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Surface acoustic wave characterization of optical sol–gel thin layers

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ABSTRACT

Controlling the thin film deposition and mechanical properties of materials is a major challenge in several fields of application. We are more particularly interested in the characterization of optical thin layers produced using sol–gel processes to reduce laser-induced damage. The mechanical properties of these coatings must be known to control and maintain optimal performance under various solicitations during their lifetime. It is therefore necessary to have means of characterization adapted to the scale and nature of the deposited materials. In this context, the dispersion of ultrasonic surface waves induced by a micrometric layer was studied on an amorphous substrate (fused silica) coated with a layer of ormosil using a sol–gel process. Our ormosil material is a silica–PDMS mixture with a variable polydimethylsiloxane (PDMS) content. The design and implementation of Surface Acoustic Wave InterDigital Transducers (SAW-IDT) have enabled quasi-monochromatic Rayleigh-type SAW to be generated and the dispersion phenomenon to be studied over a wide frequency range. Young's modulus and Poisson's ratio of coatings were estimated using an inverse method.

1. Introduction

High power lasers can damage optical components. Laser Induced Damage (LID) is characterized by craters of a few microns that appear mainly on the exit surface of the beam in fused silica when irradiated with a powerful laser beam, especially at 350 nm. The CEA (Atomic Energy and Alternative Energies Commission) suggests developing coatings on laser transmission optics to mitigate shock waves that can modify and increase the density of fused silica inducing an increase in absorption that explains the rapid increase in damage after repeated laser pulses [1,2]. To achieve this objective, a wide range of hybrid materials based on a mixture of silica and PDMS has been developed using sol–gel processes. These coatings have interesting optical and mechanical properties for the target application. Several formulations of these hybrid materials have been optimized to produce optical coatings for lasers. In order to classify these thin layers correctly, a novel technique was tested to determine the elastic properties (Young's modulus and Poisson's ratio) of these thin layers 1–2 μm thick. A surface wave dispersion technique will then be used to determine the mechanical properties of these thin layers. SAW-IDTs will be used over a wide frequency range from 10 to 60 MHz to effectively generate surface acoustic waves.

Several techniques can be used to generate Rayleigh-type surface waves. Wedge sensors are traditionally used to generate surface waves, but above 10 MHz the losses and attenuations related to this sensor technology become too significant. Another interesting technique is laser-ultrasonics, which offers numerous advantages such as the possibility of non-contact generation and broadband generation [3,4]. In recent years, several publications have demonstrated the relevance of this method for the characterization of thin films [5–7]. However, depending on the nature of the materials, the suitability of this method of generation varies according to the penetration depth and/or fragility of the layers (problem of ablation). Finally, the acoustic signature is also an interesting technique enabling measurements to be carried out at very high frequency [8]. However, it is essential to work in immersion, which in some cases is not feasible in terms of the integrity of the structure or device to be controlled.

In this study, we designed and implemented SAW transducer. This original solution is based on the development of interdigital transducers to generate quasi-monochromatic surface waves and obtain a rapid and accurate estimation of the phase velocity, key information for the characterization of the layers. Moreover, the use of SAW-IDTs allowed HF (High frequency) surface waves to

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be generated over a broad frequency range [9]. IDT are typically used in acousto-electronic signal processing devices such as surface wave filters, oscillators, and resonators. Today, most SAW applications are in the field of telecommunications and the frequencies used are typically very high and can reach several gigahertz [10]. Acoustic IDTs are used in NDT (Non Destructive Testing) applications, but are usually used at frequencies of a few megahertz [11].

It has already been shown that micrometer layers influence SAW propagation, although for frequencies in the range of megahertz, the SAW wavelengths are well above the micrometer [9,12]. In this study, we show that the micrometric sol–gel layers, with very low Young's modulus compared to the silica, also influence (dispersion phenomenon) SAW propagation. Then, through the study of this dispersion, it was possible to determine, by inversion, some important characteristics such as elastic constants.

One of the advantages of this ultrasonic technique is having SAW attenuation between 10 and 60 MHz that is not too significant. Thus, the SAW can propagate over several tens of millimeters and it is possible to characterize a large area of the sample. In addition, no specific sample preparation is required, and no metal layer is necessary, unlike with femtosecond-based techniques [13]. Therefore, samples can be tested directly with no specific preparation.

2. Sol–gel coatings

The sol–gel layers that have been developed are made with silica and PDMS elastomer. Those two materials have been chosen because of their high transparency especially around the UV wavelength [14], and for their high laser damage threshold. Indeed, silica is one of the best materials to resist to a high energy laser beam. PDMS is a silicon based inorganic polymer. It has a weak absorption coefficient ($5 \cdot 10^{-3}\,cm^{-1}$) and some good heat resistance properties, which are very interesting properties to resist to the laser beam [15,16]. The two materials have similar refractive index (1.41 for PDMS and 1.45 for silica) [14], and PDMS can be associated with silica by a sol–gel synthesis. Indeed, the hybrid silica–PDMS solution is made from the silica precursor, the tetraethylortosilicate (TEOS) and a commercial PDMS solution supplied by Sigma–Aldrich with hydroxyl groups at the end of the polymer chains. The sol–gel reaction begins with the TEOS hydrolysis in which the ethyl groups that ended the TEOS species are switched with hydrogens elements to form silanols Si–OH species. A condensation of the hydrolyzed TEOS occurs under an acid catalysis. We used two types of acid to perform this reaction, the hydrochloric acid (HCl) and the trifluoromethansulfonic acid (TFS). With the hydrolysis and the condensation of the TEOS species, a silica network is formed. The PDMS chains react with the silanols Si–OH groups of the hydrolyzed TEOS or on the surface of the silica network. The two reactions occur simultaneously, but the PDMS reaction with the silica species is not always total. Indeed, when the PDMS amount increases, some PDMS chains remain free in the organic network, which give to the high PDMS loads some viscoelastic properties. Meanwhile, weaker, autonomous and reversible hydrogens bonds can be created between the silanols and the oxygen elements of the PDMS chains. Using those two elements, the viscoelastic properties and the reversible hydrogens bonds, we aim at giving to the layers some self-healing properties.

Once the sol–gel reaction is made, a transparent and homogenous solution is obtained, and a maturing step of 4 days is made to wait for the stabilization of the species in solution. It is the maturing step used after synthesis. Indeed, after this step, the solution can be used to make coatings. The sol–gel solution has a very weak viscosity after the reaction (<5cP), but a gelling of the solutions occurs after 40 days of maturing. With a maturing time of 40 days after the synthesis, the solution tends to gelify. The solution is coated before gelling on silica polished substrates by dip or spin coating techniques. The used technique to make the materials used in this study is spin-coating, but dip coating is also possible to make silica–PPDM thin layer. Once the thin layer is obtained, the sample is dried and a heat treatment is made at 120 °C to activate the chemical bounds between the silanols surfaces groups of the substrate and the sol–gel species of the layer. A solid silica–PDMS hybrid thin coating with strong adhesion to the substrate is obtained. The transmission spectra show that the silica–PDMS coating give to the sample some higher transmission values, with the presence of Fresnel's interference that is typical for antireflective layers properties (Fig. 1). The analysis of that interference is useful to determine the refractive index of the layer and the substrate with the Fresnel laws. Once the refractive index of the layer is known, the optical and real thickness can be calculated.

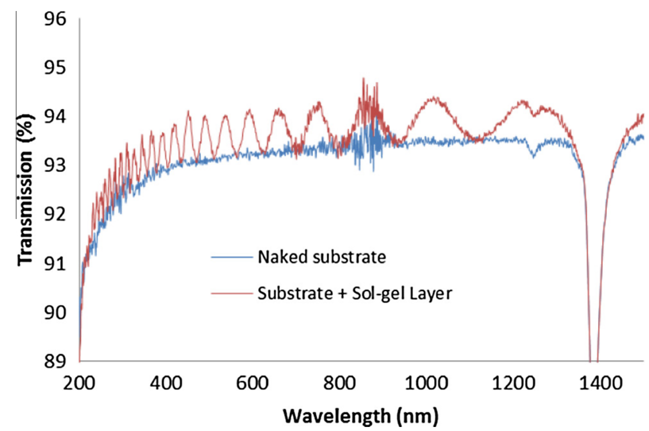

Fig. 1. Transmission spectra of a naked silica substrate and the same substrate with a silica–PDMS layer.

Density values of the layers are necessary to find the mechanical properties by surface acoustic wave characterization. At first, we estimated the sol–gel layers density with a mixture model. The density is 970 kg/m^3 for the PDMS [13] and 1920 kg/m^3 for the polymeric silica [17] in which we considered the internal porosity. Meanwhile, the literature shows that for higher amount of PDMS in the material, the structure of the hybrid is coarser with a higher internal porosity [13]. Thus, there is an uncertainty on the density of the layer at high PDMS ratio (30–40%w) that has been detected by a variation of the refractive index values between two layers on its transmission spectra. The density characterization techniques on massive materials were not fit to measure thin layers material on its substrate. For this reason, secondly, we analyzed the layers with density values that have an uncertainty of 5% and 10%. The different parameters of the layer that have been analyzed are presented in Table 1.

The unordered structure of polymeric silica confers its total isotropy and high homogeneity on a macroscopic scale. At ultrasound scale, glass appears homogenous and isotropic. The density of the silica is 2201 $kg\,m^{-3}$ and the polymeric silica porosity made by sol–gel is 12.9% [13] therefore its density was 1921 $kg\,m^{-3}$. The Young's modulus E and Poisson's ratio υ are given in the Ref. [18] (E = 73GPa and υ = 0.16).

3. Dispersion of SAW in sol gel layer on substrate structure

When a Rayleigh-type SAW propagates on the surface of a homogenous material its energy is concentrated within a thickness of about one wavelength beneath the surface. When this wave