GENERATION AND DETECTION OF QUASI TRANSVERSE WAVES IN AN ANISOTROPIC CRYSTAL BY PICOSECOND ULTRASONICS

T. Bienville and B. Perrin*
Université Pierre et Marie Curie / CNRS, LMDH, Boîte 86, 4 place Jussieu, 75005 Paris cedex 05, France
*e-mail: bper@ccr.jussieu.fr

Abstract

We present experimental results demonstrating the generation and the detection of a quasi transverse acoustic wave in picosecond laser ultrasonics. A longitudinal pulse is generated in an isotropic polycrystalline metallic film at the interface with an anisotropic transparent crystal cut along a non symmetry direction. Mode conversion at the interface gives rise to short quasi transverse and quasi longitudinal acoustic pulses which propagate in the crystal. They are detected by a time delayed laser pulse while they propagate in the transparent crystal ; these pulses give Brillouin oscillations with two different frequencies in the transient reflectivity of the metallic film.

Introduction

Soon after the laser discovery, R. M. White [1] was the first to point out that this light source could be used to generate ultrasonic waves. This non-contact method to excite longitudinal, transverse or surface waves appeared very attractive and the "laser ultrasonics" field expanded rapidly. Later on, H.J. Maris [2] proved that it was possible to generate and detect picosecond acoustic pulses using femtosecond lasers and a pump-probe optical configuration. The very high temporal resolution of this technique allows SONAR experiments [3] in very thin films (with a typical thickness between a few tens of nanometers and a few micrometers). However due to the large ratio between the source diameter (10-50 μm) and the typical acoustic wavelength (a few tens of nanometers), only longitudinal waves can be studied with such a technique. Reducing the laser spot diameter, surface waves have been successfully generated and detected [4].

Recently, it has been suggested that a mode conversion process occurring during the reflection of a longitudinal acoustic pulse on the interface between an isotropic polycrystalline metallic film and an anisotropic material could be used to generate a short transverse acoustic pulse in the film [5]. These new experiments could extend the field of picosecond laser ultrasonics to the determination of new elastic constants in thin film materials. However it can be suspected that, in such a configuration, the polish process could damage the crystalline structure of the anisotropic material over a small thickness right below the interface. In such circumstances the mode conversion efficiency for the highest frequency waves could be drastically reduced.

To avoid this problem, we chose, in this work, to use the mode conversion process which occurs during the transmission at the interface between isotropic and anisotropic materials. In this configuration it can be expected that the high frequency waves mode conversion efficiency is less affected by surface damages. Furthermore we use a transparent anisotropic crystal to obtain an easier detection of the expected quasi-transverse wave propagating in the crystal.

Generation of picosecond transverse acoustic waves

The experimental configuration is described in Figs. 1 and 2. The laser pump pulse crosses the transparent crystal and is absorbed in the metallic polycrystalline film where it generates a longitudinal short acoustic pulse which propagates along a direction normal to the interface between the film and the anisotropic transparent substrate. A part of this pulse is transmitted in

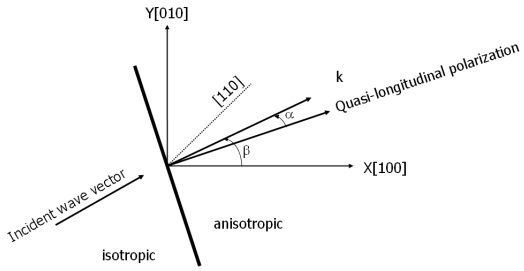

Figure 1: Mode conversion at the film/crystal interface.

the anisotropic crystal and is mode converted in quasi-transverse and quasi longitudinal waves. We chose a

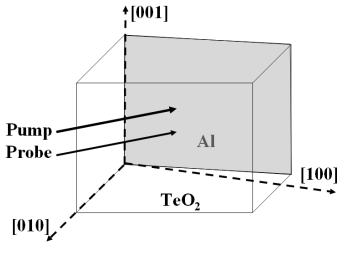

Figure 2: First experimental configuration (pump and probe normal to the crystal direction [001]).

tetragonal paratellurite crystal (TeO_2) because of its transparency and its strong anisotropy. The [001] axis of the crystal is parallel to the interface and the angle between the normal to the interface and the [100] axis is β in the plane ([100],[010]). The normal modes along this propagation direction in the crystal are a quasi-longitudinal wave with a polarization in the plane ([100],[010]) defined by the angle α with the propagation direction, a quasi transverse wave in the same plane and a pure transverse wave polarized along the [001] axis ; this last wave is not involved in the conversion process. Taking into account the boundary conditions for the stress tensor and the displacement which occur at the transducer/crystal interface, the expressions for the mode conversion coefficients can be obtained :

$$T_{l,qt} = \frac{-2z_l\left(z_t+Z_{ql}\right)\sin\alpha}{\left(z_l+Z_{ql}\right)\left(z_t+Z_{qt}\right)+\left(z_l-z_t\right)\left(Z_{ql}-Z_{qt}\right)\sin^2\alpha}$$

$$T_{l,ql} = \frac{2z_l\left(z_t+Z_{qt}\right)\cos\alpha}{\left(z_l+Z_{ql}\right)\left(z_t+Z_{qt}\right)+\left(z_l-z_t\right)\left(Z_{ql}-Z_{qt}\right)\sin^2\alpha}$$

$$R_{l,t} = \frac{-z_l\left(Z_{ql}-Z_{qt}\right)\sin 2\alpha}{\left(z_l+Z_{ql}\right)\left(z_t+Z_{qt}\right)+\left(z_l-z_t\right)\left(Z_{ql}-Z_{qt}\right)\sin^2\alpha}$$

$$R_{l,l} = \frac{\left(z_l-Z_{ql}\right)\left(z_t+Z_{qt}\right)+\left(z_l+z_t\right)\left(Z_{ql}-Z_{qt}\right)\sin^2\alpha}{\left(z_l+Z_{ql}\right)\left(z_t+Z_{qt}\right)+\left(z_l-z_t\right)\left(Z_{ql}-Z_{qt}\right)\sin^2\alpha} \quad (1)$$

$T_{l,ql}$ and $T_{l,qt}$ are the mode conversion coefficients of a longitudinal wave propagating in the metallic film and transmitted in the anisotropic crystal where it gives rise to a quasi-longitudinal wave ($T_{l,ql}$) and a quasi-transverse wave ($T_{l,qt}$). $R_{l,l}$ and $R_{l,t}$ are the mode conversion coefficients of this longitudinal wave reflected at the interface. Z_{ql} and Z_{qt} are the acoustic impedances in the anisotropic medium for quasi-longitudinal and quasi-transverse waves respectively ; z_l and z_t are the acoustic impedances in the isotropic medium for longitudinal and transverse waves respectively. The dependence of these four coefficients in terms of the angle β is displayed on Fig.3. The metallic film is made of aluminum for which the mode conversion coefficient $R_{l,t}$ exhibits one of the highest maxima among metals (24%) ; this maximum value is reached for an angle $\beta \simeq 28^\circ$ which was used in the experiments described in this paper. For this angle, the predicted sound velocities are $v_{ql} = 4.27 nm/ps$ and $v_{qt} = 1.42 nm/ps$ for the quasi longitudinal and the quasi transverse waves respectively. Values $v_l = 6.4 nm/ps$ and $v_t = 3.1 nm/ps$ were used for the longitudinal and transverse waves in the isotropic polycrystalline aluminum film. A 300 nm aluminum film was sputtered on the $Te0_2$ crystal. The pump and the probe pulses are derived from a Ti:Sapphire mode locked laser which gives a 100 fs pulse duration with

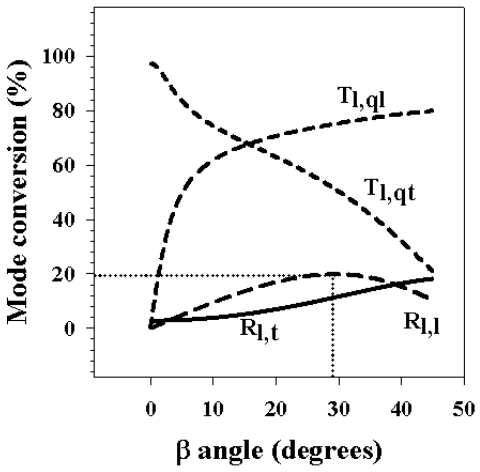

Figure 3: Mode conversion of an incident longitudinal wave in an aluminum film deposited on a TeO_2 substrate.

a 82 MHz repetition rate. The experiments were performed at a central optical wavelength of 750 nm. The pump is chopped with an acousto-optic modulator at 1 MHz. The optical probe beam propagates along a variable optical delay line, is reflected on the metallic film and collected by a photodiode. The detector output is sent to a lock-in amplifier.

The acoustic waves which propagate in the transparent crystal modify its refractive index tensor through the photoelastic effect. This change is given by [6]

$$\Delta\varepsilon_{il} = -\varepsilon_{ij}\, p_{jkmn}\varepsilon_{kl}\eta_{mn} \quad (2)$$

where η_{mn} stand for the strain tensor components, ε_{ij} the dielectric tensor and p_{jkmn} the photoelastic tensor.

$$\varepsilon_{ij} = \begin{pmatrix} n_o^2 & 0 & 0 \\ 0 & n_o^2 & 0 \\ 0 & 0 & n_e^2 \end{pmatrix} \quad (3)$$

where $n_o = 2.26$, $n_e = 2.41$ and

$$p = \begin{pmatrix} 0.007 & 0.187 & 0.34 & 0 & 0 & 0 \\ 0.187 & 0.007 & 0.34 & 0 & 0 & 0 \\ 0.09 & 0.09 & 0.24 & 0 & 0 & 0 \\ 0 & 0 & 0 & -0.17 & 0 & 0 \\ 0 & 0 & 0 & 0 & -0.17 & 0 \\ 0 & 0 & 0 & 0 & 0 & -0.046 \end{pmatrix} \quad (4)$$

in Voigt notations. These numerical values are valid for an optical wavelength of 632.8 nm. With an angle $\beta = 28^\circ$, we can evaluate the dielectric constant modifications induced by the quasi-longitudinal ($\Delta\varepsilon|_{ql}$) and quasi-transverse ($\Delta\varepsilon|_{qt}$) waves :

$$\Delta\varepsilon|_{ql} = \begin{pmatrix} -1.78 & 1.16 & 0 \\ 1.16 & -3.02 & 0 \\ 0 & 0 & -2.88 \end{pmatrix}$$

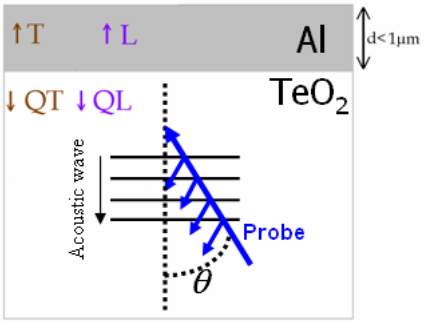

Figure 4: Acousto-optic interaction in the TeO_2 crystal.

$$\Delta\varepsilon|_{qt} = \begin{pmatrix} 1.45 & -0.32 & 0 \\ -0.32 & -3.07 & 0 \\ 0 & 0 & -0.97 \end{pmatrix} \qquad (5)$$

These perturbations give rise to a probe beam scattering by the acoustic waves. Whatever could be the polarization of the incident beam, it can be seen from eqs. (5) that there is always a component of the scattered beam which can interfere with the light beam reflected by the metallic film. These interferences should give oscillations in the transient reflectivity records [7]. The frequency of these oscillations is given by the Bragg selection rule $f = \frac{2v_a n_i}{\lambda}\cos\theta$, where the index a stands for quasi-longitudinal (ql) or quasi transverse (qt) and the index i for ordinary (o) or extraordinary (e) ; θ is the incidence angle of the probe beam on the metallic interface within the transparent TeO_2 crystal ; λ is the optical wavelength (figure 4).

Results

Time resolved reflectivity recorded with probe polarizations perpendicular and parallel to the [001] axis are displayed on Figs. 5 and 7 respectively. Oscillations can clearly be seen on these two plots. The associated Fourier transforms are given in Figs. 6 and 8 where two sharp peaks can be observed. The ratios between the frequency locations of these two peaks are $\frac{24.32}{7.89} = 3.08$, $\frac{25.69}{8.36} = 3.07$ for the two probe beam polarizations and these values are very close from the ratio $\frac{4.27}{1.42} = 3.01$ between the sound velocities of the quasi-longitudinal and quasi-transverse waves. Furthermore the frequency shift related to the polarization change between the two experiments is in very good agreement with the difference between ordinary and extraordinary optical indices. Thus these peaks in the Fourier spectrum clearly evidence the presence of both a quasi-transverse and a quasi-longitudinal wave propagating in the transparent crystal. The mode conversion process appears to be efficient at least in a transmission configuration. We

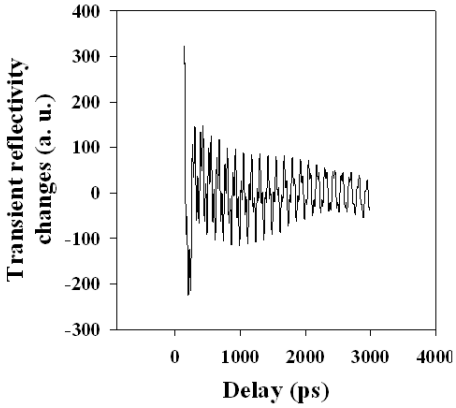

Figure 5: Brillouin Oscillations with the polarization probe normal to the [001] crystal axis (ordinary index $n_o = 2.26$).

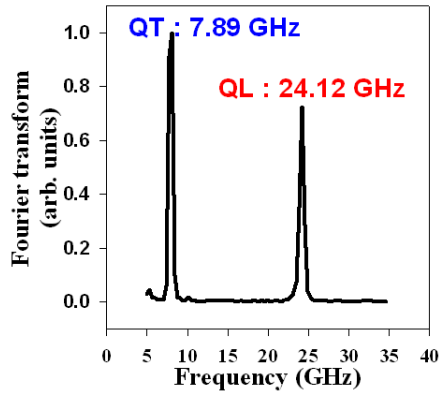

Figure 6: Fourier transform of Brillouin's Oscillations (Figure 5)

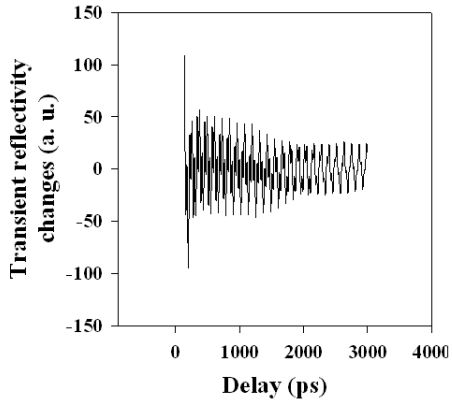

Figure 7: Brillouin Oscillations with polarization probe parallel to the [001] crystal axis (extraordinary index $n_e = 2.41$).

also investigated the mode conversion process in a reflection configuration. For this experiment, pump and probe optical beams were focused on the free surface of the aluminum film. Figure 9 shows the obtained re-

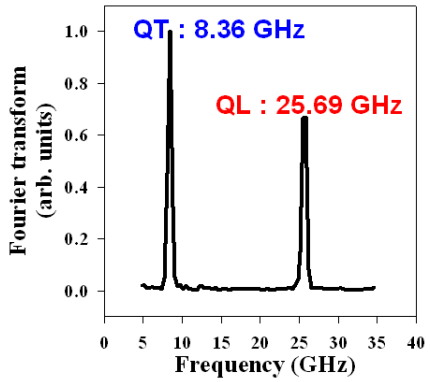

Figure 8: Fourier transform of Brillouin's Oscillations (Figure 7)

flectance variation. The first echo at 48 ps corresponds

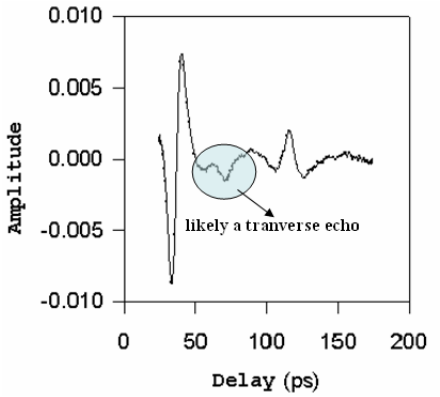

Figure 9: Relative reflectivity change with a likely transverse echo between two longitudinal echoes.

to the first longitudinal wave. The second echo could be ascribed to a transverse wave. Its arrival time is about 80 ps which could be consistent with the theoretical expectation $\frac{d}{v_t} \simeq 80$ ps, where $d = 300nm$ is the aluminum film thickness. The third echo corresponds to the second longitudinal echo. The "transverse" echo shape appeared strongly dependent on the location where the experiment was performed on the metallic film surface ; this dependence could be due to the crystalline structure being more or less affected by the polishing process.

Conclusion

To demonstrate the excitation and the detection of a quasi transverse acoustic pulse using picosecond laser ultrasonics, we used mode conversion occurring during the transmission of a longitudinal pulse between an isotropic polycrystalline aluminum film and an anisotropic TeO_2 crystal. The presence of a quasi transverse wave in the crystal is clearly evidenced by a photoelastic detection in this transparent material. In contrast, the mode conversion of the longitudinal pulse in a transverse pulse by reflection at the interface seems very dependent on the quality of the crystal surface.

References

[1] R.M. White, J. Appl. Phys. 34, pp.3359, 1963.

[2] C.Thomsen, H.T. Grahn, H.J. Maris, J. Tauc, J.J. Hauser, Phys. Rev. Lett. vol53, 1986.

[3] B. Perrin, in "Systèmes Femtosecondes", edited by P. Laporte and F.Salin, Saint-Etienne University publications, p 65-89, 2001.

[4] Y. Sugawara, O.B. Wright, O. Matsuda, M. Takigahira, Y. Tanaka, S. Tamura, V.E. Gusev, Phys. Rev. Lett. 88, 2002.

[5] Hurley D.H., Wright O.B., Matsuda O., Gusev V.E., Kolosov O.V., Ultrasonics vol 38, pp.470, 2000.

[6] D. Royer, E. Dieulesaint, Ondes élastiques dans les solides. Masson, Paris, 1996.

[7] H.-L. Lin, R.J. Stoner, H. J. Maris, and J. Tauc, J. Appl. Phys., 69 :p. 3816, 1991.