High-laser-wavelength sensitivity of the picosecond ultrasonic response in transparent thin films

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We explore from both the theoretical and experimental points of view the influence of the laser wavelength on the strain pulses detected in thin transparent layers using the picosecond ultrasonic technique. From a theoretical analysis of the displacement detection involved in such experiments, we predict amplitude and sign changes in the detected signals as the laser is tuned. Experimental results performed on various samples and at different wavelengths are found to be in excellent agreement with the theoretical description. Several applications based on the high sensitivity of the phenomenon are finally proposed.

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I. INTRODUCTION

Picosecond ultrasonics is an optical technique that makes ultrasonic measurements up to several hundred gigahertz possible. In order to reach such high frequencies, which are not accessible using the conventional transducers, one makes use of ultrafast laser pulses to generate and detect picosecond acoustic pulses.[1,2] A strong absorption of light pulses can launch acoustic pulses whose spatial extension can be less than a few tens of nanometers so that they lend themselves very well to the characterization of thin films. Picosecond ultrasonics can measure film thickness, sound velocity, and acoustic attenuation in submicrometer-thick films and multilayers.[3–6]

In a picosecond ultrasonic experiment the acoustic waves are usually excited through the absorption of a laser pulse in an absorbing part of the sample (typically a metallic layer). Two mechanisms are involved in the detection of the returning echoes. The first one is an acousto-optic interaction which is based on the modification of the optical properties of materials when they are strained. This modification affects the reflection of the probe beam. A detected echo is not the strain pulse itself but instead a measure of its impact on the optical properties of the film. The interaction between the strain wave and the optical refraction index is measured by the photoelastic couplings. The second mechanism was first introduced by Wright and mainly concerned transparent layers.[7] The strain pulse is detected through the interface displacement it induces.

Recently we have established that the acousto-optic interaction on which is based the first detection mechanism involved in picosecond ultrasonics is strongly dependent on the laser wavelength.[8–10] Phase and amplitude effects related to the acousto-optic interaction have been reported. In particular we have shown that the changes observed in the echo shape can be used to study some of the electronic interband transitions in thin metallic films.[8,9]

Here we present theoretical and experimental results which show that the second mechanism is also very sensitive to the laser wavelength. To the best of our knowledge there has been no report of the high sensitivity of this phenomenon to the laser wavelength. This high sensitivity can be used to improve the measurement precision in picosecond ultrasonics and allows us imagine new applications which are discussed.

The paper is organized as follows. In Sec. II we first present the detection mechanisms involved in picosecond ultrasonics. We then give a theoretical description of the displacement mechanism. This analytical model is shown to predict strong changes in the detected signals as the laser is tuned at different wavelengths. Section III is devoted to the experimental details, the setup, and the design of the studied samples. Section IV contains the experimental results which clearly reveal the strong effect of a change in the laser wavelength. An excellent agreement is found with the theoretical model. Possible applications are then discussed.

II. PICOSECOND ULTRASONICS AND DISPLACEMENT EFFECTS

A. Generation and photoelastic detection in picosecond ultrasonics

In Figs. 1(a) and 1(b) we show a schematic diagram of a picosecond ultrasonic experiment performed in two geometries which differ in the boundary conditions imposed on the photoacoustic generation (absorption at the free surface and in a buried layer, respectively). In both cases, a strain pulse results from the dilatation induced by the absorption of the first optical pulse (the pump pulse). Its extension is related to the absorption length which can be as short as a few nanometers in a metal. As this dimension is much smaller than the spot size (typically a few tens of micrometers in diameter) one can consider basically that only longitudinal waves are excited by the pump pulse. The resulting pulse propagates in the film at the longitudinal sound velocity, typically a few nanometers per picosecond, which explains how absorption can generate picosecond acoustic pulses. The strain pulse produced in the geometry of Fig. 1(a) has a bipolar shape due to the reflection of the strain at the free surface. In Fig. 1(a) [Fig. 1(b)] the strain pulse is reflected onto the film-substrate interface (free surface) and the result-

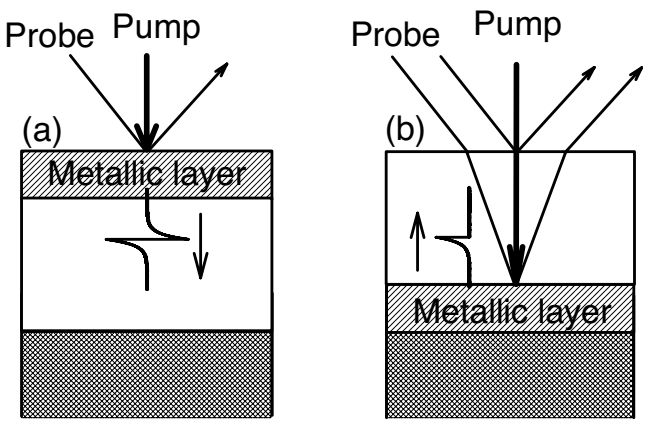

FIG. 1. Schematic diagram of the experiment. The pump light pulse generates a longitudinal acoustic pulse in the metallic layer. (a) The generation acts at the free surface and a bipolar shape strain pulse results. This is the usual case when a transducer is deposited onto the sample surface. (b) The metallic layer is buried so that the strain pulse which propagates in the transparent layer is monopolar (purely compressive). The probe beam causes interferences in the transparent layer as in a Fabry-Pérot interferometer.

ing echo returns to the metallic layer where it modifies in this way the dielectric constant. These optical changes can be detected by another optical pulse (the probe pulse) whose reflection is affected by the presence of the strain wave. By adjusting the delay between pump and probe pulses it becomes possible to monitor the successive echoes due to the strain generated by the pump pulse.

Depending on the chemical nature and on the wavelength some transparent layers can also present an acousto-optic interaction. In that case, the propagation of the strain pulse in this layer manifests itself by oscillations in the reflectivity curve. These so-called Brillouin oscillations are the result of interferences between the probe that is reflected from the interfaces of the sample and the light partially reflected from the strain pulse. Their period can be written as $T=\lambda/(2nv\cos\theta)$.[11] 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. One should note that the geometry of Fig. 1(b) is much more favorable to the detection of such oscillations since a significant part of the probe light penetrates the transparent layer. In spite of this, the metallic transducer usually used at the free surface in picosecond ultrasonics for probing transparent samples can be thin enough to permit such a detection, in particular when judicious wavelength conditions are used for the probe beam.[12]

B. The displacement mechanism and the definition of a reflectivity jump

In the case of a transparent layer another contribution to the reflectivity can be detected. The probe beam is reflected at both interfaces of the layer and these reflections interfere. If the thickness is slightly modified the phase relation between both reflections is modified and so is the resulting intensity of the reflected probe. Due to the interferometric nature of the effect, displacements as small as those carried by picosecond acoustic pulses can be detected. Displacement is the integral of the strain: The compressive part of the pulse (negative strain) acts as a slight decrease of the thickness and the tensile part (positive strain) as an increase. Let us examine the particular case of a monopolar strain that is purely compressive or tensile which thus carries a small thickness change. At the delay which corresponds to the entry of such a pulse in the transparent layer, its reflectivity experiences a sudden change due to the thickness change and keeps this new value as long as this pulse is in the layer (assuming no acoustic attenuation). If one plots the reflectivity of such a sample as a function of time delay one would thus observe a sudden change at the entry of the strain in the transparent layer. In the following, we designate such a discontinuity as a reflectivity jump. Other jumps will be detected at the reflection of the pulse if it is accompanied by a sign change in the strain and also when the pulse will leave the transparent layer.

The usual sample geometry of picosecond ultrasonics in which a thin metallic film is deposited on a transparent layer [Fig. 1(a)] is not favorable to the observation of reflectivity jumps for two reasons. First, a significant part of the probe is reflected or absorbed by the metallic layer so that only a small part of the probe experiences both interfaces of the transparent layer. Second, as the strain pulse generated at the sample surface is bipolar, it induces no thickness change and thus no reflectivity jump. On the contrary if one generates a strain pulse in a buried metallic layer [Fig. 1(b)], the upper layer will experience a pure compressive pulse immediately after the generation (jump at $t=0$). When it reaches the free surface it is converted in a pure tensile strain that leads to another jump. When this pulse exits the layer a third jump is expected. It is important to retain the needed conditions for detecting reflectivity jumps: (1) the probe beam should interact with both interfaces; (2) a jump is observed only if the strain integral is nonzero. Both conditions are satisfied in the case of a transparent layer on an absorbing substrate.

Such a geometry has been used in all the reported studies of such contributions. The first report of such jumps in reflectivity has been done by Wright *et al.* in thin silica films deposited on germanium and chromium substrates.[7] More recently, Rossignol *et al.* have reported similar effects in diamond thin films deposited on metallic substrates.[13] These authors pointed out that this contribution to the detected signal is very sensitive to the film thickness and to the incidence angle of the probe beam. They combined these jumps with the photoelastic contribution to measure the thickness and sound velocity of thin transparent layers.

Due to its nature the displacement mechanism should also be very sensitive to the optical index of the transparent layer and to the probe wavelength. In fact the transparent layer acts here as a Fabry-Pérot interferometer whose reflectivity is very sensitive to the ratio between the thickness of the layer and the wavelength inside the dielectric (λ/n). The strain pulse is detected through the small perturbation it induces on the thickness of the "cavity." In other words the pulse acts as a derivative of the Fabry-Pérot reflectivity. Due to that the reflectivity jumps are expected to be strongly dependent on the thickness, the optical index, and the probe wavelength.

From the previous description one can imagine another origin for the reflectivity jumps. Until now the acoustic pulse acts on the reflectivity through a small change in the layer thickness. But in fact it modifies the *optical path* between the two parts of the reflected probe. As the optical path is given by the product of the thickness by the optical index, a similar effect can appear if the acoustic pulse affects the optical index of the layer. One should recognize in this last condition the definition of the photoelastic effect which is described in Sec. II A. Thus if the transparent layer presents non-null photoelastic constants both contributions have to be taken into account in order to give a complete description of the reflectivity jumps.

C. The analytical model

Here we derive an analytical expression for the reflectivity jumps induced by the acoustic pulses qualitatively described in the previous part. We start from the simplest geometry that permits such effects, which is composed of a transparent layer (thickness e, optical index n) deposited onto an absorbing substrate. This sample acts on the probe beam as a Fabry-Pérot interferometer: The reflected probe results from interferences between the parts which are, respectively, reflected at the free surface and at the layer/substrate interface. One can write the complex reflectivity of such a system as

$$r = \frac{r_{01} + r_{12}\exp(2i\delta/\lambda)}{1 + r_{01}r_{12}\exp(2i\delta/\lambda)}, \tag{1}$$

where r_{01} (r_{12}) designates the reflection coefficient at the free surface (at the interface between the transparent layer and the substrate) and $\delta = ne$ is the optical path of the probe in the transparent layer.

When the strain pulse penetrates the transparent layer it induces a small change in the optical path ($\Delta\delta$). The reflectivity r is then replaced by $r+\Delta r$ and due to the small amplitude of $\Delta\delta$ one can write:

$$\Delta r = \left(\frac{\partial r}{\partial \delta}\right)\Delta\delta. \tag{2}$$

The physical quantity we have to calculate is the relative change in intensity reflectivity ($\Delta R/R$):

$$\frac{\Delta R}{R} = 2\,\mathrm{Re}\left\{\frac{1}{r}\frac{\partial r}{\partial e}\right\}\Delta\delta, \tag{3}$$

where Re{…} denotes the real part.

Combining now Eqs. (1) and (3), we obtain the requested analytical expression:

$$\frac{\Delta R}{R} = 2\,\mathrm{Re}\left\{\frac{1 - r_{01}^2 - 2r_{01}r_{12}e^{2ike}}{(1 - r_{10}r_{12}e^{2ike})(r_{01} + r_{12}e^{2ike})}\right\}\Delta\delta. \tag{4}$$

The small change in the optical path $\Delta\delta$ originates from two effects. First an acoustic pulse induces a small change in the layer thickness given by the integral of the strain pulse η:

$$\Delta\delta^{(e)} = n\Delta e = n\int_0^e \eta(z)dz. \tag{5}$$

One should note that this contribution has a non-null value here due to the sample geometry which leads to the generation of a monopolar strain pulse. Second, a strain pulse located at z induces a local change in the optical index through the acousto-optic effect:

$$\delta n(z) = \frac{\partial n}{\partial \eta}\eta(z). \tag{6}$$

The whole change it induces in the optical path is simply given by

$$\Delta\delta^{(n)} = \int_0^e \delta n(z)dz = \int_0^e \frac{\partial n}{\partial \eta}\eta(z)dz = \frac{\partial n}{\partial \eta}\Delta e. \tag{7}$$

Combining both contributions, we can write the general expression of the change in the optical path as

$$\Delta\delta = \Delta\delta^{(e)} + \Delta\delta^{(n)} = \left(n + \frac{\partial n}{\partial \eta}\right)\Delta e. \tag{8}$$

A rough estimation of Δe gives a few picometers (assuming the absorption of a 1 J/m^2 fluence beam in Al). From Eq. (4) we estimate $\Delta R/R$ in the 10^{-5} range, far above the sensitivity of our setup. As the strain pulse reaches the surface its sign is changed due to its reflection at a free surface. Due to that, the sample experiences a small change in the optical path from $-\Delta e$ to $+\Delta e$ and a jump in the reflectivity curve results. Its magnitude is given by Eq. (4) using $2\Delta e$ in Eq. (8). Concerning the sign of the reflectivity jump we can make two comments. First, according to the sign of the photoelastic constant (positive or negative) both effects have identical signs (opposite signs). Second, the sign of the reflectivity jump at the arrival of the strain pulse at the free surface is given by Eq. (4) ($\Delta\delta^{(e)} > 0$).

D. The wavelength effect

From the analytical expression established in the previous section we can now examine the effect of a change in the laser wavelength on the reflectivity jumps. On Fig. 2 we plotted the relative reflectivity changes corresponding to the jump as a function of the probe wavelength and for different film thicknesses. These curves have been obtained using Eq. (4) in silica thin films ($e = 400$ and 600 nm) and assuming $n = 1.46$. For simplicity, the presented results do not take into account the dispersion in either the dielectric or the metallic layers. We also assume that the pump produces a constant change in the film thickness (Δe).

For the same thickness one should note that the model predicts sign changes in this contribution as the laser is tuned. That means that for the same acoustic pulse, the detected jump can be positive, negative, or null. Around some cancellations the reflectivity changes are smooth, the curve presenting a low rate (for example around 600 nm for both samples on Fig. 2). One can show that they correspond to reflectivity minima. On the contrary, around other cancella-

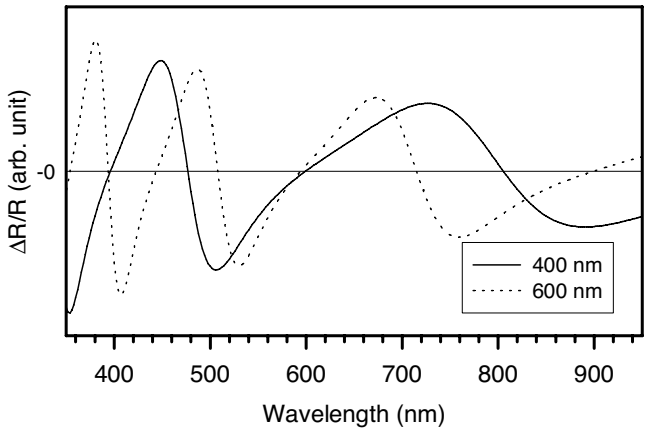

FIG. 2. Theoretical reflectivity change detected at the reflection of the acoustic pulse at the free surface as a function of the laser wavelength for two samples which differ by the thickness of the transparent layer [(a) 400 and (b) 600 nm].

tions the rate is very high (corresponding to the reflectivity maxima). Around such wavelengths the sign of the reflectivity jump is thus expected to be strongly dependent on the laser wavelength. At the same wavelength the comparison between both curves reveals that the sign and the magnitude of the jump are dependent on the film thickness. For both thicknesses one retrieves a similar dependence of the jumps on the wavelength. The theoretical curves seem to be shifted from one to another. The curves also reveal that the sign changes are more numerous in thicker samples or for shorter wavelengths.

One should finally remark that for higher wavelengths the jumps are smaller and the slopes of the curve around cancellations are smoother. This point suggests that the wavelength dependence of reflectivity jumps should be particularly strong in the blue range. We now examine experimental results which confirm all these theoretical results.

III. EXPERIMENTAL DETAILS

A. Experimental setup

The experiment is based on a conventional pump and probe setup associated with a tunable titanium:sapphire oscillator [Coherent MIRA 900f (Ref. 14)]. The laser produces 120 fs optical pulses at a repetition rate of 76 MHz centered at a wavelength adjustable between 700 and 990 nm. The laser output is split to provide pump and probe beams with crossed polarizations. The probe pulse can be delayed with respect to the pump pulse by an optical delay line based on a translation stage. Both beams are focused on the same point of the sample by a 60 mm acromat lens. The maximum incident fluence of the pump beam is 1 J/m^2. To improve the signal-to-noise ratio, the pump beam is chopped using an acousto-optic modulator and the output of the photodiode, which monitors the reflected probe, is amplified through a lock-in scheme.

Most of the experiments were performed with a blue probe obtained by focusing the laser beam into a 1 mm β–barium borate crystal.

TABLE I. Description of the samples. They all are composed of a transparent layer deposited onto a metallic layer, itself on a Si substrate. The table gives for each sample the nature, the thickness, and the optical index of the transparent layer and the nature of the metallic layer.

Sample	Transparent material	Nominal thickness	Optical index (at 400 nm)	Metallic underlayer
A	SiO_2	860 nm	1.48	Al
B	SiO_2	430 nm	1.48	Al
C	Si_3N_4	670 nm	2.08	Al
D	Si_3N_4	500 nm	2.08	Mo
E	Si_3N_4	250 nm	2.08	W

B. Description of the samples

From the previous section two conditions are needed for observing displacement effects in reflectivity measurements. First, the layer should be transparent to the probe beam. Second, the strain pulse should have a monopolar shape, i.e., a purely compressive or a purely tensile pulse. Both conditions are naturally satisfied in samples composed of a thin transparent layer deposited on an absorbing substrate. In that case both pump and probe beams go through the transparent layer and the strain pulse is generated inside the sample and presents thus the needed monopolar shape.

For convenience reasons here we study samples in which a transparent layer is deposited on a metallic film itself deposited on a silicon substrate instead of an opaque substrate. The only difference lies in the possible reflections of the acoustic pulse at the metal/silicon interface. The samples were designed in order to check the sensitivity of the displacement effects with three parameters: the optical index of refraction, the thickness of the transparent layer, and the chemical nature of the metallic layer. Table I gives the composition of the five investigated samples. The optical index depends on the nature of the dielectric layer (SiO_2 and Si_3N_4). For each kind of dielectric we present the results obtained for different thicknesses. The first three samples are composed of a transparent layer deposited on an Al film. Aluminum has been used for two reasons. First, its acoustic impedance matches well the impedance of the transparent layers (in particular SiO_2). Second, it is also well mechanically adapted to the silicon wafer so that the reflection of the acoustic pulse at the Al/Si interface is negligible. On the contrary in sample D (E) the transparent layer is deposited on a W (Mo) film. In both the last cases an acoustic impedance mismatch exists at the different interfaces.

The dielectric layers were deposited using plasma-enhanced chemical vapor deposition. The Al films were evaporated on the Si substrates and the Mo and W films were sputtered using a magnetron.

IV. EXPERIMENTAL RESULTS

A. Identifying the displacement effect

Figure 3(a) reproduces the transient reflectivity signals measured in samples A, B, and C using a 860 nm pump and

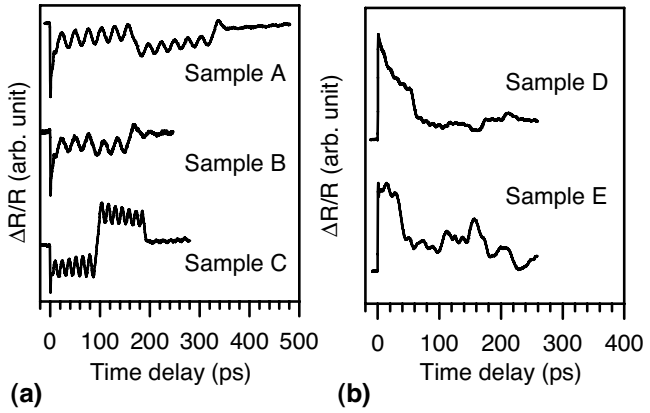

FIG. 3. (a) Reflectivity measurements on samples A, B, and C performed with an 860 nm pump and a 430 nm probe. The oscillation visible here results from an acousto-optic interaction acting in each transparent layer. As the acoustic pulse reaches the free surface a jump appears on the reflectivity curves. Its sign depends on the layer thickness and its optical index. (b) The reflectivity jumps obtained in samples D and E. Due to an important acoustic contrast at the two interfaces, acoustic reflections appear and reflectivity curves that are much more difficult to interpret are detected.

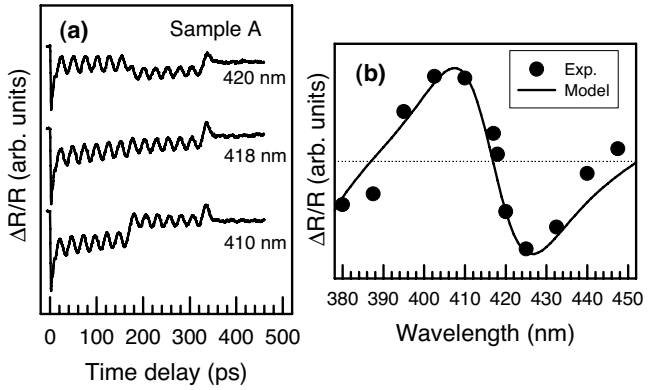

FIG. 4. (a) Reflectivity measurements obtained in sample A for three different laser wavelengths. One should remark the sign change in the reflectivity jump detected at 180 ps. (b) A comparison between the magnitude of the reflectivity jump detected in sample A as a function of the laser wavelength between experimental values and the theoretical model developed in the text. The excellent agreement has been obtained by fitting the SiO_2 thickness.

a 430 nm probe. All three samples are composed of a dielectric layer deposited onto an Al layer. They differ in the nature or the thickness of the dielectric layer (details are given in Table I). The experimental results have points in common. First, one retrieves the electronic contribution of the Al layer to the pump-probe signal at zero delay. They also all present oscillations in the reflectivity curves which are Brillouin oscillations detected through an acousto-optic interaction in the transparent film. The period of these oscillations is found to be dependent on the nature of the dielectric as expected (the period is inversely proportional to the product of the sound velocity and the optical index). On Fig. 3(a) the period of the oscillation is the same for samples A and B which are both composed of a SiO_2 layer, but is different in sample C, composed of a Si_3N_4 layer. One should note that the oscillations disappear after one round trip in the transparent film (for samples A, B, and C, respectively, close to 320, 160, and 200 ps). This is due to the small acoustic contrast at the interface between the dielectric and the Al film. At that time delay one also detects an acoustic echo corresponding to the arrival of the acoustic pulse in the Al layer. The amplitude of this echo is small due to the fact that the photoelastic constants in Al near 400 nm are not so strong as near 800 nm. The experimental signals also present a third acoustic contribution, reflectivity jumps visible at a delay equals to half the echo time position and at the echo delay also. From their delay position we can easily attribute them, respectively, to the reflection of the strain pulse at the free surface and its leaving. One clearly observes in the reflectivity curves that the magnitude and the sign of the jumps are dependent on the sample.

These last contributions are those to which this paper is devoted and in the following we will more particularly focus on those detected at the arrival at the free surface since at these delays no other acoustic contribution appears. According to the theoretical description given in Sec. II B, these jumps are related to changes induced by the strain pulse in the probe optical path. Due to the geometry of the samples, the strain pulse is generated in the buried Al layer and the part that propagates toward the free surface presents a nonnull integral, i.e., the strain pulse induces a small thickness change. Furthermore, the detection of Brillouin oscillations on the same curves shows us that the reflectivity jumps are composed of two contributions, one of them being photoelastic. This point was checked by reproducing numerically the reflectivity curves. The Brillouin oscillations are well reproduced using a real photoelastic constant equal to −0.4, a value in good agreement with the literature value.[15] One should note that the negative sign of this constant means that the reflectivity jumps detected here are smaller than expected by the displacement mechanism alone [see Eq. (8)]. Thus without the photoelastic contribution we would have obtained an overestimated value of $\partial n/\partial \eta$.

On Fig. 3(b) we present experimental results obtained in samples D and E in which the metallic layer is not composed of Al. One retrieves the Brillouin oscillations and jumps in the reflectivity curves. These results present jumps at longer delays which were not detected in samples A, B, and C. This is related to the strong acoustic contrast at the dielectric/metal interface. Due to that a part of the acoustic energy is reflected to the free surface at longer delay. Furthermore, acoustic energy is also reflected from the metal/silicon interface toward the free surface and a part of this energy enters in the transparent layer and produces this way other reflectivity jumps. These results illustrate the generality of the phenomenon and justify the design we retained for samples A–C for obtaining simple reflectivity curves.

B. Evidence of wavelength effects

We now focus on the results presented on Fig. 4(a) and which have been obtained for sample A at three different laser wavelengths. From 410 to 420 nm a sign change is

observed on the reflectivity jump localized before 200 ps. In the case of a 410 nm probe, a positive jump in the reflectivity curve is detected when the acoustic pulse reaches the free surface. On the contrary, at 420 nm the reflection of the same pulse induces a negative change in the reflectivity. One should remark on Fig. 4(a) that the acoustic echo detected in Al at 340 ps is not affected by the wavelength change. This point confirms that the observed effects are not related to a change in the strain pulse itself but instead in the detection mechanism of the reflectivity jumps. With a fine tuning of the laser wavelength, we were able to find a wavelength at which no jump can be detected [418 nm curve on Fig. 4(a)]. One should also note that this last wavelength is very close to the first one and in spite of this the reflectivity jumps detected in the two cases are strongly different. This point demonstrates the high sensitivity of the studied mechanism.

Such an observation of a sign change in the reflectivity jump corresponds to what has been theoretically predicted in Sec. II C. In fact as the laser wavelength is increased from 410 to 420 nm one first detects a positive jump, then a cancellation, and after that a negative jump. One retrieves a similar series in Fig. 2 around what we called a cancellation with a high rate. To go further in the comparison with our theoretical model, we measured the sign and the magnitude of the jump in the reflectivity for many wavelengths and calculated the theoretical curve corresponding to sample A. On Fig. 4(b) we superimpose the experimental points to the theoretical model. Jumps have been normalized to the probe and pump intensities. The experimental points very well reproduce the theoretical shape of the curve. In particular one retrieves an alternation between cancellations with slow and high rates. The best fit visible on Fig. 4(b) has been obtained for a 910 ± 3 nm thick SiO_2 layer, a value slightly different from the nominal value (7% greater) but compatible with the calibration error of the deposition chamber used here. The film thickness can also be deduced from the optical index using the Brillouin oscillation period and the echo delay.[12] Doing that the SiO_2 film thickness in sample A is 908 ± 17 nm. One should note that the uncertainty is much larger than using the method described in this paper. This illustrates how the strong dependence on the laser wavelength can be used for precise thickness measurements of thin transparent layers.

Similar effects have been observed in all the samples listed above and in others not mentioned here. In each case sudden changes in the signs of the reflectivity jumps have been observed as the laser was tuned around particular wavelengths.

C. Discussion

In the case of sample A and in the range presented on Fig. 4(b) we have obtained an excellent agreement between experimental points and the theoretical model developed in Sec. II C. Here we discuss possible improvements which could be introduced to the model especially in the case of a comparison on wider wavelength ranges.

The first point concerns the dispersion of the optical index in all the layers and in particular in the transparent layer. Dispersion effects are expected in the investigation of wide wavelength ranges. For example some of our samples present reflectivity jumps both in the blue and in the near-infrared ranges (typically 370–480 nm and 740–960 nm). In that case a comparison between experiments and theory [as on Fig. 4(b)] from 370 to 960 nm cannot be done without taking into account the dispersion: One can show that the reflectivity jump can be predicted with a wrong sign at 800 nm if one uses an index value determined at 400 nm.

Second, we examine the role of the pump pulse. In the previous part we considered that the pump only generates a strain pulse whose integral is not equal to zero. In particular we assumed that it induces a small change in the thickness of the transparent layer Δe which was supposed to be the same at each laser wavelength. But this quantity should vary with the tuning of the laser source for two important reasons. The optical index of the metallic layer varies with the laser wavelength and with it the strain pulse extension. Second, the transparent layer also acts as a Fabry-Pérot cavity for the pump pulse. The energy which is absorbed in the metallic layer is thus modulated by this effect and the amplitude of the strain pulse is thus expected to be nonconstant over a wavelength range. One can show that the first effect is negligible since, assuming that the pump energy is constant, the change in the absorption will induce a change in the pulse extension but will keep its integral constant. Thus Δe is the same for each wavelength. We have theoretically examined the effect of the modulation of the pump energy induced by the optical resonances of the transparent layer. We found significant differences with or without this effect around particular wavelengths. One should mention that for small wavelength ranges as on Fig. 4(b), the effect is not significant and only concerns the slope of the curve around cancellations. One could reveal the importance of this effect experimentally by comparing experiments performed with a blue or a red pump in the same sample.

V. CONCLUSION

We have presented theoretical and experimental results which demonstrate that the laser wavelength can strongly influence the detection of strain pulse in thin transparent layers in picosecond ultrasonics. We started from a theoretical description of the mechanism: the strain pulse generated in a buried layer carries a small change in the optical path of the probe beam in the transparent layer. Two different origins have been identified for the change in the optical path. First, in each transparent layer the strain pulse carries a small change in the layer thickness. Second, if the transparent layer presents a non-null photoelastic coupling, the strain pulse also modifies the optical index of the layer as it goes through it. The completeness of this description has been tested by adjusting the photoelastic coupling of the transparent layer from the comparison of different acoustic contributions. Then we explored the sensitivity of these jumps to the probe wavelength. Sign changes were predicted and observed experimentally. We obtained excellent agreement between the theoretical model and the experimental values.

These wavelength effects could find several applications. They mainly concern transparent thin layers for which they

can be used to improve the precision of measurements (thickness or mechanical measurements). The influence of dispersion has also been discussed and this suggests that the high sensitivity of the phenomenon can also be used to measure the dispersion of the transparent layer.

In this paper we have focused on samples composed of a transparent layer deposited on an absorbing layer. Such a geometry has been retained for several reasons. First, it permits one to generate strain pulses which carry thickness changes. Second, the reflectivity curve jumps due to the transparency of the first layer. It is important to note that the effects revealed here can also concern many other samples. In fact these so-called displacement effects will appear as soon as the probe beam experiences both interfaces of a layer. For example a very thin metallic layer acting as a transducer at the top of a sample can also experience displacement effects. For sufficiently thin films, the effect presented here is expected to influence the wavelength effects we have previously reported on photoelastic detection in metals.[8,9]

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