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Strong effect of interband transitions in the picosecond ultrasonics response of metallic thin films

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We report on the strong laser wavelength effects in the shape of acoustical pulses generated and detected in thin metallic films using ultrashort optical pulses. The experiments have been performed on aluminum and copper. In both cases, we have observed a sign change of the acoustical part in the detected signals in the vicinity of a well-known interband transition. We identify the origin of the phenomenon in the detection mechanism. These results let us demonstrate the role picosecond ultrasonics may play in the study of the electronic structure of thin films.

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I. INTRODUCTION

Thomsen *et al.*¹ have first shown that ultrashort optical pulses can generate picosecond acoustic pulses, which lead us to study the acoustic or thermal properties of thin films.^{1,2} With this so-called picosecond ultrasonic technique, it is possible to generate acoustic waves whose frequency is so high (up to several hundred GHz) that they lend themselves very well to the characterization of thin films. This technique has been used to access thicknesses, sound velocities, and attenuation measurements using ultrasonics in a frequency range that is inaccessible by conventional techniques.³ It has also motivated fundamental studies concerning the elastic properties of multilayers,^{4,5} electronic diffusion, and electron-phonon coupling in metals.^{6,7} We have recently suggested that picosecond ultrasonics can also play a role in the study of electronic interband transitions.⁸ We have reported a strong effect of the laser wavelength on the shape of detected echoes. Complementary experiments and a photoelastic model have let us ascribe this effect to an interband transition that acts on the detection mechanism. This pioneering work involved a tungsten film, and the observed changes were consistent with the 790 nm transition that falls in the wavelength range (750–900 nm) studied. However, the electronic structure of tungsten is quite complicated so it was not possible to find theoretical and experimental data that could confirm this interpretation.

The purpose of this paper is to prove that such an explanation was true. For that we study the shape of picosecond acoustic echoes detected in several metals as a function of laser wavelength around well-known interband transitions. We start from a detailed analysis of the detection mechanism of a picosecond ultrasonic experiment. This analysis reveals how the laser wavelength can affect the shape of echoes. We also draw a parallel between picosecond ultrasonics and modulation spectroscopy. We use these theoretical considerations to define the best metals to test our interpretation: we retain aluminum and copper whose electronic structure is well known and for which there exist modulation spectroscopy results. Section III presents the experimental details about the laser source and the design of the samples. In Sec. IV, we present the results of our experiments performed on both metals. As in the tungsten case, we report a strong de-

pendence of the detected echo shape on the laser wavelength. Complementary experiments show that the effect is closely related to the detection mechanism. That is supported by a photoelastic model that reproduces the detected echoes well provided that the piezo-optic couplings are strongly affected by the concerned interband transitions. This completes the demonstration that picosecond ultrasonics may play a role in the study of electronic properties of thin films.

II. PICOSECOND ULTRASONICS AND INTERBAND TRANSITIONS**A. Picosecond ultrasonics**

In a picosecond ultrasonic experiment, a first optical pulse (the pump pulse) is incident at the sample surface where it is absorbed and the resulting dilatation generates a strain pulse whose extension is related to the absorption length. In the particular case of a metal, absorption can be very strong, giving a length of a few nanometers. Such a mechanical pulse propagates into the sample at the longitudinal sound velocity (typically a few nanometers per picosecond), which explains how absorption can generate picosecond acoustic pulses. This pulse is reflected onto the film-substrate interface and the resulting echo returns to the surface and modifies in this way the dielectric constant of the film. These changes can be detected by another optical pulse (the probe pulse) whose reflection or transmission 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. Usually, these echoes are used to measure thicknesses of thin films, sound velocities, attenuation, and so on.³

B. The detection mechanism and its connection to modulation spectroscopy

Here we focus on the detection mechanism and on the physical parameters that affect the shape of detected echoes. As explained in the preceding section, the strain pulse modifies the optical properties of the film, and when it arrives near the surface this modification affects the reflection of the probe beam. It is important to notice that a detected echo is not the strain pulse itself but instead a measure of its impact

on the optical properties of the film. One thus distinguishes two major contributions to the echo shape: first, the penetration of the probe, which is measured by the optical index (real and imaginary parts, n and k , respectively); second, the interaction between the strain wave and the optical index, which is measured by the piezo-optic couplings. In general the change induced by the strain on the dielectric constant is a fourth-rank complex tensor. But in the particular case of an isotropic solid with longitudinal waves only two parameters are needed. Here we retain the first derivatives of the real and imaginary parts of the index of refraction with respect to the strain field ($\partial n/\partial\eta$ and $\partial k/\partial\eta$, where η designates the strain field). The optical properties mainly affect the extension of the echo. In contrast, we have shown that the piezo-optic couplings strongly influence the qualitative shape of echoes.⁸

An important point to notice is the derivative nature of these coefficients. It lets us bring together picosecond ultrasonics and modulation spectroscopy. In a modulation spectroscopy experiment, the band structure of a solid is modulated by means of an externally applied field and the resulting changes in the optical properties are detected synchronously.^{10,11} Piezomodulation is a particular modulation technique that uses an acoustic wave generated by a transducer to modulate the electronic structure of the studied sample. Even if both techniques have nothing in common, we see that picosecond ultrasonics and piezomodulation are responsive to the same piezo-optic couplings. These experiments take advantage of their derivative nature to probe interband transitions in the electronic structure of the solid. The basic idea behind this is that an interband transition produces a small irregularity on the optical index of a solid but a discontinuity of the first-derivative response, which should be much more easy to detect.⁹

Let us come back to the important points to keep in mind. First, the shape of the detected echoes in a picosecond ultrasonic experiment is governed by the piezo-optic couplings. Second, due to their derivative nature these coefficients can strongly vary in the vicinity of an interband transition. This sensitivity was used by modulation spectroscopy to probe the electronic structure of solids. Picosecond ultrasonics should thus be sensitive to interband transitions in that important changes on the echo shape should be detected when the photon energy of the probe beam is close to the energy of these transitions.

C. Testing this explanation

We have tested the sensitivity of picosecond ultrasonics to interband transitions by studying the shape of acoustic echoes generated and detected in thin aluminum films and copper films, which gives the reasons for the selection of metals. First, both metals have much simpler electronic structures than tungsten. In particular, each has a well-known interband transition for which modulation spectroscopy results exist. Furthermore, these transitions are accessible with our laser source.

Concerning the band structure of aluminum, it is described well by a simple nearly-free-electron model.¹² It is therefore one of the simplest theoretical situation among the

metals. The optical properties of Al can be described well using such a model.¹³ It was found that the only interband transitions that lead to significant structure in the dielectric constant were those that occur around W and Σ in the vicinity of K in the Brillouin zone and that these produce a peak near 1.5 eV. These conclusions are in agreement with optical reflectivity and absorption measurements. Modulation spectroscopy has also been used to study in detail this interband transition.¹⁴ In particular, thermoreflectance measurements have revealed the sensitivity of such measurements to this interband transition by showing a sudden inversion of the detected signal near 1.5 eV (830 nm). So in order to test our explanation we have to perform experiments between 800 and 900 nm where significant changes in the echo shape are expected. There is no difficulty to probe at the required wavelength since it falls in the titanium:sapphire range.

Concerning the choice of copper, as with any monovalent noble metal, its optical properties have been extensively studied. Despite having a more complicated electronic structure than aluminum the optical properties of noble metals are those best understood.¹² Very good agreement has been obtained between theoretical and experimental studies. In particular, it has been established that an interband transition appears in copper near 2.15 eV (577 nm) due to transitions from the top of the d band to the Fermi surface. It should be noted that modulation spectroscopy has also been used to study this transition in detail.^{11,14} As we will see in the following such a wavelength can be obtained using a femtosecond optical parametric oscillator. This is why we have chosen to test our interpretation on a copper sample.

III. EXPERIMENTAL DETAILS

A. Experimental setup

The experiment is based on a conventional pump and probe setup¹ associated with a tunable titanium:sapphire oscillator [Coherent MIRA 900f (Ref. 15)]. The laser produces 120 fs optical pulses at a repetition rate of 76 MHz centered at a wavelength adjustable between 700 nm 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 50 mm lens. The maximum incident fluence of the pump beam is 1 J/m². The ratio between the pump and probe intensities is close to 1000:1. 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. Fluctuations in the probe intensity were normalized by splitting the probe into signal and reference beams and monitoring the difference between the signals detected in identical photodiodes. Such an experimental setup allows us to detect signal changes $\Delta R/R$ of a few 10^{-7} , which is crucial in the investigation of the precise structure of acoustic echoes.

In the following we focus on the shape of acoustic echoes and its change with the wavelength of the laser. The improvement of the tunable femtosecond laser sources has

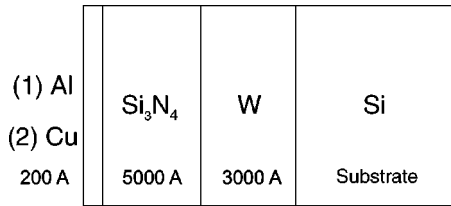


FIG. 1. Schematic diagram of the samples. The first layer is a very thin metallic layer that serves as a transducer since it generates and detects very short acoustic pulses. It is composed of the studied metal aluminum in one case and copper in the other.

greatly simplified the production of very short pulses whose wavelength can be easily changed. In the aluminum case, the pump and probe beams were centered at the same wavelength, which ranged from 800 to 920 nm. Such a wavelength range is easily reached with a titanium:sapphire oscillator. The situation is much more complicated in the copper case. In order to see the interband transition, we need to probe the acoustic signal with femtosecond pulses centered around 570 nm. We obtained such a wavelength by using an optic parametric oscillator (OPO) synchronously pumped by the titanium:sapphire laser.¹⁶ It is based on a collinear, non-critically phase-matched process in a Potassium Titanium Oxide Phosphate (KTP) crystal. This OPO converts the near infrared beam (700–850 nm) into infrared output beam (1000–1200 nm) composed of pulses whose duration is less than 150 fs and with the same repetition rate. The visible wavelength we need to study for the copper sample are easily obtained by doubling the infrared output of the OPO using a 2 mm β -barium borate (BBO) crystal.

Complementary experiments were also performed using a blue probe. In that case, the BBO doubling crystal was inserted into the oscillator beam to get a probe centered at 400 nm.

B. Design and preparation of the samples

In the case of tungsten we have studied a thin film deposited on a silicon substrate. Aluminum and copper have in common a great electron diffusion that perturbs the generation of short acoustic pulses.^{6,7} The light of the pump pulse is absorbed by the electrons of the metallic film that lose their energy to the lattice after few picoseconds. In the case of aluminum and noble metals, it has been shown that during that time the excited electrons are able to carry their energy to distances beyond the absorption length before they release it to the lattice. Very large strain pulses result, which disallow the fine analysis of echo shape we want to do. To avoid this, we have designed a multilayer structure for which a schematic diagram is given on Fig. 1. First we limit the thickness of the metallic layer in order to avoid the diffusion of electrons during the formation of the acoustic pulse. It is deposited on a transparent layer so that there is no generation after the limit of the metallic film. Here we have chosen amorphous silicon nitride (Si₃N₄) because it has a greater acoustic impedance than silicon dioxide, which is better suited for the high acoustic impedance of copper. It is deposited onto a tungsten layer in order to take advantage of the

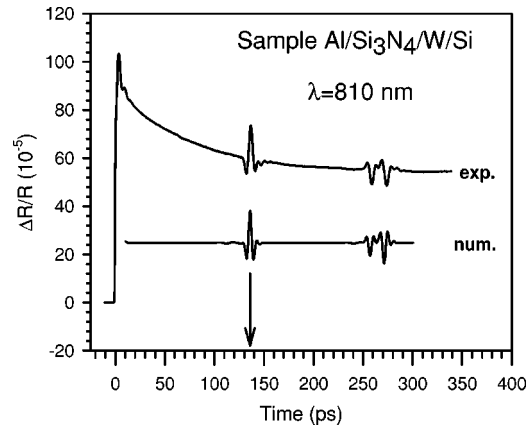


FIG. 2. Experimental and numerical results obtained in the aluminum sample. The experiment was performed using a pump and a probe, both centered around 810 nm.

reflection of the pulse (tungsten has a very high acoustic impedance). The whole structure is deposited onto a silicon substrate.

The results we present here involve samples that were composed of the same layers except that the first one is designated as the transducer. In the first case it is an aluminum layer that has permitted us to study the acoustic echo shape around 850 nm. In the second case it is composed of copper and was studied around 570 nm. Both samples only differ by the chemical nature of this first layer. A 3000 Å thick film of polycrystalline tungsten was deposited onto a silicon substrate using magnetron sputtering. A film of amorphous silicon nitride of thickness of 5000 Å was then grown on the tungsten film by using plasma-enhanced chemical-vapor deposition (PECVD). A 220 Å thick film of polycrystalline aluminum has been evaporated on the silicon nitride. A 250 Å thick polycrystalline copper film was deposited using radio frequency sputtering. Copper samples were studied a few days after the deposition in order to avoid oxidation effects.

IV. RESULTS

A. The aluminum case

Figure 2 reproduces the transient reflectivity signal measured in the aluminum sample. Both pump and probe pulses are centered at 810 nm. The signal is first composed of a jump at zero time. The pump pulse is first absorbed by the electron gas of the film, which induces a strong change in the dielectric function of the surface that affects its reflectivity. A few picoseconds later, this energy is converted into heat, which diffuses on a larger time scale. This leads to a slow decrease of reflectivity. In the first picoseconds after the $t = 0$ jump, the data of Fig. 2 have an oscillatory acoustic component. There exists an acoustic mismatch between Al and Si₃N₄ so that part of the heat deposited by the pump excites the resonance of the Al layer. The frequency is given by $v_{Al}/2d_{Al}$, where v_{Al} is the acoustic velocity of aluminum and d_{Al} is the film thickness. In the present case these oscillations have a period close to 6–7 ps, as expected. Figure 2 also shows several acoustic echoes. The first one appears

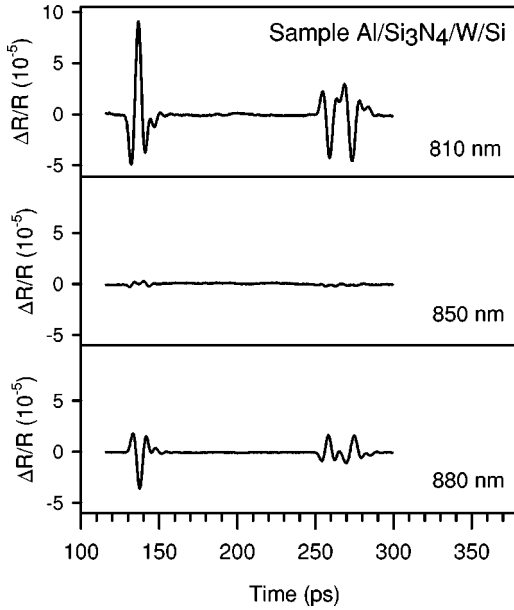


FIG. 3. Acoustic echoes obtained in a picosecond ultrasonic experiment performed in aluminum at three different laser wavelengths. The same scale has used for the three signals and the thermal backgrounds have been subtracted in each case. One should note the sign change of the acoustical contribution between 810 and 880 nm. One should also note its cancellation around 850 nm.

near 135 ps and corresponds to one round-trip of the acoustic pulse in the nitride silicon film: a sound velocity of 7.5 nm/ps (Ref. 17) in Si_3N_4 leads to a time of flight of $500 \times 2 / 7.5 = 133$ ps.

Two other echoes appear around 270 ps. They have an opposite phase compared to the previous echo. The last corresponds to a second round-trip of the strain pulse in the Si_3N_4 film. Its time of arrival is given by $2 \times 135 = 270$ ps. Its phase is opposite to the first due to the greater impedance of the tungsten compared to the nitride: the reflection is in-phase at the $\text{Si}_3\text{N}_4/\text{W}$ interface and out-of-phase at the air-film interface. The first structure that appears a few picoseconds before 270 ps results in reflection of the strain pulse at the interface W-Si. It corresponds to a round-trip in both Si_3N_4 and W films. One round trip in the tungsten film takes $2 \times 300 / 5.2 = 115$ ps [the sound velocity of tungsten is 5.2 nm/ps (Ref. 18)]. It gives a time of arrival of $135 + 115 = 250$ ps. The acoustic reflection coefficient at this interface is here negative. That explains the opposite phase of this echo compared to the echo visible at 135 ps.

Figure 3 reproduces the echoes detected in the same aluminum sample using different laser wavelengths (810, 850, and 880 nm). For each wavelength we have used the same experimental configuration with the same pump and probe intensities. The comparison of the echoes amplitude is thus meaningful. Between 810 and 880 nm one clearly observes a sign change in the detected echoes with respect to the first echo near 135 ps and also the other structures near 250 ps. One should remark that all of these signals are detected in the aluminum transducer. As in the tungsten case, we have performed complementary experiments using a frequency-doubled beam (a blue beam) to confirm the role of the detection mechanism in the present observation. Using a blue pump and a red-infrared probe, we have retrieved the same change in the sign of echoes. This last point clearly demonstrates that the observed effect is related to the detection mechanism. In the physical description we gave of the experiment the only way to reproduce such a change in the echoes sign is to modify the piezo-optic coefficients between 810 and 880 nm (see Sec. II).

To go further in this analysis, we have used a photoelastic model to reproduce the shape of detected echoes. As the sample is composed of several layers it is quite complicated to use an analytical model, as was done in the tungsten case.⁸ Here we use a numerical model that is based on the same physical model but that permits us to resolve much more complicated problems by taking into account the generation and the detection of acoustic waves in different layers. Figure 2 also reproduces the best fit of detected echoes at 810 nm, which we have obtained using this numerical model. This was obtained using literature values for all physical constants (given in Table I) except for the thicknesses of all layers, the density, and the sound velocity in silicon nitride and the piezo-optic constants. It reproduces the experimental signal well if we assume a smaller density of silicon nitride than usual and a tungsten layer larger than supposed. The echoes obtained at 880 nm can also be reproduced numerically using the mechanical parameters deduced from the 810 nm case, the literature values of optical index, and a fitting of the piezo-optic values. We have found that the piezo-optic couplings need to be changed to reproduce the experimental data well. Simulations confirm thus that the wavelength effect on the echo shape can be attributed to a strong change on the piezo-optic couplings with the laser wavelength.

Following the discussion of Sec. II, we attribute these changes in the piezo-optic couplings to the interband transition located near 830 nm. That is confirmed by two other

TABLE I. Values of physical parameters that have been used in the numerical simulation of acoustical echoes detected in copper and aluminum. The optical index values are given for 810 nm except for the last line, which is for copper at 580 nm.

		Al	Cu	Si_3N_4	W	Si[100]
Sound velocity	(nm/ps)	6.4	3.6	7.5	5.2	8.3
Density	(g/cm^3)	2.7	8.9	2.0	19.3	2.33
Optical index	(810 nm)	$2.7 + i8.3$	$0.3 + i5.3$	2	$3.5 + i2.8$	3.7
	(580 nm)		$0.8 + i2.6$			

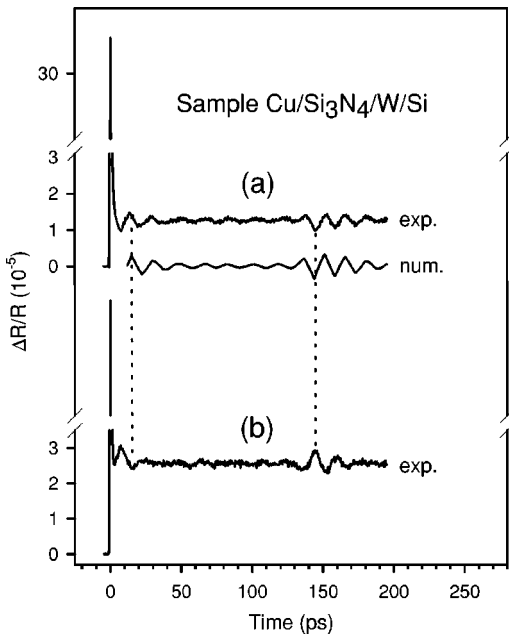


FIG. 4. Experimental and numerical results obtained in the copper sample at different laser wavelengths: (a) $\lambda_{\text{pump}} = 750$ nm, $\lambda_{\text{probe}} = 580$ nm, (b) $\lambda_{\text{pump}} = 810$ nm, $\lambda_{\text{probe}} = 555$ nm.

points. First, similar effects have been observed in many other aluminum samples in the same wavelength range. Second, this explanation is supported by a comparison with previous modulation spectroscopy results. Rosei and Lynch¹⁴ have also reported a sign change in the reflectivity of their aluminum sample near 850 nm.

As the change in the echo sign appears continuously there exists a wavelength for which no acoustical information is detected. Figure 3 also gives the experimental result obtained at 850 nm, a wavelength that almost realizes this condition. It should be noted that for such a wavelength the whole reflectivity signal (electronic and thermal contributions) has a comparable amplitude to those obtained at 810 nm but no acoustical information is detected.

To conclude, we have reported important changes in the detected echoes in the vicinity of the 1.5 eV (830 nm) interband transition. That point confirms the finding that an interband transition can strongly affect the signals detected in picosecond ultrasonics. We have exhibited a wavelength for which no acoustic echo can be detected in aluminum. This point is of particular importance in picosecond ultrasonics since aluminum is currently used as a transducer; around 850 nm, acoustic pulses could be generated but not detected.

B. The copper case

Figure 4 reproduces the experimental signal obtained in the copper sample. It has been obtained using two different wavelengths for the pump and the probe. The probe is taken from a frequency-doubled output of the OPO (555 nm–580 nm), whereas the pump is a part of the output of the femtosecond oscillator (750 nm–810 nm). Due to this complicated way of obtaining the pump and probe beams, these data have a substantially lower signal-to-noise ratio.

One first retrieves an electronic contribution to the change in the reflectivity of the sample, which manifests itself by a strong and very short peak at $t=0$. In the few picoseconds later one observes an oscillatory part due to the ringing of the transducer. It is due to the important acoustic mismatch between Cu and Si_3N_4 so that a part of the heat deposited by the pump excites the resonance of the Cu layer. One retrieves the difference of the sound velocities between aluminum and copper in the fact that the present oscillations have a much lower frequency than in the case of Fig. 2. One also observes a structure near 140 ps. It is the acoustic echo corresponding to a round-trip of the strain pulse in the Si_3N_4 layer, which is coherent with the previous results obtained in the aluminum sample that is composed of the same layers (the small difference in the time of arrival is due to the difference between the sound velocity of both metals). Its shape strongly differs from the aluminum case due to the difference of the optical and piezo-optical properties of both metals, as mentioned in part Sec. II B. In particular, we notice an important oscillatory part which can also be related to the greater acoustic mismatch between copper and nitride silicon. This is confirmed by a numerical result also presented in Fig. 4, which was obtained using the parameters extracted from the aluminum case and using the literature values of mechanical and optical constants of copper (see Table I). Numerical and experimental results are in very good agreement. This comparison confirms the possibility of producing short acoustic pulses in copper in spite of its great electronic diffusion.

Now we examine the effect of the laser wavelength on the echo shape. Figure 4 presents the experimental signals obtained for two probe wavelengths (555 and 580 nm) whose comparison clearly demonstrates a sign change in the acoustic contribution: the ringing oscillations and the first echo both have a reverse phase. One should note that the electronic contribution is not affected, which confirms that it is not due to a sign change of the whole signal. One should also note that the phase change of the ringing oscillations confirms that the observed effect is due to a change of the sign of the detection coefficient (a change in the probed thickness could also change the time of arrival of the echo and thus its phase but could not change the phase of the first picosecond oscillations). The same modification of the acoustic echo has been observed near 570 nm in different samples composed of similar layers. Here again one can find a wavelength for which there is no detection of the acoustic part of the signal (575 nm).

In these experiments both pump and probe wavelengths are modified due to the use of the OPO: probing at 555–580 nm requires a pump at 750–810 nm. One may argue that varying the pump wavelength may influence the echo shape. Nevertheless one has to remember (a more detailed discussion of this point has been given in Ref. 8) that it can only change the length over which the acoustic pulse is generated and in the present case this range is too small to see any effect. To validate this, we have also performed experiments using the same wavelength for pump and probe (750 and 810 nm). In these experiments no change was detected on the acoustic part of the signal. The observations of significant changes in the echo shape can thus be attributed to the varia-

tion of the probe wavelength near 570 nm. That conclusion is supported by numerical simulation, which requires a sign change in the piezo-optic couplings to well reproduce both echo shapes.

As in the aluminum case, the changes in the echo shape near the interband transition are strong. The role of the detection mechanism has been demonstrated experimentally and numerically. Here again the observed changes are consistent with previous results obtained in modulation spectroscopy.¹⁴ These results validate our explanation of the possible effect of laser wavelength on the shape of signals detected in picosecond ultrasonics detailed in Sec. II.

V. CONCLUSION

We have presented experimental results that confirm the connection that exists between picosecond ultrasonic experiments and electronic structure. Suggested by an observation in a tungsten film, this link was probed here in samples composed of aluminum and copper, both metals whose electronic structure is well known. In both cases we have observed strong changes of the detected acoustic echoes when the laser wavelength was tuned around an interband transition.

The role of an electronic transition in the observed effect is supported by modulation spectroscopy results.

The observed changes differ from the tungsten case in that they correspond to a sign change of echoes instead of a shape change. The origin of this difference may be related to the difference of structure of samples or to the electronic structure itself. We are carrying out numerical and experimental investigations to clarify this point. The present results have also permitted us to demonstrate that there exist wavelengths for which no acoustic information can be extracted from pump-probe experiments. This is particularly important in the aluminum case since it is usually used as a transducer in picosecond ultrasonics.

As the picosecond ultrasonic technique is contactless and nondestructive, it permits the exploration of very thin samples. The possibility of exploring electronic properties with such a method may thus be extended to very thin samples in which quantum size effects may appear.

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¹⁷This value of sound velocity was deduced from different experiments performed on the same silicon nitride. In particular, experiments with a blue probe have lead to an important oscillating part of the reflectivity due to an acousto-optic interaction in Si₃N₄ [see, for example, C. Thomsen, H. T. Grahn, J. Tauc, and H. J. Maris, *Opt. Commun.* **60**, 55 (1986)]. It lets us deduce the sound velocity from the period of the oscillations, the laser wavelength, and the real part of the index of refraction.

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