enlightened by Figure 4. The dispersion curves of the Rayleigh mode are plotted for 3 fictive gradient samples having the same characteristics of the bulk material and of the surface material (extreme materials), the same depth d (20 μm) of the gradient area but differing by the kind of profile. The set of curves present a common general shape. Between the extreme values $V_{\rm R}(n)$ and $V_{\rm R}(0)$, the variation in velocity mainly depends on the profiles of variation of the acoustic parameters. The changes of concavity are found again.

2.2.3 Role of the nature of materials

The role of the nature of materials is enlightened by Figure 5. The dispersion curves of the Rayleigh mode are plotted for 3 fictive gradient samples having the same kind of profile (exponential), the same depth d (20 μm) of the gradient area, the same bulk material, but differing by the characteristics of the surface material. The curves do not present changes of concavity. Whenever inflexion points were lacking, we focussed our study on the curvature of the dispersion curves as a function of frequency as a possible way to solve the reverse problem, namely to go back to the gradient width.

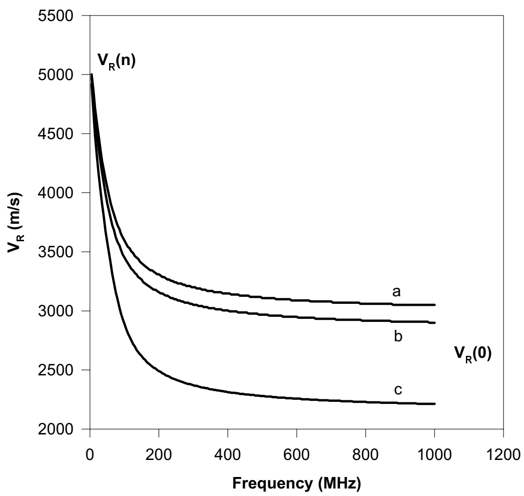

Fig. 5. Dispersion of the Rayleigh mode velocity as a function of frequency for 3 surface materials (exponential profile, $d = 20\,\mu$m, same bulk material with $V_R(n) = 5100$ m s^{-1}). (a) $V_R(0) = 3100$ m s^{-1}; (b) $V_R(0) = 2900$ m s^{-1}; (c) $V_R(0) = 2250$ m s^{-1}.

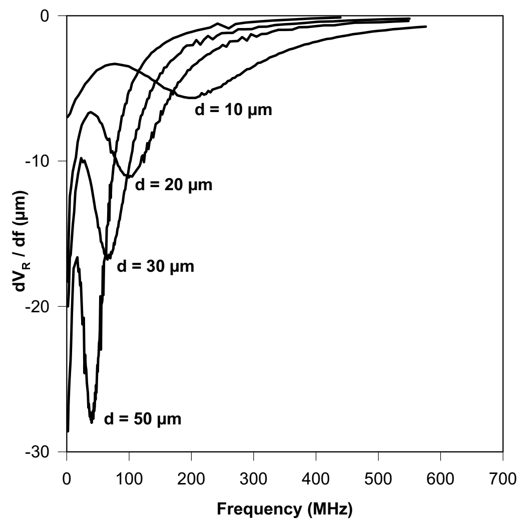

Fig. 6. Numerical derivative of the dispersion curves *versus* frequency for various thicknesses of the non-homogeneous area (common exponential profile).

Table 1. Critical points of the curves plotted in Figure 3.

Thickness (μm)	10	20	30	50
Minimum of dV_R/df (μm)	−5.66	−11.13	−16.78	−28
Location of the minimum (MHz)	199	99	65	40
Maximum of dV_R/df (μm)	−3.32	−6.64	−9.80	−16.62
Location of the maximum (MHz)	77	39	23	17

Table 2. Critical points of the curves plotted in Figure 4.

Profile	Linear	Exponential	Gaussian
Minimum of dV_R/df (μm)	−9.18	−11.18	−18.97
Location of the minimum (MHz)	103	99	89
Maximum of dV_R/df (μm)	−6.37	−6.64	−8.13
Location of the maximum (MHz)	37.2	39	35
Ratio min/max	1.44	1.68	2.33

3 Theoretical process

The study of the role of the various parameters defining the gradient area (thickness, nature of the profile and value of Rayleigh velocity of the "extreme" materials) shows that the characteristic points (either the inflexion points or the curvature maxima) are essential elements for the solution of the reverse problem.

3.1 Use of the inflexion points

We resort to the presence of the inflexion points noticed in the set of the dispersion curves (Fig. 3). The inflexion points correspond to the zero value and to the change of sign of d^2V_R/df^2 with

$$dV_R/df = [V_R(f + \Delta f) - V_R(f)]/[(f + \Delta f) - f].$$

3.1.1 Thickness characterization

Figure 6 shows the variation of the numerical derivative of the dispersion curves as a function of frequency for each considered case (various thicknesses). The values of maxima and minima of dV_R/df and their positions for the curves of Figure 3 are reported in Table 1. The negative values are due to the variation direction (decrease) of V_R as a function of f. The position of the maxima and minima (thus of the inflexion points) strongly varies with the thickness of the gradient area (Tab. 1). For the chosen example, the values of the minima and maxima roughly correspond to $d/2$ and $d/3$ respectively.

3.1.2 Profile characterization

In order to extend this approach to the role of the gradient profile, we have reported in Table 2 and Figure 7 the results and the curves achieved for 3 different profiles corresponding to a same value of d. They point out the relationship between the type of gradient profile and the location of the extrema. For the linear and exponential profiles, the location of the minimum is close to $d/2$ and the location of the maximum is close to $d/3$. For the Gaussian profile, the location of the minimum tends towards d

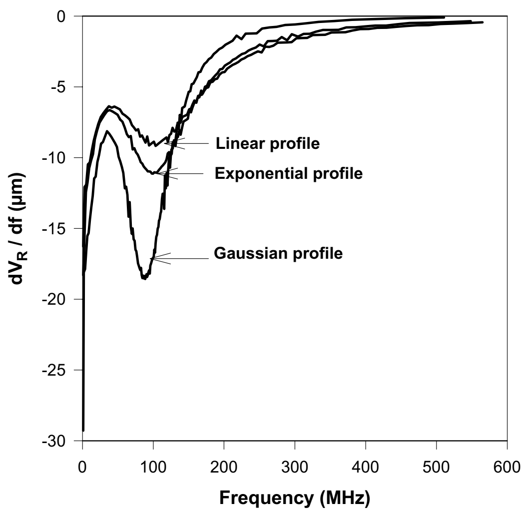

Fig. 7. Numerical derivative of the dispersion curves *versus* frequency for 3 gradient profiles ($d = 20\,\mu$m).

and the location of the maximum towards $d/2$. Table 2 shows that the various profiles can be distinguished according to the ratio min/max. Therefore, this ratio makes it possible to establish the type of gradient profile.

Thickness and nature of the gradient profile can be characterized by the study of the critical points, namely the inflexion points.

3.2 Use of the curvature

The case of dispersion curves without a change of concavity has been studied. The dispersion curves of 3 theoretical systems of gradient materials are plotted in Figure 5. The thickness d (typically 20 μm), as well as the exponential profile, are assumed to be the same. The velocity of the surface material bounding the gradient is the only parameter made to vary from one system to another.

When there is no change of concavity in the dispersion curve, the maximum of curvature is located. The curvature is defined by the function

$$C = \frac{\dfrac{\mathrm{d}^2 V_\mathrm{R}}{\mathrm{d}f^2}}{\left(1 + \left(\dfrac{\mathrm{d}V_\mathrm{R}}{\mathrm{d}f}\right)^2\right)^{\frac{3}{2}}}\,.$$

The gradient width can be recognized from the location of the curvature maximum according to the corresponding value of the derivative $\mathrm{d}V_\mathrm{R}/\mathrm{d}f$. Figure 8 shows the variation of the curvature as well as the variation of the derivative as a function of frequency for one of the dispersion curves (Fig. 5c). In each dispersion curve, the frequency corresponding to the curvature maximum can be determined. Table 3 collects these values and the corresponding values of the derivative. If the extreme materials (surface and bulk materials) are known, the gradient width is determined by the reverse operation, namely by linking it to the maximum value for $\mathrm{d}V_\mathrm{R}/\mathrm{d}f$. Indeed, Table 3 shows

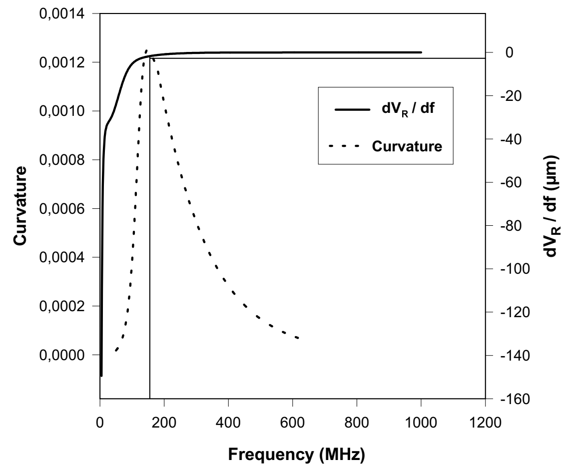

Fig. 8. Variation of the curvature and of the numerical derivative of one dispersion curve in relation with frequency (curve (c) in Fig. 5).

Table 3. Location of the maximum of curvature in the dispersion curves and corresponding derivative for the 3 pairs of materials.

$V_\mathrm{R}(0)$	2250	2900	3100
Location of the maximum of curvature (MHz)	147	121	119
$\mid \mathrm{d}V_\mathrm{R}/\mathrm{d}f \mid$ (μm)	3	3.15	3.25

that, whatever structure is studied, the value of $\mathrm{d}V_\mathrm{R}/\mathrm{d}f$ remains more or less the same, the only difference lying in the frequency shifts of the maxima. The deduced value is close to $d/6$ for the 3 considered pairs.

4 Discussion of the experimental applicability

The characterization of layers presenting a gradient in their properties is a complex problem because thickness and kind of profile should be both characterized. Generally, the "extreme" materials are known, as well as the method used to carry out the gradient (diffusion of an element in a semi-conductor material, carburization, nitriding or carbonitriding of steel, ...). So the kind of gradient is often known, the profile being typical of the process. When no information on the material to characterize is known, the sequence is as follows. It is drawn up by taking into account the sensors available in our laboratory. They work respectively at 15, 50, 130, 400, 570 MHz and 980 GHz.

The first step consist in carrying out measurements at both extreme frequencies to deduce the Rayleigh velocity of the bulk material (at 15 MHz) and of the surface material (980 GHz). Three measurements were carried out at each and every available frequency, the first one at the central value of the sensor range and the two others at 10% from this value (a sample with known Rayleigh velocity should be used as standard).

The results reveal the shape of the dispersion curve of the Rayleigh velocity (Figure 9 gives an example of

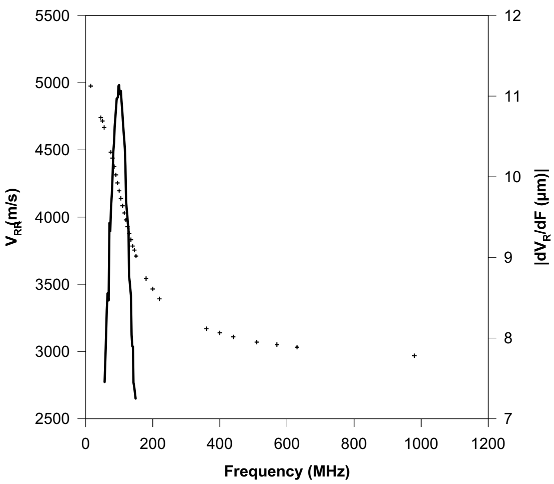

Fig. 9. Modelling (for an exponential profile and $d = 20\ \mu$m) of the results of measurements of the Rayleigh velocity in relation with frequency. The shape of the dispersion curve is revealed and is used to locate the maximum slope.

results for a thickness of the gradient area $d = 20\ \mu$m and an exponential profile). This shape is used to locate the maximum slope of the dispersion curve.

By using the sensor whose frequency is the nearest to the frequency corresponding to the maximum slope, enough measurements were carried out (fine scanning), to accurately locate the inflexion point.

In order to check the profile, the maximum and the minimum of the derivative of the dispersion curve should be determined. The method consists once again in a fine scanning by using several other sensors (typically 2).

When the inflexion point could not be determined, the method consisting in studying the curvature of the dispersion curve was resorted to. It is more awkward because it requires very fine scanning (consequently several large range sensors) allowing to calculate the second derivative.

5 Conclusion

This study, based on the modelling of a material with gradient of elastic properties, suggests processes to characterize thickness and profile of the non-homogeneous area. The method consists in plotting the dispersion curves and looking for the presence of a change of concavity. The frequency value for which the slope is maximum is a characteristic parameter of the thickness d of the gradient area. Consequently, the characterization of a gradient material and, first, of d takes place according to the determination of this frequency. The method proposed will be all the more precise, as the number of measurements of V_{R} will be higher. Practically, the method will be all the more precise, as the number of sensors used will be larger and the available range for each sensor wider.

References

1. B. Cros, H. Nounah, J. Attal, Thin Solid Films **301**, 134, (1997).
2. B. Cros, E. Gat, J.M. Saurel, J. Non-Cryst. Solids **209**, 273 (1997).
3. M. Amlouk, N. Brunet, B. Cros, S. Belgacem, D. Barjon, J. Phys. III France **7**, 1741 (1997).
4. J. Kushibiki, T. Ishikawa, N. Chubachi, Appl. Phys. Lett. **57**, 1967 (1990).
5. O. Behrend, A. Kulik, G. Gremaud, Appl. Phys. Lett. **62**, 2787 (1993).
6. J.O. Kim, J.D. Achenbach, Thin Solid Films **214**, 25 (1992).
7. H. Nounah, Ph.D. thesis, Montpellier, France, 1995.
8. A.K. Mal, P.C. Xu, Y. Bar-Cohen, Int. J. Engin. Sci. **27**, 779 (1989).
9. L.M. Brekhovskikh, *Waves in Layered Media* (Academic Press, NY, 1960).