Picosecond acoustic diffraction in anisotropic thin film (µm); application to the measurement of stiffness coefficients

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Abstract. Investigation of thin metallic film properties by means of picosecond ultrasonics has been under the scope of several studies. Generation of longitudinal and shear waves with a wave vector normal to the film free surface has been demonstrated. Such measurements can not provide complete information about properties of anisotropic films. Acute focusing of the laser pump beam (≈0.5 µm) on the sample surface has recently allowed us to provide evidence of picosecond acoustic diffraction in thin metallic films (≈1 µm) such as aluminum, gold, copper. Waveforms have been experimentally recorded in a gold layer (2 µm thick) for several distances between pump and probe on the sample surface. Due to acoustic diffraction, the acoustic wavefronts propagate at a group velocity which differs from phase velocity in the anisotropic film. However, a specified signal processing allows us analyzing the space repartition of the acoustic wave-vectors for both longitudinal and shear waves. Four stiffness coefficients of the anisotropic gold layer could thus be recovered accurately, demonstrating the feasibility of the measurement.

1. Introduction

The non-contact and nondestructive measurement of anisotropic properties of layers with thickness in the µm range is receiving growing attention for solid state physics as well as for mechanical engineering. Nanosecond laser-ultrasonic has shown that measuring group or phase velocities in several wave propagation directions allows the determination of stiffness tensor coefficients of a plate with thickness in the millimeter range [1]. Similar measurements in picosecond time domain have yet not been performed. Actually, since the end of the 1980s, plane longitudinal waves have been applied successfully to analyze nanometric layers [2-4]. Recently, the acoustic diffraction was obtained by acute focusing of the laser beams and transverse waves were observed owing to the reflection of longitudinal waves with mode conversion at the film interface. Doing so, the thermoelastic generation of shear acoustic waves, using femtosecond laser pulses in submicrometric isotropic aluminum films, was demonstrated [5].

In this paper, a gold micrometric film is considered. The picosecond acoustic wave generation in this material has already been analysed. It was shown that the electronic diffusion process was the main photo-acoustic source[6]. In addition, stiffness tensor of submicrometric gold film was determined from analysis of Brillouin scattering [7].
Table 1: Reference stiffness tensor coefficients (GPa) predicted with a Voigt’s homogenization and measured values.

|       | Voigt $C_{11}$ | $C_{12}$ | $C_{22}$ | $C_{66}$ |
|-------|----------------|---------|---------|---------|
| Voigt | 227            | 142     | 218     | 24      |
| Measured | 240±1    | 145±8   | 219±15  | 22.6±0.3 |

The film features will be firstly presented in the following with the experimental setup, and the generated acoustic waves will be commented. Notably, acoustic diffraction will be putted into relief. Then, a signal synthesis suitable to recover stiffness coefficients will be presented. Afterwards, an accurate evaluation of these coefficients will be achieved from analysis of phase arrival times.

2. Experimental results

The gold film was prepared by radio-frequency sputtering on a [100] silicon substrate. The nominal film thickness is 2.1 µm and the RMS roughness of the film has been measured by the AFM technique at about 1 nm. The film is composed of columnar crystallites with a diameter around 100 nm. The crystalline structure of the film was observed by X-Ray diffractometer. It shows that the film consists of [111] nano-crystals oriented normally to the substrate. The axes normal to [111] are uniformly oriented in the plane of the film surface.

The experiments were performed by means of a standard pump-probe picosecond set-up [4]. The shapes of pump and probe spots on the surface are measured with a knife-edge beam profiler with 0.1 µm resolution and can be represented by a Gaussian function. Their sizes at half height are measured to 0.5 and 1 µm respectively. Comparing the spot sizes to the diameter of the columnar crystallites, the sample can be considered as an homogeneous material with respect to the acoustic propagation, and acoustic diffraction at the column boundaries can be neglected. The sample is therefore considered with a transverse isotropic symmetry. Let axis $x_1$ lie along the direction normal to the sample surface. Axis $x_2$ stands along any direction included in the isotropic surface. A first evaluation of the stiffness tensor coefficients is thus performed with a Voigt’s homogenization to provide reference values. These values are reported in the first line in table 1.

The normal component of the surface displacement is optically measured since these acoustic perturbations are probed by the technique of beam deformation detection [4,8]. Then, in order to measure the arrival times of acoustic waves for several crystallographic directions, the pump spot position is displaced along the $x_2$ direction with a constant step of $\delta x_2 = 0.2 \mu m$. A set of 47 signals is recorded from the epicenter position. The measured waveforms include the signature of the bulk longitudinal waves propagating along one (2L) or two (4L) paths forth and back through the film thickness. The signature of the quasi-shear waves resulting of the reflection of longitudinal waves with mode conversion ($LT_q$) can also be detected [5].

3. Synthesis of conical waves

The waveforms are very rich as they show the signature of numerous waves with different natures and several ray paths. However, waveforms interpretation is not a trivial matter since acoustic energy travels at a group velocity that differs from phase velocity owing to anisotropy. Thus, interest is now turned to the numerical synthesis of the signal $s(t)$ that would be experimentally obtained if an array of laser point sources was used [9]. The set of $N=47$ source positions equally spaced with the constant step $\delta x_2 = 0.2 \mu m$ is considered, accordingly with sampling conditions in Ref. 9. A constant time delay $\delta t$ is applied between two successive signals. Therefore, the source slowness along direction $x_2$, $\delta t/\delta x_2$, fixes the projection of wave vectors in this direction. The so synthesized waves represent conical wavefronts which the axis is direction $x_2$. The angle of each conical refraction can merely be tuned continuously by conveniently choosing the delay $\delta t$. These bulk conical waves are reflected at the free surface with or without mode conversion giving rise to several conical wave fronts with axes
parallel to the direction $x_2$. Others waves are synthesized, such as surface waves resulting from the coherent combination of surface waves directly generated by each point source.

The signal synthesized for a source slowness along the surface $\delta t/\delta x_2$ of 0.075 s/km is shown in figure 1. On account of stiffness tensor coefficients, the source is super-ultrasonic for the two acoustic modes. The longitudinal waves ($2L, 4L, \ldots$) are clearly visible in the synthesized waveform. The signatures of the quasi-shear waves resulting of the reflection of longitudinal waves with mode conversion ($LT_q, 3LT_q$) can also be detected.

![Figure 1](image1.png)

**Figure 1**: signal synthesized for a source slowness $\delta t/\delta x_2$ along the surface of 0.075 s/km.

**Figure 2**: the points correspond to the phase arrival time obtained by the processing of signals synthesized from experiments. The solid lines represent the theoretical phase arrival time for measured stiffness tensor (Table 1).

### 4. Stiffness tensor coefficients identification

The signal synthesis is applied to the set of experimental signals. By tuning the value of parameter $\delta t$, a set of synthesized signals $s(t)$ is thus calculated providing conical waves for several wave vector directions distributed in a wide angular sector. Then, signal processing [9] allows the accurate measurement of the phase arrival times. The results are shown with points in the figure 2.

Let us now describe the stiffness coefficients identification from phase arrival time data. For wave vectors included in a principal plane of a material showing hexagonal symmetry, say $(x_1, x_2)$, and whatever the number of reflections, the number of mode conversions and the substrate properties, the phase arrival time can be written under the form:

$$t_{ph} = \left(n_L k_L^1 + n_T k_T^1\right)/\omega$$

where $\omega$ is the angular frequency, $h$ stands for the thickness, $n_L$ (respectively $n_T$) is the number of paths between two reflections with the quasi-longitudinal polarization $L$ (respectively quasi-transverse polarization $T_q$) and $k_L^1$ (respectively $k_T^1$) the component along $x_1$ of the quasi-longitudinal wave vector (respectively quasi-transverse polarization). The components $k_L^1$ and $k_T^1$ are related to the source slowness along the surface $\delta t/\delta x_2$ through the Christoffel equation.

Then, quadratic sums of the differences between the theoretical and the experimental phase arrival times over several source slownesses $\delta t/\delta x_2$, are minimized for each identified wave ($2L, LT, \ldots$). The minimization solves an over determined system of non-linear equations whose the unknowns are the four stiffness coefficients. The stiffness tensor coefficients obtained from the minimization of the functional are given in the last line in table 1 together with their 90% confidence intervals [10]. The confidence intervals are not representative of the exact errors since possible systematic errors associated with the slowness measurements are not taken into account. However, they are indicative of the uncertainty on the stiffness coefficients estimated from the set of measured phase slownesses.

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Journal of Physics: Conference Series 92 (2007) 012028 doi:10.1088/1742-6596/92/1/012028
measured stiffness coefficients are in good agreement with reference values predicted with the Voigt homogenization.

The phase arrival times calculated with these coefficients are plotted with solid lines in the figure 2. It is worth noting that a large amount of information about the quasi-transverse mode was obtained after signal processing. This result is in agreement with the one obtained in the reference [5], since the reflection of quasi-longitudinal waves allows the transfer of an important part of energy towards the quasi-transverse waves. Moreover, the information about quasi-transverse waves is sought for many reasons with the waves generated or detected with a quasi-longitudinal polarization. Firstly, the longitudinal waves are more efficiently generated than the transverse waves because of the generation mechanism. Secondly, quasi-longitudinal waves contribute more to the detected normal displacement than the transverse waves since the longitudinal polarization is mainly oriented normally to the sample surface when several reflections occur. So the information about quasi-transverse waves is accessible by means of the waves having undergone at least one reflection with conversion of polarization. However, the more there is reflections, the less crystal directions are investigated. As consequences, the identification of coefficient $C_{66}$ will result from the arrival times of the waves $LT_q$ and $3LT_q$, which correspond to the waves having undergone 1 and 3 reflections and one path with the quasi-transverse polarization. The three coefficients $C_{11}$, $C_{22}$ and $C_{12}$ will be mainly obtained from the quasi-longitudinal waves reflected once. The recovered values of the stiffness coefficients are close to their reference counterparts (Table 1). Note that stiffness coefficients $C_{11}$, $C_{22}$ and $C_{66}$ are obtained with good accuracy. Since sensitivity of phase velocities to stiffness coefficients is the least for off-diagonal coefficients, maximum relative error is for the off-diagonal coefficient $C_{12}$.

5. Conclusion
Picosecond ultrasonics has allowed nondestructive and non-contact stiffness characterization of a 2.1 micrometers thick gold film. Firstly, the point source was scanned along the film surface to provide acoustic signatures. Secondly, the signals deriving from point sources were shifted in time and summed to synthesize bulk conical waves that would have been recorded if an array had been used. The coefficients of the stiffness tensor were measured from the phase velocities with a good accuracy with experimentally recorded signals.

Acknowledgements
The authors are grateful to T. Pezeril for the help in preparing the 2.1 µm gold layer.

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