Thin layer Characterization by ZGV Lamb modes

Maximin Cès, Dominique Clorennec, Daniel Royer and Claire Prada
Laboratoire Ondes et Acoustique, ESPCI- Université Paris 7- CNRS UMR 7587, 10 rue Vauquelin, 75231 Paris Cedex 05- France
E-mail: maximin.ces@espci.fr

Abstract. Ultrasonic non-destructive testing of plates can be performed with Lamb modes guided by the structure. Non contact generation and detection of the elastic waves can be achieved with optical means such as a pulsed laser source and an interferometer. With this setup, we propose a method using zero group velocity (ZGV) Lamb modes rather than propagating modes. These ZGV modes have noteworthy properties, in particular their group velocity vanishes, whereas their phase velocity remains finite. Thus, a significant part of the energy deposited by the pulsed laser can be trapped in the source area. For example, in a homogeneous isotropic plate and at the minimum frequency of the $S_1$-Lamb mode a very sharp resonance can be observed, the frequency of which only depends on the plate thickness, for a given material. In fact, other ZGV modes exist and the set of ZGV resonance frequencies provide a local and absolute measurement of Poisson’s ratio. These non-propagating modes can also be used to characterize multi-layered structures. Experimentally, we observed that a thin (500 nm) gold layer deposited on a thick (1.5 mm) Duralumin plate induces a sensitive down-shift of the set of ZGV resonance frequencies. This shift, which is typically 5 kHz for the $S_1$-Lamb mode at 1.924 MHz, can be approximated by a formula providing the layer thickness. Thickness down to 100 nm can be estimated by this method. Such a sensitivity with conventional ultrasound inspection by acoustic microscopy would require an operating frequency in the GHz range.

1. Introduction

Coatings are commonly used in industrial processes for protecting surfaces against aggressive environment such as corroding media, high temperatures, friction or simply wear. Over past decades, various ultrasonic techniques have been developed to characterize the deposited material. Using Laser Based Ultrasonics, the measurements can be achieved without any mechanical contact with the part under test. Recently, techniques using zero group velocity Lamb modes provided local measurements of material parameters and thickness of homogeneous isotropic plates [1].

Zero group velocity Lamb modes (ZGV) are plate modes whose group velocities vanish for non-zero wavenumbers. Such non-propagative modes have been experimentally observed when source and detection are superimposed: they give rise to local resonances. Those resonances, predicted by Tolstoy and Usdin [2], have recently been highlighted [1, 3, 4, 5]. Sharp peaks can be observed in the spectrum of the normal displacement. Prada et al. [1] have shown for isotropic plates that the bundle of these ZGV resonances is related to the thickness and the material parameters.

The objective of this study is to apply this method to inhomogeneous samples like isotropic plates covered with thin layers and to determine the film characteristics.
2. Zero group velocity Lamb modes

As shown in Fig. 1, plotted for a Duralumin plate ($V_L=6.398 \text{ mm/μs}; V_T=3.122 \text{ mm/μs}$), some branches of the dispersion curves $\omega(k)$ exhibit minima for non-zero wavenumbers. At the corresponding point, the group velocity vanishes, whereas the phase velocity remains finite, hence the name *Zero Group Velocity* given to these modes. The existence and the relative values of these frequencies depend on the Poisson’s ratio. Experimentally, this interesting phenomenon can be observed by detecting the normal displacement at the source point or less than half a wavelength apart from the source. The spectrum of the signal exhibits sharp peaks whose frequencies match ZGV modes.

These sharp resonances illustrate the non-propagative property of ZGV modes: the laser generation gives rise to numerous propagating and non-propagating Lamb modes and only Lamb modes whose energy velocity, i.e. group velocity, is zero and whose phase velocity is finite remain after a few microseconds. The resonance frequency gives an absolute measurement of bulk wave velocities, if the plate thickness is known [6].

In our experiment, elastic waves are generated in the sample using a 20-ns laser pulse and the local displacement in the neighbourhood of the source is measured by an optical interferometer. The normal displacement can oscillate during a very long time. The recorded signal is shown in Fig. 2, in the case of a 1.5-mm thick Duralumin plate. The Fourier transform of the measured signal reveals the expected resonances. Apart from the continuous background due to the $A_0$ mode, each peak corresponds to a ZGV mode. Their amplitude is decaying, because the laser impulse does not excite all the modes with the same efficiency: low frequency components are more excited than higher ones due to the frequency content of the laser pulse.

The existence of ZGV modes depends on Poisson’s ratio. This phenomenon can be explained in terms of repulsion at $k \neq 0$ between modes of the same family and of different parity, such as $S_{2n+1}$ and $S_{2n}$ or $A_{2n}$ and $A_{2n+1}$ [1]. The smaller the difference between the cut-off frequencies of two modes, the stronger the repulsion between these modes. Shuvalov et al. [7] give a simple criterion to predict the ZGV modes existence from the curvature of the dispersion curves for $k = 0$. For the first ZGV mode, the ZGV frequency depends on the cut-off frequency ($k = 0$) through a multiplicative factor denoted $\beta$ in the *Impact Echo* (I. E.) method: $f_{ZGV} = \beta V_L/2d$ [4]. This coefficient depends only on Poisson’s ratio $\nu$. It can be generalized to other ZGV modes. The value of the resonance frequency obtained for a ZGV mode provides the *local* plate thickness if the longitudinal and transverse velocities are known [6]. Theoretically, in a thickness resonance the whole surface is vibrating in phase, because of the vanishing wave number, whereas a ZGV resonance is a local resonance, with a finite wave number. Compared to the use of a

![Figure 1. Lamb mode dispersion curves for a Duralumin plate of thickness $d$. Red: symmetric modes, Blue: antisymmetric modes, Arrow: ZGV point.](image-url)
Figure 2. Normal displacement detected at the source point on a 1.5 mm-thick Duralumin plate, on the left, and its Fourier transform on the right. The large low frequency oscillation is due to the $A_0$ mode, which appears in the spectrum as the continuous background. The sharpest peaks correspond to ZGV modes, the thickness resonances occur at frequencies 3.2 MHz, 7.5 MHz, 11.7 MHz. The spectrum is represented above 1.9 MHz, because at lower frequencies $A_0$ mode dominates.

thickness resonance, the relevance of such a measurement is the locality of the result.

3. Experimental setup
The experimental setup is shown in Fig. 3. The thermoelastic source is a Q-switched Nd:YAG laser (optical wavelength 1064 nm) providing a pulse having 20 ns duration and 4 mJ of energy. The spot diameter of the unfocused beam is equal to 1 mm, in order to generate efficiently the first mode [1, 8]. Lamb waves were detected by a heterodyne interferometer equipped with a 100 mW, frequency doubled Nd:YAG laser (wavelength 532 nm) [9]. This interferometer is sensitive to any phase shift along the path of the optical probe beam reflected by the moving surface. In the experiments, the source and detection points are superimposed. The normal displacement is recorded using an oscilloscope linked to a computer, which permits to process the data.

Figure 3. Experimental setup
4. Measurements

A set of measurements are performed on Duralumin samples. We assume that the plate is isotropic. The sample is placed on a motorized stage for the scanning, and three sets of measurements were carried out on the bare face using the measurement system described above. On the opposite side a thin gold layer is deposited. Sample #1 is a 150-mm square Duralumin plate of thickness 1.513 mm half-covered with a 500-nm thick gold film. Sample #2 is a Duralumin plate of the same dimensions half-covered with a 100-nm thick gold film. A 3-nm thick chromium layer was deposited before the gold layer. We assume that the values of Poisson’s ratio are \( \nu_{\text{Duralumin}} = 0.344 \) and \( \nu_{\text{gold}} = 0.421 \) [10].

On sample #1, the normal displacement was recorded at two points 3 cm apart, one on the bare region, the other on the gold-covered region.

The spectrum shown in Fig. 4 was obtained after processing the first 10 \( \mu \)s of signal. Seven peaks can be distinguished, corresponding to the \( S_1S_2, S_5S_6, S_5S_{10}, S_7S_{14}, S_9S_{18}, S_{11}S_{22} \) and \( S_{13}S_{26} \) ZGV modes. On the enlargement in Fig. 5, it is clear that the deposit modifies the ZGV frequencies. The frequencies are down-shifted for the gold-covered region.

On sample #1, a 50 mm-scan has been performed over the two different regions, with a 0.25 mm spatial step. The resonance frequency shifts are shown in Fig. 6.

The frequency step between the two regions is obvious for the three first observable ZGV modes. In Table 1, the frequency differences between the two regions are compared to simulations performed with Disperse software [11], using the following material parameters [10]:

- 500-nm thick gold layer, mass-density \( \rho = 19281 \text{ kg/m}^3 \), \( V_L = 3.240 \text{ mm/\mu s} \), \( V_T = 1.200 \text{ mm/\mu s} \)
- 1.5-mm thick Duralumin plate, mass-density \( \rho = 2795 \text{ kg/m}^3 \), \( V_L = 6.398 \text{ mm/\mu s} \), \( V_T = 3.122 \text{ mm/\mu s} \)

On sample #2, a 50 mm-scan was performed over the two different regions, with a 0.5 mm spatial step, at a 250 MHz sampling frequency. A ”reference scan” was acquired before the deposit and a second scan after, so that the effect of the 100-nm deposit can clearly be observed.
Figure 5. Enlargement of Fig. 4, for $S_1S_2$ ($S_7S_{14}$) resonance on the left (on the right). The solid (dashed) curve corresponds to the bare (gold-covered) region.

Figure 6. ZGV resonance frequencies obtained over 539 $\mu$s, for sample #1.

Table 1. Comparison between measurements and theoretical predictions on sample #1.

| Mean frequency difference (kHz) | $S_1S_2$ | $S_3S_6$ | $S_5S_{10}$ |
|-------------------------------|---------|---------|-------------|
| measured                      | 4.8     | 14.4    | 22.6        |
| theoretical                   | 5.1     | 14.6    | 23.9        |
| Relative difference            | 6 %     | 1 %     | 6 %         |

Independently of the local thickness variations of the plate. In Fig. 7, the three first ZGV resonances are plotted with respect to the distance, before the deposit, in green, and after the deposit, in blue. The curves should be superimposed on the bare region. Nevertheless, there is a tiny difference (260 Hz for $S_1S_2$, 980 Hz for $S_3S_6$ and 1.98 kHz for $S_5S_{10}$), which may be
due to a temperature variation of less than 1 °C. Indeed ZGV resonance frequencies decrease when the temperature rises [8]. We can observe that the frequency is increasing as a function of distance, which means that the plate thickness is decreasing with respect to distance. It is thus more interesting to plot the difference between the three first ZGV resonance frequencies before and after the deposit, versus the distance (Fig. 8).

In Table 2, experimental results are compared to simulations performed with Disperse software [11]. The maximum standard deviation for the frequency difference is 100 Hz for $S_1S_2$, 580 Hz for $S_3S_6$ and 1.2 kHz for $S_5S_{10}$. Thus, it could be possible to detect layers three times thinner.

5. Discussion
The dispersion curves of a Duralumin plate of thickness 1.5 mm covered with thin gold layers of different thicknesses (0, 100, 200, 300, 500, 800 nm) have been calculated with Disperse software. Then the ZGV and thickness modes frequencies were extracted. In Fig.9, the frequency variations $\Delta f$ due to the deposit are drawn for $S_1S_2$, $S_3S_6$ and $S_5S_{10}$ ZGV Lamb modes, for both thickness resonances ($f_C$, with circles) and ZGV resonances ($f_{ZGV}$, with crosses), with respect to thickness deposit. Three important features can be noticed:
Frequency variations in ZGV resonances and in thickness resonances are quite similar: 
\[ \Delta f_C \approx \Delta f_{ZGV} \], especially for higher order modes: the higher the order of the mode, the weaker the curvature of the dispersion curve \( \omega(k) \), so that ZGV and thickness resonance frequencies have closer and closer behaviour;

- for both ZGV and thickness resonances, the frequencies variations are proportional to the thickness of the deposit;
- for both ZGV and thickness resonances, the frequencies variations are proportional to the order of the mode.

![Figure 9](image1.png)

**Figure 9.** Frequency variations \( \Delta f \) due to the deposit with respect to the gold layer thickness, for \( S_1 S_2, S_3 S_6 \) and \( S_5 S_{10} \) ZGV Lamb modes (theory).

This behaviour can be experimentally observed. In Fig. 10, the \( \Delta f_{ZGV}/f_{ZGV} \) ratio due to the 500 nm gold deposit is drawn for \( S_1 S_2, S_3 S_6, S_5 S_{10}, S_7 S_{14}, S_9 S_{18}, S_{11} S_{22}, S_{13} S_{26} \) ZGV modes, with respect to the ZGV resonance frequencies. The data are extracted from Fig. 4. It can be noticed that the frequency shift is proportional to the mode order, as expected.

![Figure 10](image2.png)

**Figure 10.** \( \Delta f/f \) due to the 500 nm gold layer, with respect to the bare plate ZGV resonance frequency (experiment).

Actually, thickness mode frequencies must be proportional to the mass of the deposit. This assumption is used in quartz crystal microbalances to determine the mass of a thin layer. A more precise relation between masses and cut-off frequencies is [12, 13]:

\[
\frac{\Delta m}{m} = -\frac{1}{\pi} \frac{Z_{layer}}{Z_{plate}} f_c \frac{\Delta f_{ZGV}}{f_{ZGV}} \arctan \left[ \frac{Z_{plate}}{Z_{layer}} \tan \left( \pi \frac{f_c}{f_c'} \right) \right]
\]

(1)

where:
- \( \Delta m \): mass of the layer ; \( m \): plate mass
- \( f_c \): cut-off frequency for the bare plate; \( f_c' \): cut-off frequency for the gold-covered plate
- \( Z_{layer} \): layer acoustic impedance; \( Z_{plate} \): plate acoustic impedance

The formula can be modified. Using \( f_c = f_{ZGV}/\beta \) and \( \Delta f_c \approx \Delta f_{ZGV} \), it is possible to write:

\[
\frac{\Delta m}{m} \approx -\frac{1}{\pi} \frac{Z_{layer}}{Z_{plate}} \frac{f_{ZGV}}{f_{ZGV} + \beta \Delta f_{ZGV}} \arctan \left[ \frac{Z_{plate}}{Z_{layer}} \tan \left( \pi \frac{\Delta f_{ZGV}}{f_{ZGV}} \right) \right]
\]

(2)
Applying this formula to $S_1S_2$ Lamb mode, it was found that sample # 1 layer thickness was 530 nm instead of 570 nm measured with an optical profilometer, and 90 nm instead of 100 nm expected for sample # 2. The error may be caused by the uncertainty on the layer mass-density. As predicted in [12, 13], the deposited mass is too low to allow the determination of $Z_{layer}/Z_{plate}$.

6. Conclusion
Using laser-based ultrasonic techniques, we have investigated the effects on zero group velocity resonance frequencies of a thin gold layer deposited on a thick Duralumin plate. Experimental results are in good agreement with theory. The temperature has not been measured in the presented results, but further measurements reveal that in our material, a temperature change of 1 °C causes a frequency shift of about 430 Hz for $S_1S_2$ resonance, 1.5 kHz for $S_3S_6$ resonance and 2.5 kHz for $S_5S_{10}$ resonance. It was found experimentally that the ZGV resonance spectrum is down-shifted by the deposit, and is quasi proportional to the deposited mass. The higher the order of the mode, the larger the frequency shift. This frequency difference allows to estimate the layer thickness, in agreement with optical measurements. Thicker deposits may enable the measurement of the layer elastic moduli.

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