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Theory and illustration of the three-dimensional elasto-optic interaction in picosecond ultrasonics

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Abstract. The three-dimensional (3D) photoelastic interaction involved in the detection mechanism of picosecond ultrasonics is investigated in micrometric metallic films. In pump-probe experiments, the laser source beam is focused to a spot size less than $1\ \mu\text{m}$. A 3D diffracted acoustic field is generated at high frequencies of several tens of gigahertz, containing longitudinal and shear waves altogether. Their propagation changes the dielectric permittivity tensor and the material becomes optically heterogeneous. Consequently, the detection process is modeled through the propagation of the laser probe beam in a material with dielectric properties varying in all directions. Thus, the solving of Maxwell equations leads to a differential system, the source term of which is proportional to the acoustic field itself. The scattered electromagnetic field is described using the matricant and the ensuing analytical solution allows then analyzing the 3D photoelastic interaction. Good agreement with experiments performed in a $1\ \mu\text{m}$ thick aluminum film is found.

1. Introduction

Picosecond ultrasonics allow the generation and detection of acoustic waves in nanostructures, by means of ultra-short laser pulses. Until recently, only longitudinal waves could be generated through a thermoelastic process. However, in order to access more information about the sample's elasticity, transverse acoustic waves are of great interest. The generation of plane transverse waves has been achieved pumping through a transparent layer directly inside an anisotropic medium with tilted axis [1, 2]. When focusing the laser beam through a microscope objective, the lateral size of the generation laser spot can be reduced, and a localized thermal source is created. While relaxing, the latter launches a three-dimensional (3D) diffracted acoustic field. Thus, due to mode conversion at an interface, diffracted shear waves were observed by interferometry in a strongly absorbing material [3]. In this case, the probe light is almost entirely reflected from the free surface and only its phase is modified by the surface displacement [4]. As the optical penetration increases, the probe beam senses the in-depth optical variation due to the strain propagation. To model the reflectometric signals, the interaction between the laser probe beam and the 3D acoustic field must be studied. A semi-analytical model is here developed to account for such phenomena.

The acoustic field is described by a 3D equation of motion coupled with thermal diffusion [5]. The propagation of the elastic perturbation into the bulk creates a local deformation, and changes the optical properties of the sample, through the linear acousto-optic interaction [6]. When introduced in Maxwell's equation, the change of dielectric properties leads to a differential

system with varying coefficients. Using the theory of small perturbations, the space dependence of these coefficients can be neglected [7]. In Sect. 2 of this paper, attention is drawn on the matrix formalism of the resulting linearized equations in the Fourier domain. Then, this differential system is solved using the matricant method [8]. Finally, in Sect. 3, the calculated results are compared with the experimental data.

2. Elastically induced transient optical heterogeneity

From the impulse response to a laser line source, a point source can be reconstructed. Therefore, the system formed by the thermal and mechanical equations is written in cartesian coordinates (x_1, x_2, x_3) for a 2D line source focused on the surface along the x_3 direction. A free isotropic film, the thickness of which is along x_1 , is modelled. To obtain ordinary differential equations, a spatiotemporal Fourier transform is applied in the (x_2, t) space. The associated dual space is referred as (k_2, ω) . Considering the thermoelastic process [9] as the dominant generation phenomenon in low-intensity radiation interaction with metals, the analytical expression of the spectrum of the strain tensor $[\eta(x_1, k_2, \omega)]$ is calculated analytically using the method described in a recent paper [5]. The propagation of the acoustic field changes the optical properties of the sample. The variation $[\varepsilon^s]$ of the dielectric tensor $[\varepsilon]$ can be related to the strain thanks to the fourth order photoelastic tensor $[P]$ through the linear acousto-optic interaction [6]:

$$[\varepsilon(x_1, k_2, \omega)] = \varepsilon^h[I] + [P] : [\eta(x_1, k_2, \omega)] = \varepsilon^h[I] + [\varepsilon^s(x_1, k_2, \omega)] = \varepsilon^h[I] + \begin{bmatrix} \varepsilon_1^s & \varepsilon_6^s & 0 \\ \varepsilon_6^s & \varepsilon_2^s & 0 \\ 0 & 0 & \varepsilon_3^s \end{bmatrix} \quad (1)$$

In absence of any acoustic perturbation, the dielectric matrix is given by its homogeneous component $\varepsilon^h[I]$, where $[I]$ is the identity matrix.

The perturbed dielectric tensor (1) is now introduced in Maxwell's equations to describe the propagation of the probe pulse. The homogeneous electric and magnetic fields $\mathbf{E}^h$ and $\mathbf{H}^h$ are separated from the scattered fields $\mathbf{E}^s$ and $\mathbf{H}^s$. In the frame of linear acoustics, the amplitude of the elastic strain remains small. Therefore, the formalism of the problem can be reduced to the following system by applying the perturbation theory [7]:

$$\begin{aligned} \nabla \wedge \mathbf{E}^h(x_1) - jk_0 \mu \mathbf{H}^h(x_1) &= 0 \\ \nabla \wedge \mathbf{H}^h(x_1) + jk_0 \varepsilon^h \mathbf{E}^h(x_1) &= 0 \\ \nabla \wedge \mathbf{E}^s(x_1, k_2) - jk_0 \mu \mathbf{H}^s(x_1, k_2) &= 0 \\ \nabla \wedge \mathbf{H}^s(x_1, k_2) + jk_0 \varepsilon^h \mathbf{E}^s(x_1, k_2) &= -jk_0 [\varepsilon^s(x_1, k_2)] \mathbf{E}^h(x_1) \end{aligned} \quad (2)$$

where μ and k_0 are the magnetic permittivity and the optical wave number in vacuum, respectively. This system is written under the following matrix form:

$$\begin{aligned} \frac{\partial \mathbf{f}^h(x_1)}{\partial x_1} - [A^h] \mathbf{f}^h(x_1) &= 0 \\ \frac{\partial \mathbf{f}^s(x_1, k_2)}{\partial x_1} - [A^s(k_2)] \mathbf{f}^s(x_1, k_2) &= [a(x_1, k_2)] \mathbf{f}^h(x_1) \end{aligned} \quad (3)$$

with:

$$\mathbf{f}^h(x_1) = \begin{Bmatrix} E_3^h \\ H_2^h \\ E_2^h \\ H_3^h \end{Bmatrix}, \quad [A^h] = jk_0 \left[\begin{array}{cc|cc} 0 & \mu & 0 & 0 \\ \varepsilon^h & 0 & 0 & 0 \\ \hline 0 & 0 & 0 & -\mu \\ 0 & 0 & -\varepsilon^h & 0 \end{array} \right] = \begin{bmatrix} [A_3^h] & 0 \\ 0 & [A_2^h] \end{bmatrix} \quad (4)$$

$$\mathbf{f}^s(x_1, k_2) = \begin{Bmatrix} E_3^s \\ H_2^s \\ E_2^s \\ H_3^s \end{Bmatrix}, \quad [A^s] = jk_0 \left[\begin{array}{cc|cc} 0 & \mu & 0 & 0 \\ \varepsilon^h - k_2^2/k_0^2\mu & 0 & 0 & 0 \\ \hline 0 & 0 & 0 & k_2^2/k_0^2\varepsilon^h - \mu \\ 0 & 0 & -\varepsilon^h & 0 \end{array} \right] = \begin{bmatrix} [A_3^s] & 0 \\ 0 & [A_2^s] \end{bmatrix}$$

$$[a] = jk_0 \left[\begin{array}{cc|cc} 0 & 0 & 0 & 0 \\ \varepsilon_3^s & 0 & 0 & 0 \\ \hline 0 & 0 & -k_2\varepsilon_6^s/k_0\varepsilon^h & 0 \\ 0 & 0 & -\varepsilon_2^s & 0 \end{array} \right] = \begin{bmatrix} [a_3] & 0 \\ 0 & [a_2] \end{bmatrix} \quad (5)$$

The upper-left 2x2 matrices $[A_3^h]$, $[A_3^s]$ and $[a_3]$ represent the propagation of the optical mode polarized along the axis $\mathbf{x}_3$, whereas the lower-right 2x2 matrices $[A_2^h]$, $[A_2^s]$ and $[a_2]$ stand for the polarization in the plane $(\mathbf{x}_1, \mathbf{x}_2)$. As the other submatrices are null, there is no coupling between these two modes, and no rotation of the scattered polarization with respect to the incident one is to be expected. The upper-left matrix system will be denoted with the index $p = 3$, and the lower-right system with $p = 2$. Thus, the solving of the problem is split to study separately the influence of each polarization p . The system (3) is then rewritten to manipulate smaller 2x2 matrices:

$$\frac{\partial \mathbf{f}_p^h(x_1)}{\partial x_1} - [A_p^h] \mathbf{f}_p^h(x_1) = 0$$

$$\frac{\partial \mathbf{f}_p^s(x_1, k_2)}{\partial x_1} - [A_p^s(k_2)] \mathbf{f}_p^s(x_1, k_2) = [a_p(x_1, k_2)] \mathbf{f}_p^h(x_1) \quad (6)$$

with

$$\mathbf{f}_p^q = \begin{Bmatrix} E_p^q \\ H_{5-p}^q \end{Bmatrix} \quad (7)$$

where the index $q = h, s$ stands for the homogeneous and scattered solutions, respectively. The differential systems (6) are then solved using the matricant [8], and the change of reflectivity r_p^s is obtained for each polarization $p = 2, 3$. Finally, r_p^s is multiplied by the spectrum of the functions defining the spatial distributions of intensity along $\mathbf{x}_2$ of the pump and probe pulses. A combination of the change of reflectivity r_p^s for the p polarization is made to determine the variation of scattered amplitude in the plane $(\mathbf{x}_2, \mathbf{x}_3)$. A point source generation is then obtained by applying a Hankel transform to the spectrum of the response to a line source. Finally, to return in the time domain, a numerical inverse Fourier transform is performed over ω .

3. Theoretical and experimental results

Experiments are carried out with a classical reflectometric pump and probe setup on an isotropic aluminum film of thickness $d \approx 900$ nm, deposited on a mylar substrate. The acoustic wave is generated by the absorption of radiation of a Ti:Sapphire laser at the wavelength $\lambda = 796$ nm. The pump beam passes through a doubling crystal to divide its wavelength by 2. Both beams are focused through a $\times 100$ magnification microscope objective to a spot on the surface of the film. The spatial distributions of intensity for pump and probe beams are described by Gaussian functions, the FWHM of which define the diameters of the spots on the surface $\chi_b \sim 0.5 \mu\text{m}$ and $\chi_r \sim 1 \mu\text{m}$, respectively. Reflectometric measurements are plotted in Fig. 1(a) for several pump-probe positions. The thermal background, due to the slow relaxation of the material, is removed by subtraction of a polynomial. From their times of arrival, the longitudinal pulses which have performed one and two round trips inside the material, namely $2L$ and $4L$, can be

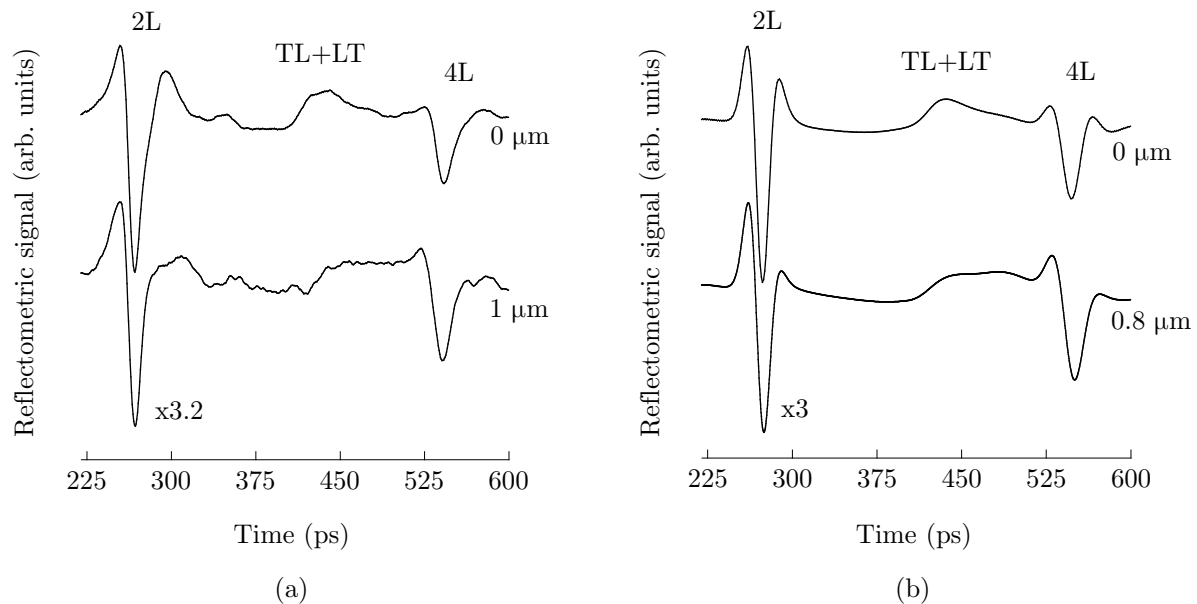


Figure 1. (a) Measured and (b) calculated reflectometric signals in a 900 nm thick isotropic aluminum film deposited on a Mylar substrate for several pump-probe distances.

identified. The time shift between the two corresponds to the ratio $2d/v_L \approx 281$ ps, with the longitudinal velocity in polycrystalline aluminium $v_L \approx 6.4$ nm/ps [10] and the thickness of the film $d \approx 900$ nm. An additional echo can also be observed between the two longitudinal ones. This echo, containing shear information, is the sum of two waves TL and LT. They arise from the conversion of the longitudinal waves on the front and back surfaces of the film, respectively.

As the acoustic impedance of the Mylar substrate is small, a free isotropic aluminium film is modelled, using the values given in a previous paper [3]. The photoelastic coefficients $P_{11} = -10 + 5j$ and $P_{12} = -10 - j$ are adjusted to match the experimental results. As the material is isotropic, it is assumed that $P_{66} = (P_{11} - P_{12})/2$. Calculations are performed for different pump-probe distances, as plotted in Fig. 1(b). Very good agreement with experimental data can be observed. The acoustic diffraction, together with the detection of shear waves is fully described by the model.

Using this theoretical approach, the optical detection of the 3D acoustic propagation in reflectometric measurements has been represented. This work could be applied to the elastic characterization of opaque materials by reflectometric measurements. To a greater extend, the acoustic imaging of objects buried in transparent films could be performed.

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