Tilted columnar metal film as transducer of transverse coherent acoustic phonons in Picosecond Acoustics

Revised Version

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Abstract

Picosecond acoustics has been widely used to study thin-film elasticity, hypersound attenuation and adhesion of thin-films to substrates. A major limitation of the technique is its restriction to longitudinal waves only. Although work has reported on the ultrafast generation and detection of transverse waves, a general method compatible with thin films deposited on silicon is still missing. In this work, we show that by depositing a tilted columnar metal film and using an optical detection sensitive to light polarization, it is possible to excite and detect optically both types of bulk acoustic waves in thin films. The protocol is first established on metallized glass substrates, then applied to a range of transparent films deposited on silicon (silica, AlN, AlScN, SiC). In each case, Brillouin oscillations are detected at two frequencies, one being the longitudinal mode, the other the transverse. The film thickness and two sound velocities are measured in each thin-film. Transverse coherent phonons as high as 116 GHz are observed in the SiC thin film.

Keywords: Picosecond Acoustics, Brillouin oscillation, Transverse acoustic waves, Glancing Angle Deposition,

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Picosecond Acoustics (PA) provides a way to make elastic measurements at several hundred of gigahertz, a frequency range the conventional physical acoustics cannot reach. PA is a purely optical, non-contact and non-destructive technique. First used to measure thin film thickness, it is particularly useful for complex stacks of various materials, since acoustic waves are compatible with all materials, whether opaque or transparent.[1][2][3] The technique also offers a unique way of measuring elastic properties of thin film materials which often differ from bulk materials[4][5]. Adhesion has also been investigated using PA.[6][7][8]

In practical terms, the technique combines a femtosecond laser and a pump–probe scheme to time-resolve the acoustic propagation at the nanoscale. In a metal film, an acoustic pulse results from light absorption at the free surface, where returning echoes from any buried interface, are also detected. From an accurate measurement of the time of-flight, one deduces either the film thickness or the sound velocity. In a transparent sample, a metal film is deposited on top of the sample to ensure acoustic emission. If that metal film is thin enough, light penetrates the sample, and interacts with the acoustic pulse during propagation, causing so-called Brillouin oscillations. In this case, propagation is time-resolved and the sound velocity is derived from the oscillation period, and the film thickness is then deduced from the time-of-flight. [9]

PA is essentially a 1D technique as the emission zone is much larger than it is deep (typ. 1000 times). The light pulse then excites only longitudinal phonons which are ideal for thickness control but a major limitation for elasticity measurement. The simplest case of an isotropic film already requires two moduli, Young modulus and Poisson ratio, which means two different sound velocities to measure. Over the years, various approaches have been proposed to overcome this limitation. (see Ref. [10] as a detailed review).

One method is to generate transverse waves by mode conversion at the interface between an isotropic film and an anisotropic substrate whose crystalline orientation is misaligned with respect to the directions of high symmetry. The longitudinal wave generated in the isotropic

material, at normal incidence to the interface, is partially converted into transverse waves by reflection at (or transmission through) the interface with an anisotropic substrate.[11][12][13] In other works, transverse acoustic (TA) waves have been generated by strongly focusing the emitting laser beam; here, the source is less directive and transverse waves can be diffracted and detected in the far acoustic field.[14][15][16] Another approach has been to use non-thermoelastic acoustic photoexcitation which facilitates the symmetry breaking needed for shear wave emission. Piezoelectric, ferroelectric and magnetic materials have been used to generate transverse acoustic pulses directly from ultrafast optical pulses. These studies are based on epitaxial films of specific materials (such as nitride semiconductor epitaxy, single-domain BiFeO3 epitaxy or iron epitaxy) which cannot be grown on just any substrate. [10][17] Deposition of a nanostructured metal film has also been proposed to excite and detect surface acoustic waves whose velocity can be used to deduce the transverse acoustic velocity. If such an approach can be applied to any thin-film, its main limitation is the sample preparation, based on an e-beam lithography process.[18][19]

These works illustrated possibilities for shear wave generation, but their requirements for specialized material fabrication reduce their versatility for use with a wide range of materials. In this letter we show that a simple deposition process combined to a modified pump-probe setup facilitates access to the mechanical properties of any thin transparent layer. We demonstrate the efficiency of our process on several samples made of a thin transparent layer deposited on a silicon substrate. After deposition of a tilted metal film, the propagation of both longitudinal and transverse waves is detected in the transparent film. Shear waves of frequency as high as 116 GHz are reported.

The experiments were conducted using a non-degenerate pump and probe configuration (Fig.1). The laser source is a Ti-sapphire oscillator which produces 120 fs optical pulses at a repetition rate of 80 MHz, tuned in this work at 816 nm. A first part of the laser output, designated in the

following as the pump, is focused onto the sample surface using a microscope objective with a spot diameter of ~ 1.7 µm at normal incidence to generate the acoustic pulse. The probe beam, which is issued from the same laser, is first frequency doubled with a β-barium borate crystal and then time-delayed with respect to the pump using a mechanical delay line. The probe is focused onto the same spot as the pump using a 50 mm lens. Following the pioneer work of Matsuda *et al.*, to detect TA wave, the probe angle of incidence is fixed to 45° and the change in reflected probe polarization is monitored.[20][21] To confirm the need for such a condition, some specific measurements were carried out using a conventional set-up, i.e. at normal incidence for both pump and probe, and measuring only the reflected probe intensity.

Two first samples are made of a metal film deposited on a glass substrate (microscope slide). One is covered with a 20 nm thick Al film. Since no specific deposition conditions are applied to Al deposition, this sample should not reveal a TA signal and is used as a reference sample. On the second glass substrate, a tilted columnar 400 nm thick W film is deposited by the Glancing Angle Deposition technique (GLAD).[22] The substrate holder is tilted at α=80° with respect to the substrate normal. The thickness and the columnar microstructure (column angle β = 39°) is confirmed by the observation of the cross-section using Scanning Electron Microscopy (SEM) (Fig. 1 inset).

Five others samples are made of a thin transparent layer deposited on a Si(100) wafer. We selected different materials for their different elastic properties: two SiO2 layers, one pure AlN film, one 15% Sc doped AlN film and an epitaxial 3C-SiC(100) layer. The 3C-SiC layer was deposited on Si(100) substrate by atmospheric pressure chemical vapor deposition (CVD) using a standard two-step process, including 10 min carbonization at 1165°C under 12 sccm propane followed by 60 min epitaxy at 1350°C under a C/Si ratio of 4 using 1.5 sccm silane and 2 sccm propane.[23]

A thin tilted metal film is deposited on top of each silicon sample, acting as a transducer. An important difference with the glass substrate is the non-transparency of the silicon substrate. The transducer must therefore be thin enough to allow the probe light to penetrate the sample and follow the acoustic propagation through Brillouin oscillation. The results presented in this work have been obtained with 20-25 nm thick metal film deposited by GLAD sputtering using two different metals and inclination angles: the incident angle α was 70° for Ti and Mo and 60° for W films. Table 1 summarizes the description of the samples and the tilted metal film deposited on each sample.

Figure 2(a) reproduces the transient reflectivity signal, obtained with a laser excitation and detection at the back surface of the two glass samples. In both cases, the pump laser excites an acoustic pulse in the metal film, then transmitted to the glass substrate where a Brillouin oscillation is expected to be detected as the probe light and acoustic pulse coexist in the transparent medium. In the case of the Al-coated glass substrate, a pure oscillating signal is detected at a frequency close to 40 GHz. It starts from t=0 and lasts as long as the time window. The Brillouin nature of such an oscillation is confirmed by its frequency, which is related to the sound velocity v and the probe wavelength λ through:

$$f = 2nv\cos\theta/\lambda, \quad (1)$$

where n is the Refractive Index (RI) at the probe wavelength and θ is the incidence angle of the probe beam. Assuming n=1.5, v=5970 m/s and an external incidence angle of 45°, one gets 38 GHz.

The signal obtained on the W/Glass sample appears to be less regular. Compared to the Al sample, the W film is much thicker and a significant elastic contrast exists between glass and metal. A part of the acoustic energy keeps confined in W and several strain pulses are transmitted to the transparent substrate after each round trip in the W film (154 ps for a 400 nm

thick W film). However, Fig.2(a) shows that the signal already presents a non-regular character between 0 ps to 150 ps, a range in which only one pulse is propagating through the glass.

The Fourier transform of the time-domain signals is given in Fig.2(b). For the Al sample, a single frequency at 39 GHz is observed as expected for a pure sinusoidal signal. On the contrary, in the W sample, two peaks are identified around 39 GHz and 25 GHz. The highest is exactly at the same frequency for both samples and corresponds to the longitudinal Brillouin frequency of the glass substrate. The second peak which falls well above the round-trip frequency (here close to 6 GHz) corresponds to the detection in the glass substrate of an acoustic wave that propagates at the transverse velocity. This is supported by extracting the sound velocity from the frequency using Eq.(1): 5870 m/s and 3740 m/s (using n@408nm=1.536 [24] and an angle of incidence of 45°). These values are close to the expected velocities of the longitudinal and shear sound velocity in glass (5970 m/s and 3750 m/s respectively) even if the glass substrate is not pure fused silica here. As two frequencies are detected simultaneously during propagation, their superposition generates a complex signal. In Fig. 2(a), the measured signal on W/Glass is very well reproduced with a simple superposition of the two frequencies. A final argument in favor of this interpretation, is the dependence we observed of the position of these frequency peaks on the laser wavelength. The change in wavelength is accompanied by a shift of both peaks according to Eq.(1).

The presence of a clear Brillouin frequency close to 25 GHz, demonstrates that transverse acoustic waves are propagating in the glass substrate. The comparison to the Al film case confirms that this is the use of a tilted film made with GLAD technique that allows the breaking of the shear symmetry in the direction normal to the sample and subsequently ensures the direct thermoelastic generation of shear in addition to longitudinal acoustic waves. Pézeril et al. reported simulations results of thermoelastic emission of shear waves by gold crystalline with particular orientations.[25] The same mechanism has previously been invoked to support the

observation of shear resonances in thin canted gold films.[26] According to this generation mechanism, the transverse waves emitted have a displacement governed by the orientation of the columns in the metal film. Consequently, the transverse mode with a perpendicular polarization should not be excited. Probing transverse waves in that specific direction would give nothing. This is indeed what we observed: as the polarization of the probe light is modified, the transverse signal disappears for a certain direction (see SI Fig. 1).

For glass samples, measurements were performed from the uncovered side to allow the probe to penetrate the sample. There was therefore no limit to the thickness of the metal film. In case of Si samples, the wafer is opaque and the metal layer must be thin enough to allow optical detection inside the sample while keeping its GLAD structure. Figure 3(a) compares the transient reflectivity measured on the same W/SiC/Si sample obtained with and without the detection sensitive to polarization change. Both traces are dominated by a long-lived oscillation and an oscillating echo detected around 300 ps.

The long-lived oscillation corresponds to the Brillouin oscillation detected in the SiC layer. The high speed of sound of SiC (typ. 11000 m/s) justifies the high frequency (150 GHz) we obtain. The structure detected around 300 ps is the photoelastic signal (a damped Brillouin oscillation) detected in Si as the pulse reaches the substrate: 300 ps is exactly the time-delay needed to travel across a 3.3 µm thick layer in which sound velocity is 11000 m/s.

The Si signal is lower than what we would have obtained with a 12 nm thick Al film on top. This is first due to the nature of the transducer. Tungsten is mechanically not well adapted to SiC, the related resonance of the transducer is visible in the first picoseconds. From the ringing period, we estimate the W film thickness close to 25 nm. The thickness of W limits the penetration of light into the sample. Despite this, the arrival of the acoustic pulse in Si is easy to detect.

When the setup detection is sensitive to change in probe polarization, the Fourier transform reveals a second contribution at a lower frequency (116 GHz). To confirm its origin, we compute the sound velocities from the two peaks detected here and using the RI of 3C-SiC at the probe wavelength (408 nm). According to Lothetidis and Petalas[27], n@408nm=2.79 and then using Eq.(1), we get 11187 m/s and 8760 m/s for longitudinal and transverse velocity, respectively. To compare with literature, those sound velocities are converted into elastic moduli using the mass density (3210 kg.m-3): C11=402 GPa and C44=246 GPa in very good agreement with Ref. [28] (C11=397 GPa, C44=259 GPa). Elasticity is different from one polytype to another in SiC. The comparison is easy here as the sample is an epilayer grown on a Si(100). From the arrival in Si (302 ps) we can also extract the SiC film thickness: 3325 nm which is in excellent agreement with the value of 3.3 µm deduced from IR reflectance measurement.

These results thus confirm that it is possible to make a metal film thin enough to permit photoelastic detection in the sample and the observation of both longitudinal and transverse waves. The same scheme has been applied to other Si samples using different metal films (Time-resolved signals and Fourier Transforms are visible in SI Fig.2). In each case, the combination of a GLAD transducer and an optical detection of polarization change allows us to detect two contributions in the Fourier transform. Table 1 presents the measured frequencies, the longitudinal and transverse sound velocity deduced from the RI, the delay at which the arrival in Si is detected and the film thickness deduced from the longitudinal velocity. In each case, an excellent agreement is found between nominal and measured thickness. On silica, the measurement was possible with either a Ti film or a Mo film on top. The comparison between AlN and AlScN is a particularly interesting case. Both materials are currently used as a piezoelectric layer in electronic device (radiofrequency filter). Doping with scandium increases the coupling factor of AlN but degrades the elastic properties.[29][30] As shown in Table 1,

our protocol allowed us to reach both longitudinal and transverse sound velocity in the two materials and make the difference between the doped and non-doped film.

Elastic moduli and film thickness are here deduced from the Brillouin frequencies and the arrival of the longitudinal pulse at the interface between the film and the substrate. An arrival of the transverse pulse may also be detected. Let's focus back on the SiC sample. Combining thickness and transverse sound velocity, the arrival in Si is expected around 380 ps. As shown in Fig.3(a), no echo is observed around that delay. Two reasons can explain this. First, it could be related to the photoelastic coefficient that governs the efficiency of the optical detection of the strain pulse. While the coupling is huge for longitudinal waves in the wavelength range close to 400 nm[31], it may be much lower for transverse. Second, an angle is needed to detect the transverse component (no transverse contribution at normal incidence in Fig. 3). But the Si RI around 400 nm is so strong (typ. 5.36@408nm) that the external incident angle is not large enough to provide a significant angle inside the substrate where the Brillouin signal is detected.

Other photoelastic contributions are also expected to be detected. One is the usual returning longitudinal echo detected in the metal film after a round trip in the layer. This is the acoustic echo usually used to deduced the film thickness in PA. Here, it is expected around 604 ps and it is clearly visible in Fig. 3(c) which presents the transient response at long delay. An interesting contribution, which does not exist in common PA, is the transverse echo expected when the transverse pulse returns to the metal film on top of the sample, here expected around 760 ps. In Fig.3(c) a structure is visible around 750 ps and may correspond to the transverse echo. A similar transverse echo has already been observed in previous work dedicated to observation of shear waves in PA.[10][11][14] This last result shows that the present work can be extended to non-transparent layers for which Brillouin oscillations do not exist. In that case,

longitudinal and transverse velocity can be obtained by measuring the time of flight of the corresponding echoes.

To conclude, we show that it is possible to prepare, using a standard deposition tool, a tilted thin metal film that can be used as a transducer exciting both longitudinal and transverse hypersonic waves when submitted to a femtosecond laser pulse. Combined with an optical detection sensitive to light polarization, the PA technique is extended to longitudinal and transverse domains. We have demonstrated this capability in a wide variety of transparent materials, SiO2, AlN, SiC, using different metals as transducers Ti, Mo and W. In each transparent thin-film, we detect a Brillouin oscillation for each component from which the longitudinal and transverse velocity is extracted. The observation of a returning transverse echo in the same transducer suggests that the protocol is applicable to opaque layers in which no Brillouin oscillation can be detected.

Supplementary Material

Two additional figures are presented in the Supplementary Material. SI Fig.1 shows the influence of the probe light polarization on the acoustic spectrum. SI Fig. 2 presents the time-resolved signals (and Fourier Transform) measured on samples #1, #2,#4 & #5.

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Institutional Review Board Statement

Not applicable.

Informed Consent Statement

Not applicable.

Data Availability Statement

The data presented in this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare no conflict of interest.

References

[1] C. Thomsen, J. Strait, Z. Vardeny, H. J. Maris, J. Tauc, J. J. Hauser, Phys Rev Lett. 53, 989–92 (1984).

[2] A. Devos, Ultrasonics 56, 90 (2015).

[3] O. B. Wright., J. Appl. Phys. 71, 1617 (1992).

[4] H. Ogi, M. Fujii, N. Nakamura, T. Yasui, and M. Hirao, Phys. Rev. Lett. 98, 195503 (2007).

[5] K. Tanigaki, H. Ogi, H. Sumiya, K. Kusakabe, N. Nakamura, M. Hirao, and H. Ledbetter, Nat. Commun., 4, 2343 (2013).

[6] J. Costa Dantas Faria, P. Garnier, and A. Devos, Appl. Phys. Lett. 111, 243105 (2017).

[7] A. Devos, and P. Emery, Surf. Coat. Tech. 352, 406 (2018).

[8] A. Vital-Juarez, L. Roffi, J.-M. Desmarres and A. Devos, Surf. Coat. Tech. 448, 128926 (2022).

[9] A. Devos, R. Côte, G. Caruyer, and A. Lefebre, Appl. Phys. Lett. 86, 211903 (2005).

[10] T. Pezeril, Opt. Laser Technol. 83, 177 (2016) and references therein.

[11] D.H. Hurley, O.B. Wright, O. Matsuda, V.E. Gusev, and O.V. Kolosov, Ultrasonics 38(1-8), 470–474 (2000).

[12] T. Bienville, and B. Perrin, Proceedings of the World Congress on Ultrasonics 2003, Paris, pp. 813–816. (2003). Hal-00016487

[13] Y. Wang, and M. Khafizov, J. Am. Ceram. Soc., 104, 2788 (2021).

[14] C. Rossignol, J. M. Rampnoux, M. Perton, B. Audoin, and S. Dilhaire, Phys. Rev. Lett. 94, 166106 (2005).

[15] Matsuda, O. B. Wright, D. H. Hurley, V. Gusev, and K. Shimizu, Phys. Rev. B 77, 224110 (2008).

[16] T. Dehoux, N.Chigarev, C. Rossignol, and B. Audoin, Phys. Rev. B, 77, 2143047 (2008).

[17] R. Gu, V. Juvé, C. Laulhé et al., Sci. Adv. 9, eadi1160 (2023).

[18] P. A. Mante, J. F. Robillard, and A. Devos, Appl. Phys. Lett. 93, 071909 (2008).

[19] O. Matsuda, K.Tsutsui, G. Vaudel et al., Phys. Rev. B 101, 224307 (2020).

[20] O. Matsuda, and O.B. Wright, Anal. Sci. 17 s216 (2001).

[21] O. Matsuda, O.B. Wright, D.H. Hurley et al., Phys. Rev. Lett. 93, 095501 (2004).

[22] A. Chargui, R. El Beainou, A. Mosset *et al.*, Nanomaterials 10, 1–18(2020).

[23] G. Ferro, T. Yeghoyan, F. Cauwet *et al.*, Mat. Science Forum, 1062, 23 (2022).

[24] M. Rubin, Sol. Energy Mater. Sol. Cells 12, 275-288 (1985).

[25] T. Péreril V. Gusev, D. Mounier, N. Chigarev and P Ruello, J. Phys. D 38, 1421 (2005).

[26] T. Pézeril, F. Leon, D. Chateigner et al., App. Phys. Lett. 92, 061908 (2008).
[27] S. Logothetidis and J. Petalas, J. Appl. Phys. 80, 1768 (1996).
[28] X.J. Liu, L.F. Li, and F. Lu, Optical properties and Mechanical properties of C, Si, Ge and 3C-SiC Materials Calculated from First Principles, Theory https://doi.org/10.48550/arXiv.1301.1745
[29] M. Akiyama, T. Kamohara, K. Kano, A. Teshigahara, Y. Takeuchi, and N. Kawahara, Adv. Mater. 21, 593–596 (2009).
[30] F. Tasnádi, B. Alling, C. Höglund, G. Wingqvist, J. Birch, L. Hultman, I.A. Abrikosov, Phys. Rev. Lett. 104, 137601 (2010).
[31] A. Devos, and R. Cote, Rev. B 70, 125208 (2004).

Figure 1

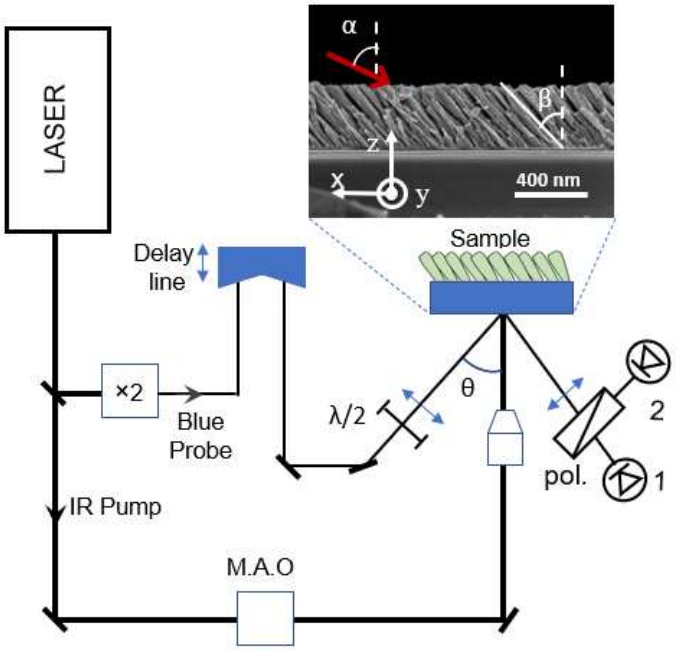

Figure 1– *Schematic view of the experimental transverse setup. The femtosecond laser output is split into two parts: 1) the pump that excites acoustic pulses in the sample, and 2) the probe whose polarization is monitored as a function of the time-delay between pump and probe. (λ/2, half-wave plate; pol., polarizer; M.A.O, acousto-optic modulator). Inset: SEM view of cross-section of 400 nm thick W thin film sputtered by GLAD with an incident angle α = 80°. The red arrow indicates the incoming flux of W atoms, which leads to an inclined columnar architecture (β = 39°).*

Figure 2

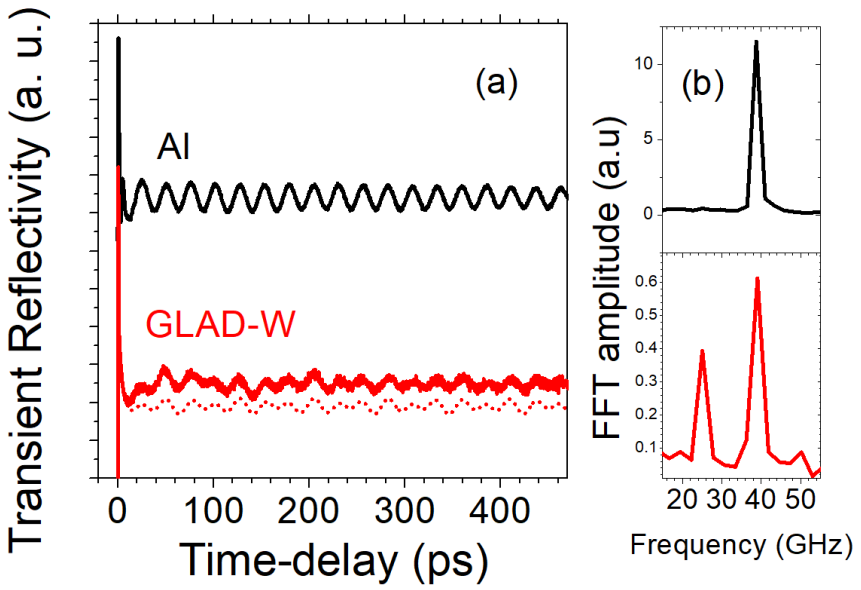

Figure 2: *(a) Transient relative reflectivity change in the glass samples covered with Al and W-GLAD metal transducers using the same setup. The dotted line is a superposition of two pure cosine functions. (b) Fourier transform of the transient reflectivity change.*

Figure 3

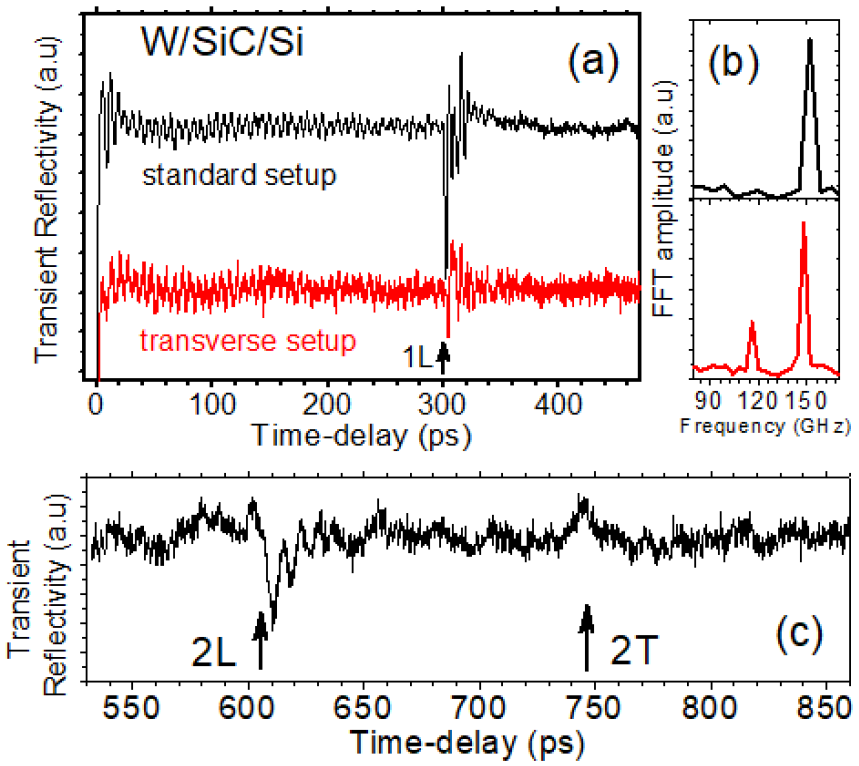

Figure 3 – *(a) Transient relative reflectivity change in W/SiC/Si sample measured with two distinct setups: standard means normal incidence with no sensitivity to polarization change; transverse setup means probe incidence at 45° with an optical detection sensitive to change in light polarization. (b) Fourier Transform of the transient reflectivity change. (c) Transient signal measured at longer time-delays: 2L (resp. 2T) is identified as the round-trip of the longitudinal (resp. transverse) pulse.*

Table 1 – *Description of the silicon samples, made of a transparent layer deposited on a Si substrate. A tilted columnar metal film is deposited by the GLAD technique on top of each. The quantities measured by PA are high and low Brillouin frequency, and the time-delay at which the acoustic pulse reaches the substrate. Longitudinal, and transverse velocity and film thickness are deduced from the measured data and the refractive index of the layer.*

Sample	Thin-film Material X (X/Si)	Nominal Thickness (nm)	Refractive index @408nm	Tilted Metal layer - α	High Freq. (GHz)	Low Freq. (GHz)	Longitudinal Sound Velocity (m/s)	Transverse Sound Velocity (m/s)	Longitudinal Arrival in Si (ps)	Thickness (nm)
1	SiO2	800	1.48	Mo-70°	38	24.5	5960	3840	138	822
2	SiO2	1000	1.48	Ti-70°	38	24.4	5960	3830	165	983
3	3C-SiC	3300	2.70	W-60°	148	116	11187	8768	302	3325
4	AlN	2000	2.15	W-60°	111	62	11150	6230	195	2174
5	AlScN 15%	1050	2.17	Ti-70°	104	59.5	10340	5910	103	1065