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A different way of performing picosecond ultrasonic measurements in thin transparent films based on laser-wavelength effects

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We present a way of using picosecond ultrasonics to simultaneously get the thickness and elastic properties of thin dielectric layers. This is based on the use of a blue probe which is shown to improve the detection of acousto-optic oscillations in the dielectric from which the sound velocity can be measured from the refractive index. At the same wavelength a strong response of the silicon is used to detect the arrival of the acoustic pulse. We apply this scheme to various materials deposited on silicon substrates. © 2005 American Institute of Physics. [DOI: 10.1063/1.1929869]

In picosecond ultrasonics ultrashort optical pulses are used in a pump and probe scheme to make ultrasonic measurements at very high frequency (up to several hundred gigahertz).¹ In an absorbing sample, a first optical pulse (the pump pulse) generates a thermal strain pulse whose extension can be less than 10 nm. The resulting wave packet of longitudinal phonons propagates in the sample normal to the surface at the corresponding sound velocity. The returning echoes are detected at the surface by another delayed optical pulse (the probe pulse) via an acousto-optic interaction acting along the absorption length. By adjusting the delay between pump and probe pulses it is possible to get a very precise measurement of the round-trip time from which the sound velocity can be deduced from the thickness of the film or conversely the thickness from the sound velocity.² In the case of a transparent sample, a thin metallic film is deposited onto the sample surface. This so-called transducer layer serves first as an absorbing layer for the generation of the acoustic pulse; second the metal should be chosen to also provide a sensitive detection, which means presenting a strong acousto-optic interaction at the probe wavelength. Aluminum (Al) is the best choice commonly retained due to its performance in both mechanisms.

If part of the probe beam passes through a transparent layer its interaction with the acoustic pulse can lead to oscillations in the measured signal whose period is given by $T = \lambda / (2nv \cos \theta)$.³ Here λ is the probe wavelength, n is the index of refraction of the layer at this wavelength, v is the sound velocity, and θ is the angle of incidence. These so-called Brillouin oscillations are the result of interference between the probe light that is reflected from the interfaces of the sample and the light partially reflected from the strain pulse. A few conditions are required to observe such oscillations. First, the layer should be at least partially transparent at the probe wavelength. Second, the probe beam should reach the concerned layer. Third, an acousto-optic interaction should act in this layer. The major interest in Brillouin oscil-

lations is the possibility of extracting the sound velocity independent of the film thickness. Until now the mentioned conditions have been satisfied by chance in different dielectric or semiconductor samples.³⁻⁷ It has mainly involved samples in which the transparent layer was deposited on an absorbing film. Such a geometry fully satisfies the second requirement.^{4,6,8} Here we propose to use wavelength conditions to force the detection of Brillouin oscillations.

Recently we established that the acousto-optic interaction involved in picosecond ultrasonics is strongly dependent on the laser wavelength.⁹⁻¹¹ Here we show that such a dependence can be used to satisfy the third condition even in the usual geometry where a thin metallic layer is deposited at the top of the sample which widens the applicability of picosecond ultrasonics. We apply this to thin dielectric films deposited on silicon (Si) whose elastic properties and thicknesses are much different. As we will see, another wavelength effect that was recently reported in the blue range in Si is used to detect the arrival of the strain pulse in the substrate.¹¹ This way both the thickness and the sound velocity of thin transparent layers deposited on Si can be extracted in the usual geometry with no need for usual acoustic echoes.

The experiment is based on a conventional pump and probe setup associated with a tunable Ti:sapphire laser. Details can be found elsewhere.¹¹ All the samples are composed of a transparent layer deposited on a (100) Si substrate. Details are given in Table I. A very thin layer of Al (100–150 Å) was evaporated onto each film in order to generate the picosecond acoustic pulses. All the studied materials are useful for microelectronic applications: amorphous silicon oxide (SiO₂), fluorinated silicate glass (FSG), silicon nitride (Si₃N₄), aluminum nitride (AlN), and a carbon-incorporated silicon oxide (SiOC). One should notice that the mechanical properties of the involved materials are strongly different: a Si₃N₄ layer is expected to have a Young's modulus greater than 150 GPa whereas a SiOC layer should present a Young's modulus lower than 10 GPa.

Figure 1(a) reproduces the transient reflectivity signal obtained in sample (1) using the usual experimental conditions, i.e., both pump and probe pulses are centered at the

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TABLE I. The data presented here were obtained in experiments performed with a pump wavelength of 804 nm and a probe wavelength of 402 nm. The Young's moduli have been obtained assuming a Poisson's ratio equal to 0.17 for each sample.

Sample	Material	Nominal thickness (nm)	Brillouin period (ps)	Silicon arrival (ps)	Refractive index	Sound velocity (mm/ps)	Thickness (nm)	Density (g cm^{-3})	Young's modulus (GPa)
1	Si_3N_4	850	11.2	99.5	2.08	8.63	858	3.1	215
2	SiO_2	500	22.8	86	1.48	5.95	514	2.2	72
3	SiO_2	1200	22.7	202.5	1.48	5.98	1212	2.2	73
4	AlN	2000	8.5	191	2.16	10.96	2090	3.2	357
5	FSG	500	27.2	106	1.46	5.09	540	2.2	53
6	FSG	2000	27.0	431	1.46	5.10	2200	2.2	53
7	SiOC	500	61	257	1.45	2.29	589	1.4	6

same wavelength (804 nm). The signal is first composed of a jump at zero time, which is the electronic contribution to the pump-probe signal. A few picoseconds later, the pump energy is converted into heat, which diffuses on a larger time scale leading to a slow decrease of the reflectivity. In the first picoseconds the measurement shows an oscillation, which corresponds to a small ringing of the Al film (period close to 3.3 ps corresponding to a thickness of 106 Å). Figure 1(a) also shows an acoustic echo that appears near 203 ps from which we can calculate the sound velocity following the usual procedure of picosecond ultrasonics and assuming a nominal thickness for the film. One should note that the time of arrival of the echo corresponds to the round trip of the acoustic pulse through both the transducer and the transparent layers. Thus a precise measurement of the ringing period is needed to get a precise measurement of the sound velocity in the transparent layer. As a consequence a well-adapted transducer, which would facilitate the penetration of the acoustic energy in the film, is not favorable for fine measurements since the ringing would then disappear.

Figure 1(b) reproduces the signal obtained in the same sample using the same infrared pump but a blue probe. Electronic, thermal, and acoustic contributions are strongly affected by the change in the probe wavelength. Figure 1(b) also reveals strong oscillations: the first starts at $t=0$ and has the lower frequency ($T \approx 11$ ps). The second starts near 100 ps and has a much higher frequency ($T \approx 5$ ps). Both oscillations result from acousto-optic interactions acting, respectively, in the Si_3N_4 layer and Si substrate. This is supported by the value of the measured periods and their starting time:

in Si_3N_4 the oscillations start at $t=0$ as expected since the acoustic pulse penetrates this layer immediately after the generation and the high frequency oscillations appear when the strain pulse penetrates the Si substrate. Both oscillations can be well reproduced by a numerical calculation based on an analytical model (details may be found in Ref. 11) visible in Fig. 1(c). Furthermore the model can reproduce both experimental results using the same mechanical parameters for the Si_3N_4 layer, literature parameters for Al and Si,^{12,13} but different piezo-optic couplings for each experiment. This point confirms that between 800 and 400 nm of these couplings strongly vary in both materials.

Following our interpretation the starting time of Brillouin oscillations in Si exactly corresponds to the time of flight of the acoustic pulse in the transparent layer. It should be noted that it is not exactly equal to the half time of arrival of the echo detected in the first experiment. The measured delay of an echo detected in Al corresponds to a round-trip of the acoustic pulse in both Al and Si_3N_4 layers. Instead of that, Brillouin oscillations in Si start as soon as part of the acoustic pulse penetrates the substrate, a delay which corresponds to the crossing of the single Si_3N_4 layer. This is supported by a fine comparison of the data presented in Fig. 1: the echo arrives at 203 ps whereas the Brillouin oscillations start at 100 ps. As shown in the following this difference is particularly interesting for precise measurements.

Figure 2 presents the signals obtained in samples (2) and (3), which are composed of a SiO_2 layer of different thicknesses. On both curves one retrieves low and high frequency

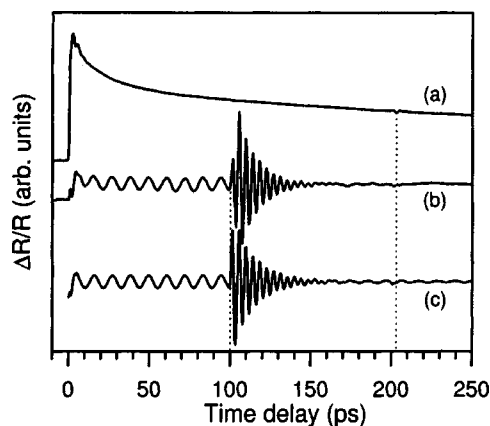


FIG. 1. (a) Experimental result obtained in a Al/ Si_3N_4 /Si sample in picosecond ultrasonics with pump and probe both centered at 804 nm; (b) idem with a probe centered at 402 nm; (c) theoretical simulation of (b).

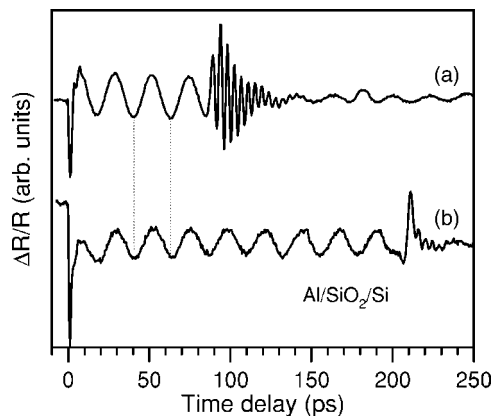


FIG. 2. Experimental results obtained in Al/ SiO_2 /Si samples which differ by the SiO_2 thickness [(a) 500 nm, (b) 1200 nm]. One should note that both low frequency oscillations have the same period and the different starts of the high frequency oscillations.

oscillations as in Fig. 1(b). One first notes that the period of the low frequency oscillations in both curves is close to a 23 ps greater value than that obtained in sample (1) which was close to 11 ps [evident from the comparison of Figs. 1(b) and 2(a)]. This is consistent with our interpretation: the Brillouin oscillations period is related to the $n \cdot v$ product which almost differs by a factor 2 between Si_3N_4 and SiO_2 . The high frequency oscillations have the same period in the three samples as expected since they all are composed of a (100) Si substrate. They start at different delays in samples (2) and (3) which are consistent with the difference of thicknesses of the SiO_2 layers (respectively, 500 and 1200 nm).

Similar signals were detected in all the samples described in Table I and also in others not mentioned here. We present in Table I the periods of the low frequency oscillations and the start of the high frequency oscillations. Each experimental result was obtained using the same laser-wavelength conditions (pump 804 nm and probe 402 nm). As expected the Brillouin period is found to vary between hard and soft materials (60 ps in SiOC and 8.5 ps in AlN). These data can be used to calculate the thickness and the sound velocity of each sample with no need for any echo. The sound velocity was first deduced from the Brillouin period in the dielectric and the index of refraction measured by ellipsometry. Then the film thickness is deduced from the start of the high frequency oscillations.

Let us examine now which advantages this scheme offers. One gets both thickness and sound velocity from the index of refraction. Of course from a metrological point of view it is not a major gain but in most practical cases the index of refraction is known that gives a rough value of the sound velocity and the thickness. It also provides an independence versus the transducer: provided that the film is thin enough to let the probe light go partially through the sample the precise value of its thickness does not play any role in the determination of the thickness of the transparent layer. The full temporal window needed for an experiment is reduced by a factor 2 since we only need to detect the arrival of the acoustic pulse in the substrate and not its return to the surface. One can thus study similar samples quicker or thicker samples with the same delay stage. Finally the method can easily probe samples for which the acoustic impedances of the dielectric and the Si are close. This is not the case for the traditional method since little acoustic energy is reflected at the film/substrate interface.

The Brillouin oscillations are not detected in experiments performed with a red probe (detected in some cases but with a lower amplitude) neither in dielectrics nor in Si. In the last case we have related this to the effect of an electronic interband transition on the piezo-optic couplings.¹¹ In dielectrics it is difficult to involve a similar argument due to the amorphous nature of the layer. Nevertheless, in the use of a blue probe the photon energy is higher and less far from the energy gap of glasses. This explanation also supports some historical observations of Brillouin oscillations in the visible range due to the use of a mode-locked dye laser which delivered pulses centered near 600 nm.⁴⁻⁶

In conclusion, it has been shown that, when one uses a blue probe, a strong acousto-optic interaction acts in many dielectrics that enables one to deduce the sound velocity from the index of refraction of the layer. The thickness of the film is deduced from the strong oscillations which are detected in the substrate in the same wavelength range. This method has been applied to numerous thin films of various elastic properties. As both quantities can be measured without the use of the Al layer in the detection of acoustic waves, different metals can be used as transducer, some of them being better mechanically adapted to the transparent film. Furthermore if one finds an efficient way of launching acoustic pulses in the substrate, picosecond ultrasonics could operate directly in transparent layers that would widen the applicability of the technique.

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