# EXPERIMENTAL VERIFICATION OF KRAMERS-KRONIG RELATIONSHIP IN ANISOTROPIC COMPOSITE MATERIALS

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## INTRODUCTION

Measuring elastic constants ( $C_{ij}$) of composite materials from ultrasonic velocities is a very well known technique [1,2], using the propagation of bulk modes generated at the fluid-solid interface. There is still a question about the validity of these measurements at lower frequencies or for static stress fields. These materials made with viscoelastic matrix are anisotropic and viscoelastic. Hence, the attenuation is also anisotropic. In an absorbing medium, the propagation of waves is dispersive: the phase velocity depends on the frequency. Attenuation and velocity are linked together through the Kramers-Kronig relations that are deduced from the principle of causality [3].

A way to validate the $C_{ij}$ measurements in the static domain, is to build a model of their variation to prolong it toward lower frequencies. In that effort, this paper presents the first aspect of this problem that is the connection between anisotropic attenuation and anisotropic dependence of phase velocity versus frequency.

A well - known set-up to measure attenuations and velocities in any direction of propagation, is the immersion method .This furnishes the whole set of complex viscoelastic constants, ${}^{*}C_{ij} = C'_{ij} + iC''_{ij}$ , where the real parts are deduced from velocities and the imaginary parts from attenuations [1].

In an orthotropic material, this complex matrix has nine independent constants. Orthotropic materials have three planes [4] of symmetry. Composite materials made of superimposed plies are shaped like a plate, which defines the principal plane $P_{23}$ (see Fig. 1).

$$^*C_{ij} = C'_{ij} + i\ C''_{ij}$$

$$\begin{bmatrix} ^*C_{11} & ^*C_{12} & ^*C_{13} & 0 & 0 & 0 \\ & ^*C_{22} & ^*C_{23} & 0 & 0 & 0 \\ & & ^*C_{33} & 0 & 0 & 0 \\ & & & ^*C_{44} & 0 & 0 \\ \text{Sym} & & & & ^*C_{55} & 0 \\ & & & & & ^*C_{66} \end{bmatrix}$$

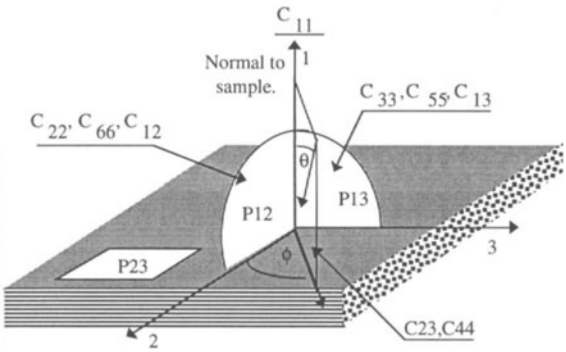


Fig. 1 Axis and planes of symmetry in long fibers composite materials.

Using the immersion technique, it is possible to measure seven of these constants in two planes of symmetry and the two others in an another plane [1] .

The purpose of this paper is to present an experimental verification of the Kramers-Krönig relations in an anisotropic plane (here $P_{13}$) of a unidirectional composite made of glass fibers and epoxy matrix, taking into account the heterogeneous structure of wave generated through the liquid-solid interface.

## THEORY

A plane wave propagating in a solid can be represented by a displacement field as :

$$\vec{U} = \vec{P}\exp\left(+i\left(2\pi\nu\left(t - \vec{S}.\vec{M}\right)\right)\right) \qquad [1]$$

with $\vec{S} = \vec{S}' - i\vec{S}''$, where $\vec{S}'$ is the wave propagation vector and $\vec{S}''$ the damping vector.

In a viscoelastic material , the dispersion relation between any modulus like Young's or Coulomb's modulus , viscoelastic moduli $^*C_{ij}$ in an axis of symmetry, etc., is given by [3] :

$$G(\nu) = G_1(\nu) + i\ G_2(\nu) = \frac{\rho}{\vec{S}^2} \qquad [2]$$

In the general case of an arbitrary direction of propagation in an infinite medium, three heterogeneous modes are the solutions of the Christoffel's equations $\left|{}^*\Gamma_{ij} - \frac{\rho}{{}^*\vec{S}.{}^*\vec{S}}\delta_{ij}\right| = 0$ where the Christoffel's tensor $^*\Gamma_{ij}$ is built with the viscoelastic moduli $^*C_{ij}$ and the direction of propagation [4] . With the hypothesis of small attenuation [1] , these equations take the shape: $G\left({}^*\Gamma_{ij}\right) = \frac{\rho}{\vec{S}^2}$. The decomposition of the complex $G$ function in real part $G_1(\nu)$ and imaginary part $G_2(\nu)$ leads to the same equation [2].

The damping vector is assumed to be smaller than the wave vector. This hypothesis needs to be satisfied if one wants to propagate waves through materials in order to measure their properties. Indeed, if the signal attenuation is too large, no signal can be transmitted through the sample.

Then :

$$\rho \approx G_1(\nu)\vec{S}'^2 + i\left(G_2(\nu)\vec{S}'^2 - 2i\ \vec{S}'\vec{S}''G_1(\nu)\right) \qquad [3]$$

$$G_1(\nu) = \frac{\rho}{S'^2} \quad \text{and} \quad G_2(\nu) = 2\frac{\rho\vec{S}'\vec{S}''}{S'^4}\ . \qquad [4]$$

The principle of causality [3] imposes that $G_2$ is the Hilbert transform of $G_1$:

$$G_2(\nu) = \frac{1}{\pi}\int_{-\infty}^{+\infty}\frac{G_1(u)}{u-\nu}\,du = Hi(G_1(\nu)) \ \text{, and reciprocally: } G_1(\nu) = -Hi(G_2(\nu)). \qquad [5]$$

Because the temporal representation $g(t)$ of the modulus is real, its Fourier transform is such that $G_1(u) = G_1(-u)$ and $G_2(u) = -G_2(-u)$. This property leads to the classical Kramers-Kronig relations :

$$G_1(\nu) = -\frac{2}{\pi}\int_0^{+\infty}\frac{uG_2(u)}{u^2-\nu^2}\,du \quad \text{and} \quad G_2(\nu) = \frac{2\nu}{\pi}\int_0^{+\infty}\frac{G_1(u)}{u^2-\nu^2}\,du. \qquad [6]$$

In the case of heterogeneous wave generated at the liquid-solid interface [5], let us denote $S_r$ the projection of the damping vector onto the wave vector (Fig. 2). $S_r = S''\cos(\theta_r)$ where $\theta_r$ is the refraction angle.

The relationship between real and imaginary parts of the slowness vector become:

$$\frac{1}{S'^2} = \frac{4}{\pi}\int_0^{+\infty}\frac{S_r}{S'^3}\frac{u}{u^2-\nu^2}\,du \qquad \frac{S_r}{S'^3} = \frac{\nu}{\pi}\int_0^{+\infty}\frac{1}{S'^2\left(u^2-\nu^2\right)}\,du \qquad [7]$$

M.O.Donnell and al. [6] prove that those relations can be written in a "local" form , over a limited frequency bandwidth :

$$S' = S'_0 - \frac{2}{\pi}\int_{\nu_0}^{\nu}\frac{S_r}{u}\,du \quad \text{where } S'_0 = \frac{1}{C(\nu_0)} = \frac{1}{C_0} \qquad [8]$$

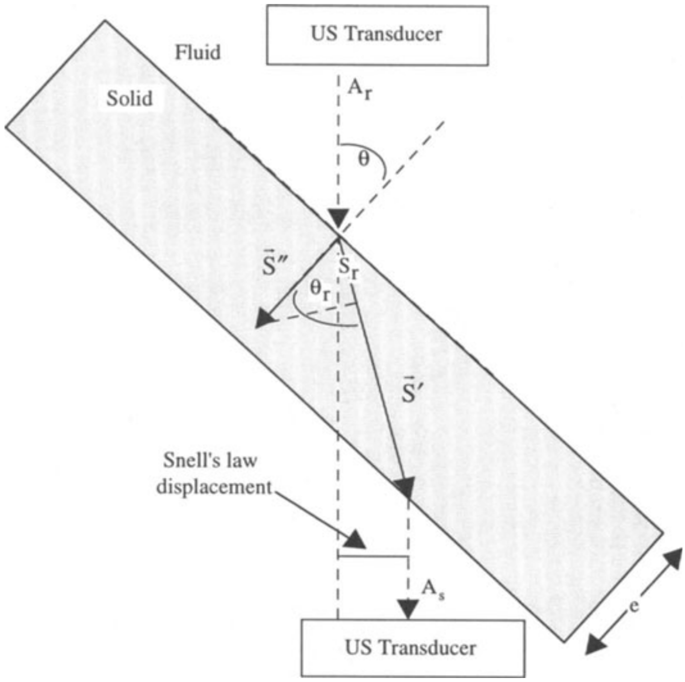


Fig. 2 Heterogeneous wave generation at liquid - solid interface.

In many papers [6-9] , it was shown that the attenuation factor $\alpha = 2\pi\nu S_r$ is almost varying linearly with frequency in the ultrasonic frequencies range. The slope of the curve is noted $a_1$. This model is simple and useful because it implies that the $C''_{ij}$ are constant [7] . To take into account that this curve does not pass through the origin, it is mandatory to increase the degree of the polynomial. In this paper, a simple second order polynomial is used :

$$\alpha = a_1\nu + a_2\nu^2 \quad . \qquad [9]$$

Then the velocity dispersion can be easily linked to the attenuation by the formula :

$$S' = S'_0 - \frac{a_1}{\pi^2}\mathrm{Ln}\left(\frac{\nu}{\nu_0}\right) - \frac{a_2}{\pi^2}(\nu - \nu_0) \qquad [10]$$

ATTENUATION MEASUREMENT

The incident wave amplitude $A_r(\nu)$ is defined by the Fourier transform of the acquired waveform resulting of propagation between an ultrasonic transmitter and an ultrasonic receiver (Bandwidth $\approx$ 0.5 - 3 MHz ) when the solid is not present.

The amplitude of the transmitted waveform $A_s(\nu)$ is normalized by $A_r(\nu)$ to get the transfer function of the two interfaces and the propagation through the solid. This amplitude $A(\nu)=\dfrac{A_s(\nu)}{A_r(\nu)}$ corresponds to the normalized amplitude of the heterogeneous bulk mode first transmitted through the plate . An important feature to measure correctly this amplitude, is to move the receiver with a shift computed from the Snell'law [Fig. 2] . The modulus of this complex amplitude permits to compute $S''$ from the formula :

$$|A(\nu)| = T \quad \exp(-\ 2\pi\nu\ S''e\ ) \qquad [11]$$

T is the transmission coefficient of both interfaces , computed from the real part of $^*C_{ij}$ [1] and e is the thickness of the sample.

In Fig. 3, the measurement of $S_r$ in the $P_{12}$ plane and the value of $S_r$ computed from the $^*C_{ij}$ values (Table 1) for heterogeneous modes are plotted. As expected, this plane looks almost isotropic. From the $^*C_{ij}$ values, it is also possible to compute the values of $S''$ for homogeneous modes. For comparison, these values are also shown in Fig. 3 for homogeneous modes propagating in the same direction than the heterogeneous modes. (For them, the incidence has no meaning; the direction of propagation is the correspondant refraction angle). As already proved [5] , in an isotropic plane, if the attenuation is small, the value of $S_r$ for heterogeneous modes is close to the value of $S''$ for homogeneous modes.

As shown in Fig. 4, if the plane is anisotropic, there is a noticeable difference between these two values. This permits to check experimentally which of the two values has to be used in the Kramers-Kronig relations.

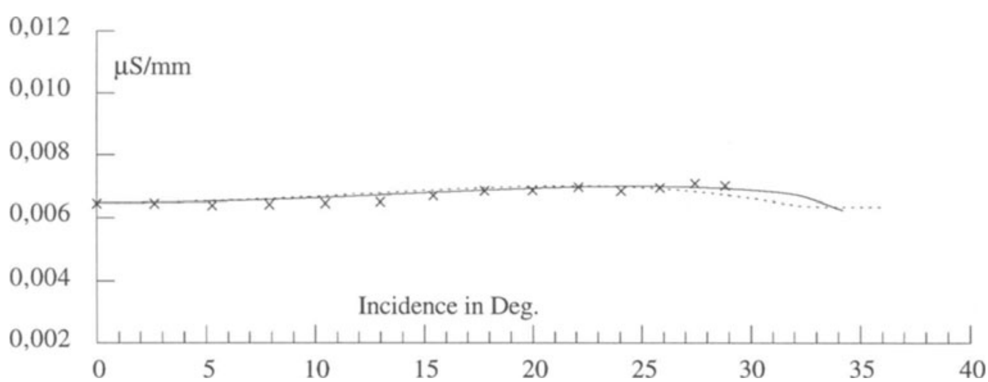


Fig. 3 $S_r$ versus incidence for quasi-longitudinal mode in the plane $P_{12}$
X : Experimental data ; —— : $S_r$ for heterogeneous mode;
----- : $S''$ for homogeneous mode.

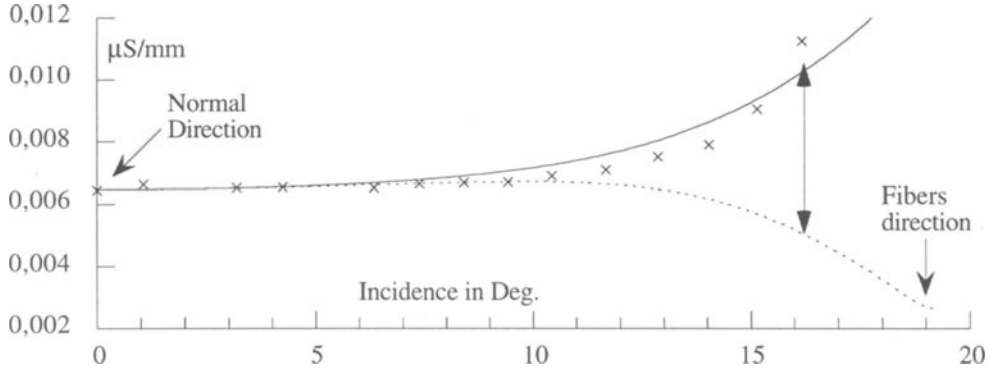


Fig. 4 $S_r$ versus incidence for quasi-longitudinal mode in the plane $P_{13}$
X : Experimental data ; —— : $S_r$ for heterogeneous mode;
----- : $S''$ for homogeneous mode.

The two coefficients $a_1$ and $a_2$ [9] are identified by a Newton-Raphton's procedure to get the best fit between the polynom and the attenuation measurements.

The evolution of $\alpha$ versus frequency is presented at Fig. 5 at normal incidence and an incidence of 16°. For this incidence, the wave propagates in the solid at an angle of $\theta_r = 38°$ . It is surprising that the attenuation factor is larger when the direction of propagation approaches the fiber direction. As explained at Fig. 4, the attenuation factor is larger for heterogeneous waves than for homogenous waves propagating in the same direction. That effect can be verified with the velocity variation versus frequency.

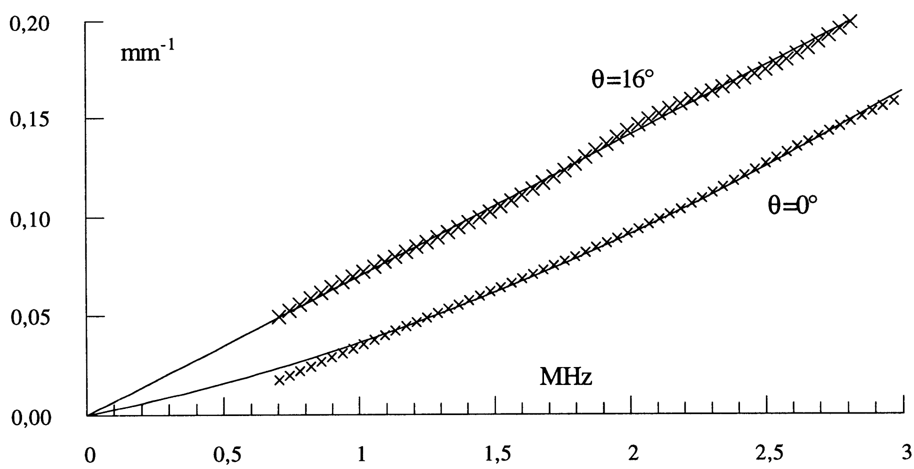


Fig. 5 Attenuation factor $\alpha = 2\pi\nu S_r$ versus frequency of the quasi-longitudinal mode in the plane $P_{13}$ for $\theta = 0°$ and $\theta = 16°$

According the classical ultrasonic spectroscopy technique [10], The delay introduced by the propagation of the wave from one interface to the other is given by :

$$\tau(\nu) = \frac{\varphi(\nu)}{2\pi\nu} = \frac{e}{\cos(\theta_r)} S' , \qquad [12]$$

where $\varphi(\nu)$ is the phase of the complex amplitude $A(\nu)$. Then, it is straightforward to deduce the evolution of the phase velocity versus frequency, from the $S'$ measurement. This is shown in Fig. 6, for the quasi-longitudinal heterogeneous mode propagating in the direction $\theta_r = 38°$ in the frequency range of 0.5-3 MHz.

The velocities are also computed from the values of coefficients $a_1$ and $a_2$ deduced from the attenuation [10] .The frequency $\nu_0$ is the lower frequency in the bandwidth and $S_0'$ at this frequency is identified by a Newton-Raphton's procedure from the velocity measurements.

The good correspondence between measured and computed velocities validates the Kramers-Krönig relations between velocity and the attenuation factor of the heterogeneous waves. We observed the same good fitting in any other direction. In Fig. 6, we also plotted the dispersion curve computed from values of $S''$ for homogeneous waves propagating in

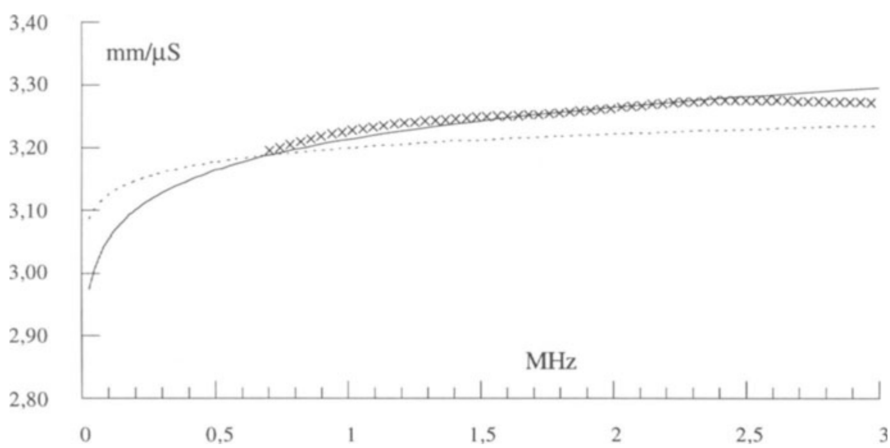


Fig. 6 Velocities versus frequency of the quasi-longitudinal mode in the plane $P_{13}$
for $\theta = 16°$ x : Experimental velocities ;
__ : Velocities computed from heterogeneous wave attenuation;
---- : Velocities computed from homogeneous wave attenuation.

Table 1 $^*C_{ij}$ values in the plane $P_{13}$, in GPa at 1.5 MHz

| $^*C_{11}$ | $^*C_{33}$ | $^*C_{55}$ | $^*C_{13}$ |
|---|---|---|---|
| 14.7 + i 0.5 | 40 + i 0.9 | 3.9 + i 0.3 | 7.3 + i 0.2 |

the same direction. This smaller factor leads to a smaller velocity dispersion. The difference is large enough to prove the need to take into account the wave heterogeneity .

## CONCLUSIONS

The Kramers-Krönig relations between attenuation and velocity in anisotropic medium, are verified, as in isotropic medium, if the heterogeneity of the quasi-longitudinal modes is taken into account; the same study for quasi-shear heterogeneous modes will be presented in a further paper. Then, these relations could permit to reach material properties at low frequencies, from ultrasonic measurements.

But the question of how far can the model be extended towards lower frequencies, using these relations, has still to be solved.

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