Imaging, non-destructive evaluation and characterization of complex materials and structures by laser ultrasound

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« *La dernière démarche de la raison est de reconnaître qu'il y a une infinité de choses qui la surpassent.* »

Blaise Pascal

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Foreword

This manuscript provides a global view of the two primary research axes that have occupied me at the Laboratoire d'Acoustique de l'Université du Mans (LAUM, UMR CNRS 6613) since I took up my position in September 2014.

Part I of this document encapsulates my scientific contributions, delineated into three concise chapters. Chapter 1 delves into my investigation of non-destructive testing and evaluation (NDT&E) by laser ultrasonics, highlighting resonances, especially the local resonances associated with zero-group velocity (ZGV) Lamb modes. Herein, I demonstrate how laser-induced ZGV resonances can quantitatively and locally monitor material fatigue and assess adhesive bonding. Chapter 2 unravels my focal research domain of picosecond laser ultrasonics, specifically the time-domain Brillouin scattering (TDBS) technique. I detail the evolution of TDBS, its applications in elastic property characterization, three-dimensional imaging, and real-time monitoring of transient processes. Alongside the exposition of our published findings, I outline our current and expected progress in this field. In chapter 3, I provide a succinct glimpse into additional research works in which I am actively engaged, yet have chosen not to explain extensively within this manuscript. Lastly, chapter 4 concludes with reflections and prospective directions within the domain of laser ultrasonics, notably accentuating future efforts I intend to pursue in picosecond laser ultrasonics.

Part II of this document encompasses my curriculum vitae, with a traditional CV, an overview of my academic and pedagogical duties, and an enumeration of publications.

Part ?? contains a selection of publications discussed in part I, serving as samples of my work. These publications are sequenced in chronological alignment with part I.

The work presented in part I of this manuscript was made possible thanks to strong collaborations. I am deeply grateful to my colleagues at LAUM and IMMM, as well as to national partners from LSPM, SAFRAN, ILM, GREMAN, Institut Langevin, and C2N. I also extend my thanks to international collaborators from Hokkaido University (Japan), Idaho National Laboratory (USA), and the Department of Physics and Astronomy at the University of Nottingham (UK). Unfortunately, modern research relies heavily on external funding, which has been crucial for advancing our work in laser ultrasonics. Details about the funding that supported these efforts are provided in my curriculum vitae presented

in part II.

As the reader will quickly notice, I have chosen to include citations in this manuscript only for published articles where I am a co-author. This decision aims to maintain conciseness, though readers should be aware that none of the presented works would have been possible without the foundational investigations on which they rely. These prior studies are, hopefully, well-documented in the references of the cited articles. I believe that each manuscript is part of a larger scientific effort, contributing to our understanding of new phenomena and inspiring the development of innovative methods. The interconnectedness of different minds, shared through conferences and peer-reviewed articles, is what makes research so rewarding and why I was inspired to pursue this journey in the first place.

PART I

Presentation of the main scientific results

Chapter

1 Laser ultrasonics for NDT&E:

Zero-group velocity Lamb modes and other plate resonances

Using laser pulses with durations ranging from sub-nanosecond to a few nanoseconds, focused on a sample surface with a fluence below the ablation threshold of the material, generates ultrasound in the low GHz or MHz frequency range, remotely and non-destructively. In metals and carbon-fiber-based composite materials, the main generation mechanism at play is thermoelasticity. The laser pulse is locally absorbed by the material, within its optical skin depth at the used laser wavelength, giving rise to a local temperature rise. This local heat source introduces a volume temperature distribution, giving rise to local thermoelastic stresses perturbating the sample mechanical equilibrium. This perturbation plays the role of a local source for broadband longitudinal and shear elastic waves that then propagate away from the laser-heated area. The laser pulse duration, the time of flight of the longitudinal elastic waves within the optical skin depth, or the heat diffusion length, depending on which one is the longest, dictates the frequency range of the generated ultrasound wavefield. The spatial distribution of the absorbed electromagnetic energy density dictates the spatial frequency, *i.e.*, the distribution of the elastic wavevector components. This spatial spectrum of the source modulates the directivity of the delta-localized optoacoustic source, which is determined by the boundary conditions. Using a second laser, that can be pulsed or continuous (our case), the remote detection of the mechanical disturbances of a sample surface associated with the propagating elastic waves can be performed using deflectometry, beam distorsion detection, or interferometry techniques. These laser-based, non-destructive, and remote generation and detection of ultrasound are the fundamental basis of a field of non-destructive testing and evaluation (NDT&E) techniques.

Overall, my contributions to this field of research are twofold: (i) to contribute in developing more local and quantitative characterization methods through experiments and their numerically-driven (semi-analytic and finite element modeling) analyses/processing, and (ii) to help tackling the challenging evaluation of fatigued/cracked materials and bonded structures, all in plate-like structures. To do so, I have focused on elastic guided waves known as Lamb waves, especially the specific Lamb modes with zero-group velocity points, which I have developed as a new research direction at LAUM.

1.1 Applications of laser-based zero-group velocity Lamb modes to evaluate plates

In this section, I present three applications of laser ultrasonics for NDT&E by zero-group velocity (ZGV) Lamb modes we have developed to detect flaws, characterize attenuation, or evaluate fatigue in plates. ZGV Lamb modes have been introduced as the particular solutions of the Rayleigh-Lamb dispersion relation in plate-like structure with free boundary conditions having a zero-group velocity and a finite lateral wavelength. Such a mode can hence be understood as a local resonance of the plate-like structure, in opposition to the more classic solution of the Rayleigh-Lamb dispersion relation having also a zero-group velocity but an infinite lateral wavelength, that could be referred to as plate resonances. Contrary to a plate resonance, a ZGV resonance is very sensitive to the local mechanical properties and local geometry of the plate-like structure due to the finiteness of its wavelength. This specific feature, associated with the very high quality factor of such a resonance, has triggered many applications (*e.g.*, thickness variation characterization, Poisson's ratio measurement, adhesive disbond imaging, anisotropy evaluation, etc.), following the pioneer work by C. Prada et al. in 2005, to which I have started to contribute during my postdoctoral work in the team of C. Prada in 2013-2014.

1.1.1 Impact flaw detection in composite materials

When I joined the LAUM in September 2014, I incorporated an on-going project named LUCITA (for "Laser Ultrasonics for Composite Inspection Totally Automated"), managed by the IRT Jules Verne (French Institute in Research and Technology in Advanced Manufacturing Technologies for Composite, Metallic and Hybrid Structures), in which LAUM and Airbus Groups Innovations were two of the partners. In the frame of this collaborative projects, we have explored the use of ZGV Lamb modes for flaw detection in composite materials. The main challenges to address when looking for laser generation and detection of ZGV Lamb modes in composite plates are that the composite plates usually have a low damage threshold, their surfaces are matt and diffusive for light, challenging the optical detection of ultrasound, and the resin-based composite are strongly viscoelastic, hence reducing the ZGV resonance's quality factors. To circumvent these drawbacks and still benefit from the ZGV resonance features, we have proposed a method enabling ZGV Lamb modes enhancement thanks to the beam shaping of the generation laser, making use of a mask with slits spaced by the ZGV wavelength. Depending on the spacing between the mask slits, it has been experimentally demonstrated that ZGV

Lamb modes interfere constructively when the resulting spacing between the opotacoustic sources distributed on the sample surface equals the ZGV wavelength [Fig. 1.1(a)] whereas they interfere destructively when the spacing is halved. This technique has been effectively applied on a composite sample, allowing to distinguish between healthy zones, where the expected ZGV frequency was indeed detected, and an impacted zone, where the frequency content was drastically changed [Fig. 1.1(b)], even when flaws were not visually detectable. This study hence suggested a practical way for flaw imaging in composite materials with a lateral resolution dictated by the ZGV wavelength. Pending the development of a realistic model that considers membrane resonance effects or local defect resonances to better utilize the detected frequency content near the ZGV frequency, it could evolve into a quantitative imaging technique, offering more insight into the flaw geometry. These results have been presented in Ref. [1].

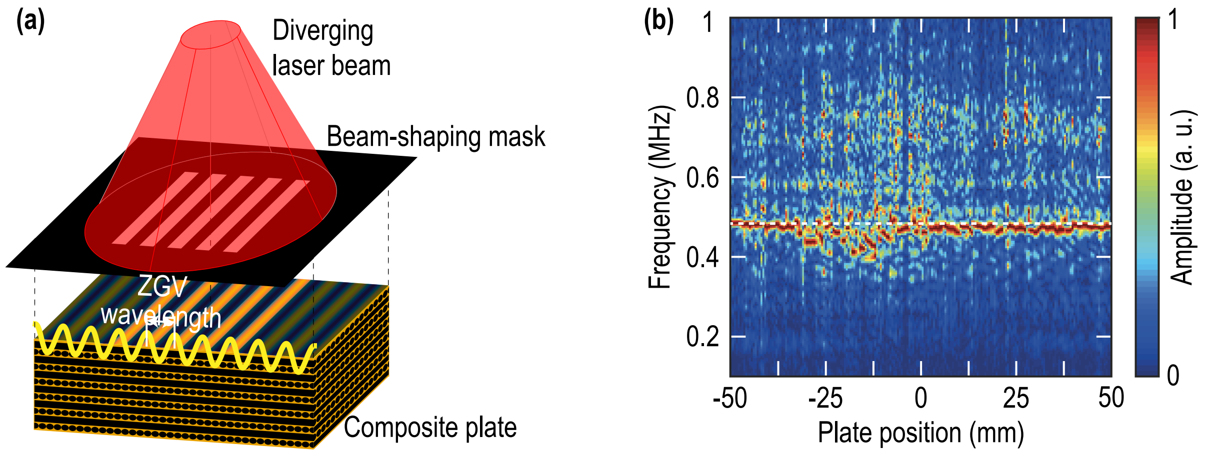

Figure 1.1: (a) Illustration of the ZGV Lamb mode generation using a beam-shaping mask. The generation laser beam is shaped with periodic slits whose spacing matches the ZGV wavelength. (b) Frequency spectra as a function of the plate position relative to the beam-shaping mask center for a one-dimensional scan of the sample away from the zone where the impact flaw was optically detectable. The deviation of the detected frequency content from the frequency peak associated with the expected ZGV resonance (white dashed horizontal line) demonstrates the ability of the proposed approach to detect the presence of, and potentially characterize, sub-surface flaws in composite materials.

1.1.2 Attenuation characterization in metallic materials

At the same time, benefiting from the rich environment at LAUM and a visit of Nicholas Boechler[1], a decision was made to apply a new processing method initially developed by Alan Geslain, Aroune Duclos and Jean-Philippe Groby at LAUM to different scenarios. The goal was to present this method while demonstrating its versatility [2]. Named

[1]At that time, Nicholas Boechler was at the Department of Mechanical Engineering, University of Washington. He is now Associate Professor in the Department of Mechanical and Aerospace Engineering at the University of California, San Diego.

SLaTCoW (Spatial Laplace Transform for Complex Wavenumber recovery), this method allows for the recovery of complex wavenumber via spatial Laplace transforms of spatiotemporal wave propagation measurements. One of the analyzed scenario involved the leakage of a ZGV Lamb mode generated and detected in a Duralumin plate using a linear array ultrasound transducer probe coupled to the plate by a thin layer of echographic gel. The SLaTCoW analysis revealed that the leakage introduced by the echographic gel layer leads to the separation, in the complex wavenumber plane, of the two interfering counter-propagating Lamb modes that compose the ZGV resonance. In the theoretical lossless case (no leakage, no material attenuation), these two modes precisely coincide at the frequency/wavenumber pair of the ZGV Lamb mode, known as the ZGV point. However, in the presence of any type of losses, this exact coincidence is lost, but the local ZGV resonance effect remains effective and detectable because of the near-to-zero group velocity of both modes in the vicinity of the ZGV point.

The SLaTCoW method, free of the usual restrictions/assumptions required by other methods to recover the complex wavenumber, was further used in order to extract the ultrasonic attenuation of a thin plate of polycrystalline aluminum. For this quantitative analysis, a ZGV mode in a free-standing plate was monitored by laser ultrasonics to limit mode attenuation due to leakage in the surrounding medium. Using the SLaTCoW method, the dispersion curves in the complex wavenumber plane were recovered and used to extract the ultrasonic attenuation (Fig. 1.2), which was successfully compared with two reported values measured by other means and also agreed with another estimate based on the decay time of the ZGV resonance. Further details on this application for characterizing attenuation can be found in Ref. [3].

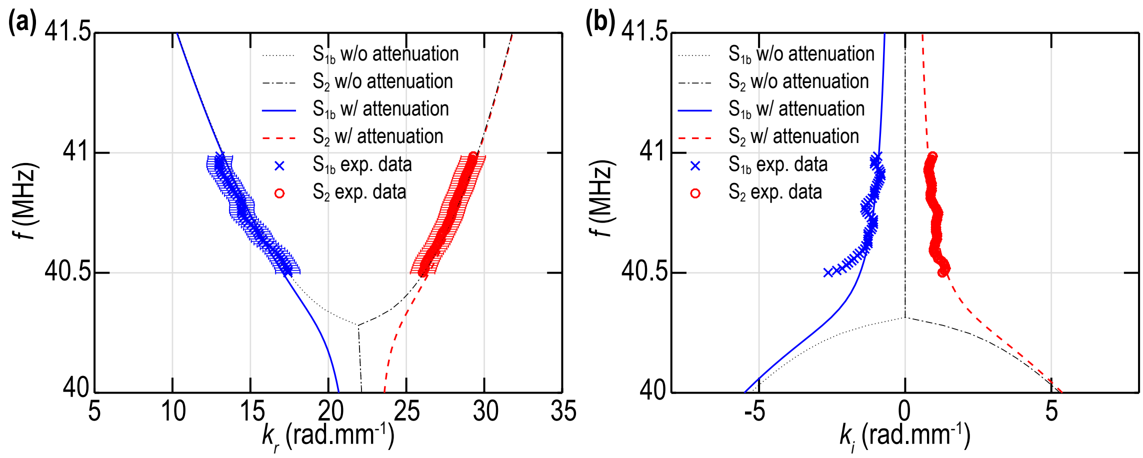

Figure 1.2: Dispersion curves in (a) the plane of the real part k_r of the wavenumber and the frequency f and (b) the plane of the imaginary part k_i of the wavenumber and f. The lines stand for the theoretical dispersion curves with (w/) or without (w/o) taking into account the material attenuation. The symbols stands for the experimental data. In (a), the error bars of the experimental measurements in the recovery of k_r is depicted by horizontal segments.

1.1.3 *In situ* monitoring of transient fatigue process in ductile materials

Another field of interest where ZGV resonance features can be highly beneficial is the monitoring of fatigue processes in ductile materials. In various industrial contexts, non-destructive assessment of cumulative fatigue damage in solid structures used in manufactured products is of paramount importance for ensuring reliability, quantifying health, and certifying security. This challenging achievement has been continuously recognized as of scientific importance over the past decade. Expecting an increasing signature of nonlinear phenomena due to material changes associated with cumulative fatigue, our primary goal was to combine my skills related to ZGV Lamb modes with those of the historical members of the optoacoustic research team at LAUM[2], who developed advanced laser ultrasonics techniques for NDT&E based on nonlinear phenomena.

While we did not succeed in demonstrating nonlinear phenomena with ZGV resonances in mechanically- or thermally-fatigued metallic plates without any ambiguity, we did make an experimental observation during the process that holds importance for material scientists interested in the study of material fatigue. For decades, several parameters such as damage index, Young's modulus, density, and cross-sectional area have demonstrated changes along the fatigue lifetime, and thus, they have been proposed as measurable physical parameters that provide information about the fatigue stage of a material undergoing fatigue testing. However, the variation of these parameters were all reported to be monotonous (either decreasing or increasing) with the material fatigue, which often necessitates an arbitrary definition of a quantitative threshold to denote a specific fatigue stage. In our experimental study, we demonstrated the capability of monitoring the frequency of a ZGV Lamb mode for (i) locating damage, (ii) predicting fatigue lifetime, and (iii) qualitatively and potentially quantitatively assessing cumulative fatigue damage levels during the fatigue process. This was achieved through three fatigue monitoring tests [Fig. 1.3(a)] conducted on three samples (75 µm-thick aluminum membranes), which exhibited identical relative variations of the ZGV frequency as a function of the normalized fatigue lifetime scale [Fig. 1.3(b)], despite experiencing failure at different amounts of loading cycles. This reproducible, nonmonotonous variation in the parameter, attributed to the existence of an extremum just before specimen fracture (approximately 80% of the fatigue lifetime), has led to the proposal of a quantitative empirical law characterizing a fatigue stage of a sample and might serve as an effective tool for predicting fatigue life. The details of these findings have been reported in Ref. [4].

To gain further insight into the experimental observations, we developed an empirically-inspired theoretical model based on cumulative damage theory, supplemented by finite element simulations that account for local geometrical changes in the plate during the

[2]Namely Nikolay Chigarev, Vitalyi E. Gusev, and Vincent Tournat

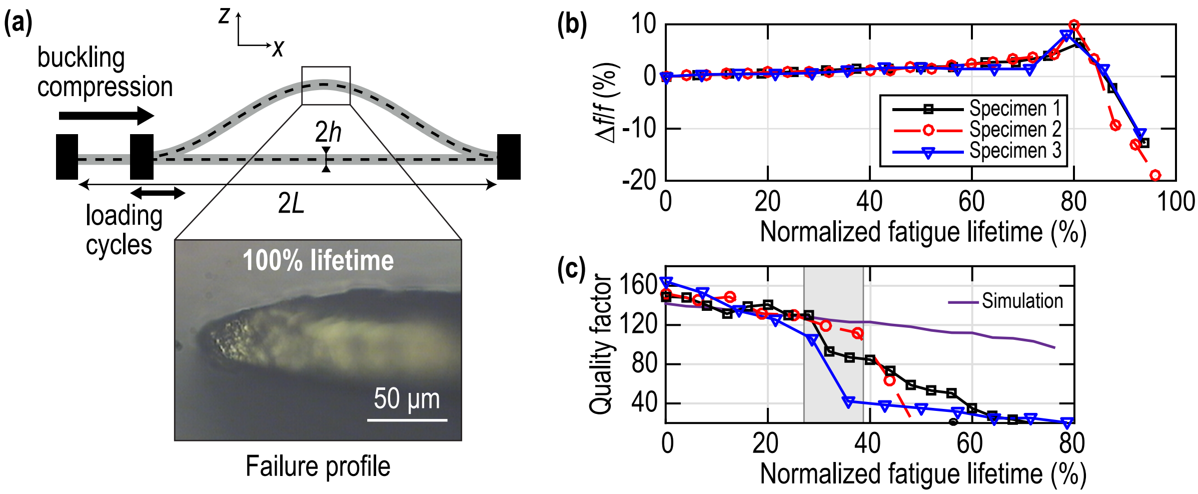

Figure 1.3: (a) Schematic presentation of the loaded and unloaded plate subjected to a buckling compression state at which loading cycles are realized. The inset picture shows the profile of one fatigued specimen after failure. Before laser ultrasonics measurements at different stages of the fatigue tests, the tested specimen was completely unloaded, returning to a plate with parallel free surfaces normal to the z-axis, allowing measurements to be conducted at several positions along the x-axis. (b) Comparison of the $\Delta f/f$ evolutions measured at the middle position (along x) of the plate in specimens 1, 2, and 3 (failure at approximately 16 000, 12 500, and 14 000 cycles, respectively) as a function of normalized fatigue lifetime. (c) Comparison of the evolutions of the quality factor measured in specimens 1, 2, and 3 with that obtained from the simulation, as a function of normalized fatigue lifetime.

fatigue process. This was done to compare with the experimental measurements. The study not only showed good agreement between the simulations and experiments concerning the ZGV resonance frequency, but it also helped elucidate the start time of the change in mechanical properties during the early fatigue stage, after which the changes cannot be ignored anymore. This observation was made possible through the analysis of the quality factor of the ZGV resonance, which demonstrated a disagreement between the simulations considering only geometrical changes and the experiments after about 30% of the fatigue lifetime [Fig. 1.3(c)]. The model-assisted analysis of the results from Ref. [4], where we primarily focused on the frequency of the ZGV resonance, underscored the importance of analyzing both the frequency and quality factor of ZGV resonances to achieve more quantitative monitoring of fatigue processes in ductile materials. Specifically, it revealed that the quality factor, despite varying monotonously, might serve as a better index for early detection of fatigue if continuous monitoring is performed throughout the structure life. Additional information on the implemented models and the conclusions drawn can be found in Ref. [5].

1.2 Evaluation of bonded structures: from adhesive-bonded parts to van der Waals heterostructures

Bonding between subparts in assembled structures is achieved through various means, depending on the scale or targeted applications for example. For aeronautical or automotive applications, adhesive bonding is increasingly favored over conventional methods like welding or riveting. It offers minimal added mass compared to rivets, eliminates the need for drilling and prevents stress concentration, which weakens the structures by producing better stress distribution between assembled structures. It is particularly efficient for composite material structures, aiming for a high strength-to-weight ratio. Additionally, adhesive bonding contributes to reducing environmental impact by mitigating fuel consumption through weight reduction. On the other end of the application spectrum are van der Waals heterostructures, which consist of stacked two-dimensional (2D) layers of different materials held together by van der Waals forces. These heterostructures show great promise in designing versatile electronic, optical, and elastic properties at the nanoscale, generating significant interest in research. However, in this case, not only the uniformity of the individual 2D layers but also the uniformity of their adhesion to the substrate and/or to another 2D layer is crucial, as it can influence the stability, reliability, and performance of heterostructure-based electronic devices that require uniform layers and interfaces.

Both types of bonded structures require non-destructive evaluation techniques after assembly. We have used laser ultrasonics for such evaluation in both cases, which I will briefly summarize in this section. However, due to substantial differences in the spatial scale of the tested structures (adhesive bonding and van der Waals heterostructures), different experimental techniques are employed. Adhesive bonding and van der Waals heterostructures are evaluated using elastic waves in the MHz and GHz ranges, respectively, generated with laser pulses of nanosecond or sub-picosecond durations. Resonances are the basis of two methods we have investigated for both cases, which is why they are discussed together in this section. The first two subsections summarize the results obtained for adhesive bonding evaluation in the frame of a collaboration with Safran Tech through a CIFRE[3] PhD thesis. The last subsection describes the evaluation of van der Waals heterostructures performed in collaboration with colleagues from the School of Physics and Astronomy at the University of Nottingham, in the frame of the scientific visit of Prof. Andrey V. Akimov to our team.

[3]CIFRE is a French acronym standing for an industrial agreement for training through research ("Convention Industrielle de Formation par la REcherche").

1.2.1 Evaluation of adhesive bonding by zero-group velocity Lamb modes

In our first paper on adhesive bonding evaluation [6], we investigated such evaluation by focusing on the attenuation (temporal decay) of ZGV resonances, contrary to previously reported results that primarily focused on the frequency of ZGV resonances. The study involved five trilayer assemblies composed of two asymmetric aluminum alloy plates bonded with a structural epoxy adhesive film. These assemblies included one nominal assembly, one with cohesive defects, and three with adhesive defects. Cohesive and adhesive defects were intentionally introduced to degrade the practical adhesion. To lower the cohesive strength of the adhesive, we reduced the curing time of the epoxy adhesive by half. For introducing adhesive defects, we disrupted the practical adhesion between one or both aluminum plates and the adhesive layer by applying a layer of release agent (Frekote® 44-NC™, Henkel, Germany) on the degreased aluminum alloy surface in contact with the adhesive layer before assembly and curing of the epoxy under nominal conditions.

Analyzing the frequency spectra [Fig. 1.4(a)] of the measured timel signal, we made two main observations. First, the sample in which both aluminum plates had been treated with the release agent behaved like a free-standing aluminum plate having the same thickness as the aluminum plate of the assembly on which the laser beams were focused to perform the experiments. In this configuration, the ZGV resonances displayed characteristics similar to those of a standalone aluminum plate. Second, all other four samples had similar but different ZGV frequencies, while the nominal bonded sample [black solid line labeled Nom. in Fig. 1.4(a)] presented a peak resembling two very close resonance frequencies, as expected from theoretical analysis. This latter observation precluded the use of a time-varying model signal associated with the normal displacement of a free surface due to a ZGV resonance, previously proposed by C. Prada et al., to fit the data. In addition to the product of an exponential decay, of time constant τ_1, representing viscoelastic losses and a $t^{-1/2}$ power-law decay due to the energy that is not trapped under the source, and thus can propagate at non-zero group velocity in the medium away from the inspected zone, we also needed to consider a beating phenomenon between two close resonances. To account for this, we introduced an extra cosine term, $\cos(\delta\omega t/2)$, dependent on the frequency difference between the two close ZGV resonances $\delta\omega$, in the model signal. By fitting the signals measured in the five trilayer assemblies [Fig. 1.4(b)], we were able to discriminate the different samples using the time constant τ_1 and $\delta\omega$, which quantified the differences in the temporal decay between each sample and paved the way for quantitative assessments of adhesive bonding. Finally, by performing a 2D scan of a trilayer assembly with areas containing adhesive defects and areas without introduced defects, we demonstrated the imaging capability of this contactless NDE method for adhesive bonding evaluation, which is made possible thanks to the local nature of ZGV resonances.

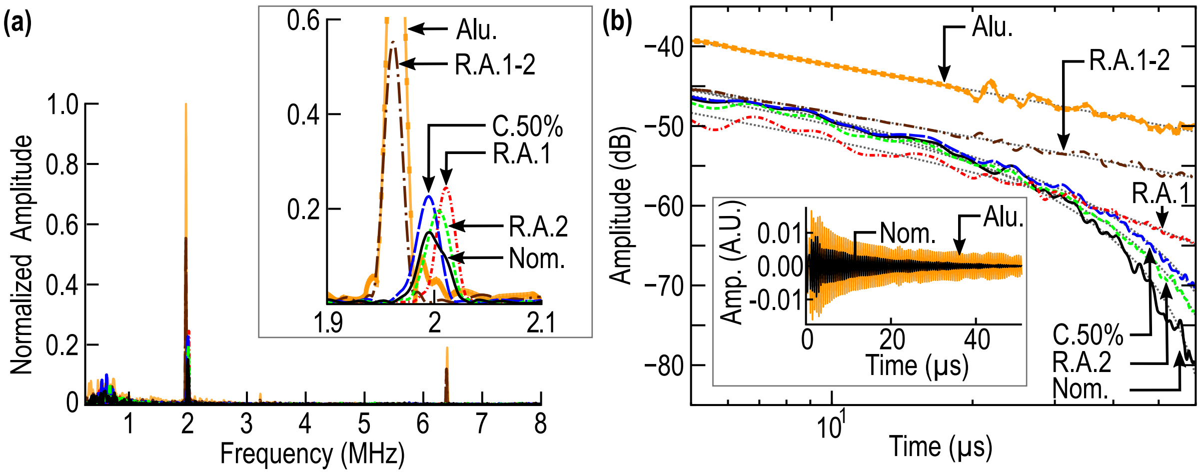

Figure 1.4: (a) Experimental frequency spectra in the range 0–8 MHz, with a closer view around 2 MHz (inset). (b) Experimental attenuation as a function of time, presented on a logarithmic scale. For each sample, the average attenuation for 11 different measurement points on the sample is plotted. The fitting lines are shown as dotted lines. The inset displays the temporal signals acquired on the aluminum alloy plate and the nominal bonded sample after filtering the data with a bandpass filter centered around the ZGV frequency at approximately 2 MHz.

1.2.2 Evaluation of adhesive bonding by plane wave synthesis

Besides ZGV resonances, we have also investigated another laser ultrasonics technique for evaluating adhesive bonding. This method, less local than the previous one, is based on the synthesis of elastic plane waves developed by Reverdy and Audoin for determining elastic constants. In this approach that we have reported in Ref. [7], elastic plane waves are numerically synthesized from experimentally detected propagating cylindrical waves, obtained using a laser beam shaped as a line. From the spatio-temporal data acquired by scanning the distance between the generation and the detection lasers, various angles of incidence with respect to the bonding interface are achieved by varying the delay in the delay-and-sum synthesis step. The synthesized plane waves are then used to solve an inverse problem to identify the normal and transverse interfacial stiffnesses (K_N, K_T) that model the mechanical coupling between the two bonded parts (Fig. 1.5). A semi-analytic 2D model, developed and detailed in a companion paper we published in the same journal issue [8], was used to create a database of simulated laser-generated ultrasounds required to solve the inverse problem. The developed method was first validated with semi-analytic simulated input data containing added Gaussian noise. Next, experimental signals acquired on an aluminum alloy plate and on two bonded assemblies (with and without adhesion defects) were used as input data. This method successfully distinguished the three specimens by finding the minima of cost functions based on the differences between the input data and the database composed of semi-analytic simulations for a large set of K_N and K_T. The differences between the identified values of interfacial stiffnesses provided quantitative values to characterize the adhesive bonding.

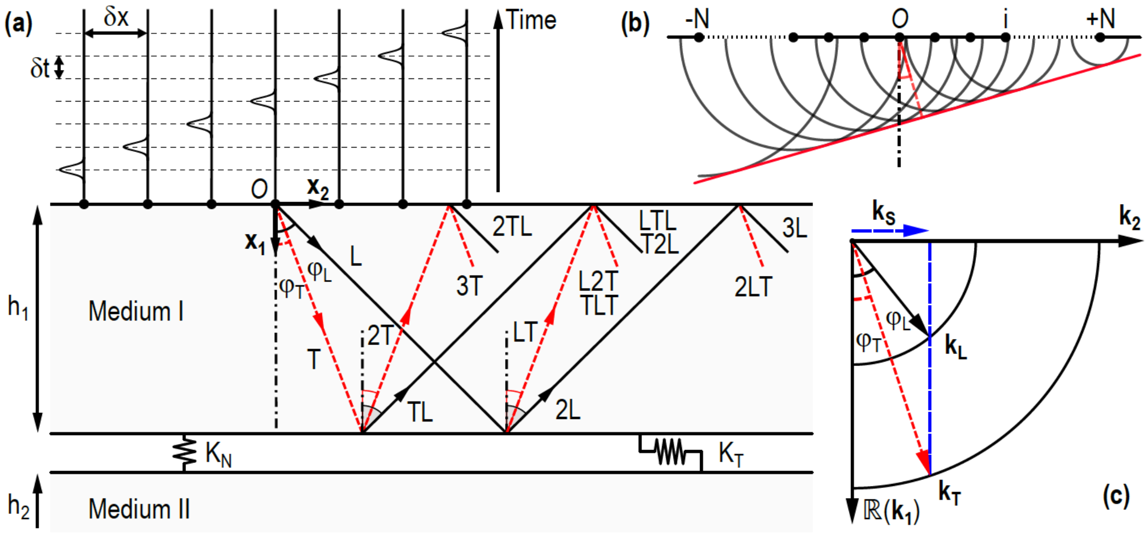

Figure 1.5: (a) Representation of the elastic plane waves that are synthesized in medium I. First, a laser line source of infinite length in the x_3 direction is moved over the sample surface in $2N + 1$ positions with a constant δx step. Second, a time delay δt is applied between the laser pulses in postprocessing to synthesized longitudinal (L) and transverse (T) plane waves. (b) The application of delays between the $2N + 1$ sources leads to constructive and destructive interferences between the divergent elastic waves, resulting in the generation of a plane wave. (c) Slowness diagram of the L and T plane waves that are synthesized by imposing the wave vector along x_2 of magnitude $k_s = \omega \delta t / \delta x$, with ω the angular frequency.

The main limitation we faced with this approach was that the mechanical properties of the bonded parts must be precisely known to obtain accurate simulations of the ultrasound propagation necessary for creating the database. Additionally, it was demonstrated that accounting for all physical phenomena involved in the ultrasound generation process was of utmost importance. In this specific case, disregarding the heat diffusion in the aluminum alloy precluded the inverse problem to be satisfactorily solved. Furthermore, this method provides average values of K_N and K_T for the scanned area (which was 32 mm long in our experiments), making it unsuitable for detecting local bonding defects. Last but not least, the top plate, where the laser generation and detection are performed, must be free of imperfections as assumed in the model. A fair question would be to discuss the interest of using the plane wave synthesis to compare the input data to the database instead of directly compare the simulated and measured spatio-temporal data. The reasons for this are twofold: the delay-and-sum process leads to reduce the noise level and, at the same time, it filters the contribution of surface acoustic waves (here essentially Rayleigh waves) which are of little interest as they propagate at the free surface of ultrasound generation without interacting with the bonding.

Note that the semi-analytic 2D model developed in the frame of this study exceeds the necessary requirements for such use. We have developed a multilayer semi-analytic model that successively solves electromagnetic, thermal, and elastodynamic problems. It accounts for optical penetration of the laser line source, as well as thermal conduction and convection. The model also considers optical, thermal, and mechanical coupling conditions between the upper and lower media of the multilayer. All layers are assumed electromagnetically linear, isotropic, and homogeneous, while they are thermally and elastically linear, homogeneous, temperature independent, and orthotropic. The model can also account for viscoelasticity. However, it is assumed that the direction normal to the layer surfaces [x_1-axis in Fig. 1.5(a)] and the direction defining the lateral infinite extension of the layers [x_2-axis in Fig. 1.5(a)] must be aligned with the principal axes of the orthotropic material symmetry. Despite these limitations, we believe that this versatile model is of interest to researchers working with multilayer structures in laser ultrasonics and hence have shared it on GitHub (`https://doi.org/10.5281/zenodo.4301720`).

1.2.3 Interface coupling in van der Waals heterostructures

As a last example of bonding evaluation, although taking place at a far smaller scale, I present the results we obtained in collaboration with colleagues from the University of Nottingham by picosecond laser ultrasonics and reported in Ref. [9]. Technical details explaining how elastic waves in the GHz range are able to be generated and detected with lasers are left for the next chapters. I want to emphasize here only the possibility offered by the use of lasers to generate and detect elastic waves/mechanical vibrations at the nanoscale, providing a great opportunity to evaluate bonding at that scale, such as between a 2D material layer and a substrate or between different stacked layers of 2D materials, held together by van der Waals forces. The techniques and forces involved in this case are quite different from the cases discussed in Secs. 1.2.1 and 1.2.2. Yet, these structures can be seen as multilayer structures with elastic coupling between the layers or with the substrate that can be described by interfacial stiffnesses. Due to the very large ratio of the lateral dimension of the generation and detection laser foci (hundreds of nanometers at best when using visible light) to the thickness of the multilayer structure (one to few atomic layer for each layer composing the structure), it is hardly possible to generate elastic waves with a wavevector having non-zero component along the layer surfaces. Therefore, this is usually treated as a one-dimensional (1D) problem in which displacement normal to the layer surfaces is the only non-zero component of the displacement field. In that case, the only resonances that could be generated, detected and used for bonding evaluation are plate resonances with an infinite lateral wavelength matching the 1D problem requirement. ZGV resonances are, of course, excluded since they are defined as local resonances of a plate-like structure with a finite lateral wavelength.

While examining the signals obtained on an InSe/InSe homostructure on a sapphire substrate [Fig. 1.6(a)] and that on an hBN/InSe/graphene heterostructure on a SiO_2/Si substrate [Fig. 1.6(b)-(d)], we have been able to explore different elastic coupling conditions. In the first case, the acoustic impedance of InSe and that of sapphire are significantly different, the former being four times smaller than the latter. Because of this, if the elastic coupling (the bonding) between InSe and sapphire is very good (perfect interface), the homostructure in which elastic waves are generated is expected to behave as a quarter wavelength resonator with fixed-free boundary conditions. On the opposite, if the elastic coupling is very bad (broken interface), the homostructure is expected to behave as a half wavelength resonator with free-free boundary conditions. Those two behaviors have been observed, as well as a mixed behavior revealing a position where the laser foci

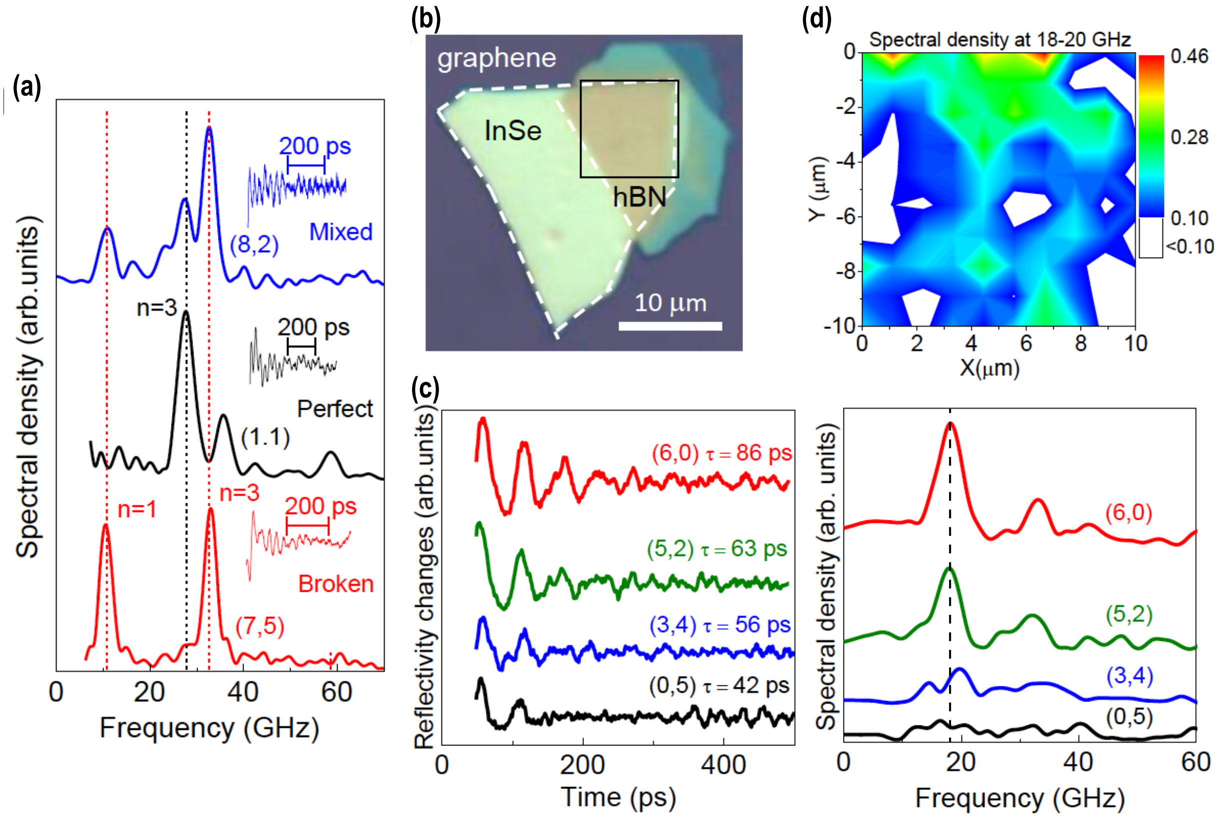

Figure 1.6: (a) Temporal evolutions (insets) and their frequency spectra measured at different positions on an InSe/InSe homostructure on a sapphire substrate. The lowest red spectrum corresponds to a broken interface; the middle black one to a perfect interface; the top blue one to a position where both broken and perfect interfaces coexist. Black and red dotted lines show the calculated frequencies of the resonances for perfect and broken interfaces, respectively. (b) Optical image of an hBN/InSe/graphene heterostructure on a SiO_2/Si substrate. The black square indicates the area of the heterostructure imaged by picosecond laser ultrasonics in parts (c) and (d). (c) Temporal traces of reflectivity (left) and related frequency spectra (right) at different positions on the heterostructure. The dashed vertical line indicates the main peak centered at 18 GHz. (d) Spectral density images at frequencies of 18–20 GHz. The white areas correspond to the spectral density values below the noise level. In (a) and (c), position coordinates inside brackets are in micrometers.

are encompassing an area where both perfect and broken interfaces laterally coexist [see frequency spectra depicting resonances in Fig. 1.6(a)]. In the case of the heterostructure on the SiO_2/Si substrate, the difference in acoustic impedances is small. In that case, the resonance frequency does not change much with the elastic coupling, whereas the lifetime of the resonance (τ) strongly depends on this coupling. The degradation of the elastic coupling from a perfect to a broken interface has been, therefore, seen in this case by the increase of the resonance lifetime [see Fig. 1.6(c)]. With that study, we demonstrated the possibility of mapping [Fig. 1.6(d) and further examples in Ref. [9]], by picosecond laser ultrasonics, the elastic coupling between multilayer nanostacks and different substrates. Additionally, the bonding between the van der Waals layers was shown to be as perfect for frequencies up to several tens of GHz, as the effect of such bonding defects had been ruled out. This work introduced a novel technique enabling a nondestructive assessment of van der Waals layers and heterostructures, with a demonstrated higher sensitivity to broken interfaces than AFM imaging, which we believe could be important for identifying and sampling areas for specific studies and applications in the field of van der Waals layers and interfaces.

1.3 Concluding remarks

In this chapter, I have presented the research activities conducted at LAUM on laser ultrasonics for NDT&E, with a particular emphasize on resonances, especially the local resonances associated with zero-group velocity Lamb modes. From my point of view, our main achievements in this field are the proofs of concept that laser-based ZGV resonances can be used to locally and quantitatively monitor the fatigue of materials and evaluate adhesive bonding. As discussed in chapter 4, we have recently initiated, in collaboration with Sylvain Mézil and Claire Prada (both CNRS researchers at the Langevin Institute, Paris), the investigation of another type of guided elastic waves known as edge waves, which also exhibit zero-group velocity points and resonances. These edge waves are elastic waves guided at the free edge of a laterally finite plate structure, and our aim is to better understand resonance and guiding phenomena that might occur at a crack in a plate. These research activities, now encompassing both ZGV Lamb and edge waves, were my primary focus for my first five/six years at LAUM and continue. However, today, the center of mass of my research activities has shifted to picosecond laser ultrasonics. Far from being left behind, my expertise in elastic guided waves and resonances remains central and contributes to the overall direction of my research plan, as briefly discussed in Chapter 4.

CHAPTER

2

Time-domain Brillouin scattering:

A tool for elastic property characterization, three-dimensional imaging, and real-time monitoring of transient processes

In the last section of the previous chapter, picosecond laser ultrasonics, the branch of laser ultrasonics dealing with the generation and detection of GHz elastic waves by lasers, has been used to measure GHz resonances of multilayer nanostacks in order to evaluate their bonding to the substrate, giving a fair introduction to the scales involved in this method. The lateral extend of laser foci controls the lateral resolution of the method, which is important for property mapping/imaging purposes. The elastodynamic transient effects last about hundreds of picoseconds to several nanoseconds, with features of duration in the picosecond range. When dealing with nanometer-thin layers, GHz plate resonances can be used for their non-destructive evaluation. For thicker layers, as soon as GHz acoustic pulses can be launched in the layer, typical pulse-echo measurements (time of flight, amplitude of echoes, etc.) can be used for the evaluation. This is already a well established technology for control applications in electronic industry (see for example the Echo™ system proposed by Onto Technology®). When layers are transparent to the wavelength of the laser used for detecting elastic waves, it opens up the possibility of detecting picosecond acoustic pulses not only when they reach and reflect at the surface between the layer and a transparent material (such as air for free surfaces) through which the lasers are focused, but also for monitoring the acoustic pulses along their propagation path via the photoelastic effect. When acoustic pulses of picosecond duration are coherently generated by very short laser pulses and monitored in time, as they propagate, by other very short laser pulses, the technique is named *time-domain Brillouin scattering*[1] (TDBS).

The development of the TDBS technique and its applications to elastic property characterization, three-dimensional imaging, and real-time monitoring of transient processes is at the core of this chapter and is today my main research topic, which I started when I joined the LAUM and started collaborating with my colleagues from LAUM, especially

[1]In the literature, it is also called picosecond acoustic interferometry or time-resolved Brillouin scattering.

Vitalyi E. Gusev and Nikolay Chigarev. I will now summarize our main achievements on that matter and, by doing so, will hopefully share the enthusiasm of mine for pushing further the technical and theoretical developments needed for this technique to become a widely used technique in many fields of sciences, from material sciences to biology, where it is definitely able to provide complementary insights on material elastic, optical, acousto-elastic properties or structures/textures compare to more classic and well established techniques of characterization, such as x-ray-based techniques, electron microscopy, scanning probe microscopy, or else Brillouin light scattering spectroscopy, to name but a few. After a rather technical introduction, I will span the recent developments and applications using TDBS achieved at LAUM over the past 10 years, in an attempt to sketch the roadmap guiding both our short-term and long-term perspectives discussed in the following chapters.

2.1 Technical introduction to pump-probe experiment and time-domain Brillouin scattering

The purpose of this section is not to provide an exhaustive introduction to the theoretical and experimental aspects of TDBS, which would require much more than few pages, but rather to offer an overview of key concepts which understanding is crucial for comprehending the developments described in the subsequent sections and the factors that may contribute to the wider adoption of TDBS as a universally-used characterization and imaging technique.

2.1.1 Pump-probe experiment

In order to generate and detect elastodynamic transient of several nanoseconds with picosecond time resolution to resolve ultrafast features, picosecond laser ultrasonics experimental setup are based on *pump-probe experiments*. The principle is as follows. An ultrashort (typically sub-picosecond long) laser pulse, called the *pump*, is used to launch a picosecond coherent acoustic pulse (CAP) in the sample volume through a generation mechanism that depends on the material. Another, time-delayed, ultrashort laser pulse, called the *probe*, is used to detect this CAP. Although different mechanisms or optical setups can be involved or used for CAP detection, we will primarily focus our attention on the photoelastic effect. In this case, the CAP strains the material and acts as a local disturbance of the complex dielectric tensor of the material. The probe pulse is sensitive to this local disturbance as a local perturbation in the material reflectance, which can be observed by monitoring the change in probe intensity after its reflection from the sample[2].

[2]Note that the measurement is also possible with the transmission of the probe if the latter can be monitored.

Indeed, the total reflected probe electric field from the material containing the perturbation can be seen as the sum of an electric field reflected from the unperturbed material and an electric field transmitted inside the material backscattered by the acoustic strain perturbation and transmitted back to air towards the photodetector where both electric field parts interfere. Note that the backscattered light, from now on denonimated simply scattered light, is intrinsically very weak and can be only detected via optical heterodyning detection, here realized through the superposition of the reflected and scattered probe light field at the photodetector[3]. By precisely controlling the time delay between the arrival of the pump and probe pulses at the sample, and assuming the transient response of the sample to the pump pulse action is repeatable, the collected probe intensity for each time delay will contain transient changes associated with ultrafast elastodynamic transients. Note that the received signal will also include other effects of the pump pulse action on the material, such as electronic and thermal effects. The former typically depicts much faster dynamics, while the latter exhibits much slower dynamics than those associated with the CAP. This distinction usually allows to filter out these effects and focus on the ones related to acoustics.

2.1.2 Control of the pump-probe time delay: mechanical delay line vs. asynchronous optical sampling (ASOPS)

Precise control of the pump-probe time delay can be achieved through various means, two of which are commonly used today. One approach involves using a single ultrafast mode-locked laser cavity and splitting its output laser beam, which consists of a periodic train of ultrashort laser pulses, into two parts for the pump and probe. One of these paths includes a controllable micrometric-precision mechanical delay line that can adjust its length and, thus, the arrival time of the laser pulses passing through it compared to those passing through the other path. The second method employs two ultrafast mode-locked laser cavities in a leader-follower configuration. The repetition rates of both cavities are stably offset using a phase-locked loop, and real-time control of a stepper motor and piezo control in the follower laser cavity precisely drive the position of one cavity mirror. State-of-the-art synchronization electronics provide a jitter time shorter than, or comparable with, a single laser pulse duration, enabling time-resolved experiments.

The solution with two synchronized laser cavity is known as ASOPS, standing for asynchronous optical sampling, and it allows high-speed scanning of the pump-probe time delay without the need for a mechanical delay line. The scanning speed is controlled by the repetition rate difference, which also determines the time resolution. For instance, with a leader cavity repetition rate of 42 MHz and a repetition rate difference of 500 Hz, the

[3]This is sometimes shortened by saying that the scattered light is heterodyned by the reflected light and the reflected light is hence called the heterodyning light.

achievable transient lasting time is 23.8 ns, and this time can be scanned over in 2 ms. In comparison, if one were to scan 2 ns of the transient response with a mechanical delay line and assume the beam path is four times the distance between the beam entrance on the delay line and the retroreflector position (by reflecting back the beam to the retroreflector once), the mechanical delay line would need to be moved at a speed of 75 m/s to achieve the same scanning speed of the pump-probe time delay, while for such "long" pump-probe time delay scanning range it is usually moved at a few cm/s. Fast mechanical delay lines based on shakers can be used for scanning "short" pump-probe time delay range faster.

However, it is important to keep in mind that both solutions have their advantages and disadvantages. For instance, the use of a mechanical delay line provides the opportunity to modulate the pump laser intensity in the radio frequency range and feed the output of the photodector to a lock-in amplifier synchronized at the pump modulation frequency. This allows for a drastic improvement in the signal-to-noise ratio, which will then be limited by shot noise. As a result, good signal-to-noise ratio is usually achieved after hundreds of averages. On the contrary, the acquisition time needed to obtain a single time trace with ASOPS is incredibly fast, allowing for direct averaging of signals to improve the signal-to-noise ratio. In this case, other sources of noise (technical, environmental) might become the limiting factors. Yet, from our experience, only thousands to a few tens of thousands of averages are commonly needed, which corresponds to a minute of acquisition or less. Going back to the previous example, with the delay line speed set at 3 cm/s and assuming that data are acquired during both scan directions (negligible or compensated backlash) for a total of 100 averages (50 round trips), the acquisition time with the setup using a mechanical delay line would be 500 s. In comparison, this would correspond to acquiring 250,000 averages with the ASOPS-based setup, ten to fifty times more than what is usually needed.

The advantages of ASOPS-based setup are clear: faster pump-probe delay time scanning speed, no pointing instability or defocusing due to an optical path change, reduction of mechanical noise, and recording long lasting transient without increasing the acquisition time. It makes this type of experimental setup the perfect one to perform imaging experiments. However, the time resolution, Δt, in ASOPS is determined by the repetition rate difference, Δf, and the leader cavity repetition rate, f_r: $\Delta t \approx \Delta f / f_r^2$. This expression sets boundaries on the achievable range of Δt once the laser system and synchronization electronics are chosen. In the example case, with $f_r = 42$ MHz and $\Delta f = 500$ Hz, $\Delta t \approx 0.28$ ps. Considering transient signal events to be well resolved with five times better time discretization than that suggested by Shanon criterion gives an upper limit to the frequency bandwidth of the system at about 360 GHz. In comparison, for a setup based on a mechanical delay line, the delay line scanning speed, v_{line}, and the lock-in amplifier integration time, $\tau_{\text{lock-in}}$, set the time resolution by $\Delta t = v_{\text{line}} \tau_{\text{lock}} / c_0$, with c_0 being the speed of light in air. For $v_{\text{line}} = 3$ cm/s and $\tau_{\text{lock}} = 3$ ms, it gives $\Delta t \approx 0.3$ ps. The great advantage of the setup with a delay line, therefore, comes from the fact that the balance between acquisition time, time resolution, and signal-to-noise ratio of a single time trace can be adjusted at will; in other word, it is more adaptable to any sample *a priori* than an ASOPS-based setup. Yet, it is not suited for laterally well-resolved imaging purposes in a limited/reasonable acquisition time.

Last but not least, the photodetector is a crucial element of the data acquisition and must be chosen wisely to avoid deteriorating the expected performance, having in mind that sensitivity is inversely correlated to bandwidth. For instance, because a larger bandwidth is needed to access the same high-frequency transient content when using an ASOPS-based setup compared to one based on a mechanical delay line, the sensitivity of the photodetector is usually lower. Clearly, the key word here is *trade-off*, as is the case for all experimental designs, but further discussion on this topic goes beyond the scope of the points I intended to address in this section.

2.1.3 Time-domain Brillouin scattering: principle and challenges

Up to this point, the technical considerations discussed have been general to the field of picosecond laser ultrasonics. Now, we will focus on specificities that arise when dealing with a material transparent to the probe laser wavelength.

In this scenario, the probe intensity at the photodetector includes additional components related to the CAPs, which scatter the probe light through the photoelastic effect all along its propagation path. This implies that the interaction between the probe light and the acoustic fields, driven by photoelasticity, is not limited to a volume close to the surface where the pump and probe laser beams are focused. Instead, it extends as far from that surface of CAP generation as the scattering of light by sound and its detection are effective. Assuming a collinear propagation of the probe light and sound in a direction normal to the surface, this volume is constrained along the propagation direction of light and sound by the shortest distance among the following ones: the probe laser pulse coherence length, the diffraction lengths of the probe light and acoustic beams (tightly controlled by the laser foci), and the characteristic lengths of probe light and acoustic wave absorption/scattering.

When considering a single longitudinal δ-localized plane CAP propagating in an optically isotropic semi-infinite material along the same direction as a plane probe light pulse, the additional component to the probe intensity time variation takes the form of a decaying sine modulation. This decay occurs due to the loss of scattering efficiency caused by at least one of the factors mentioned earlier. This oscillating component results from the interference of the probe light reflected at the unperturbed/steady sample surface and the weak probe light scattered inside the sample by the acoustic strain pulse through the photoelastic effect, as previously explained. As the pump-probe time delay changes, the position of the CAP also changes as it propagates, leading to a varying relative phase at the photodetector between the scattered and reflected light fields. The path of the scattered probe light is longer than that of the reflected probe light by twice the distance between the sample surface and the CAP. Consequently, both light fields presents the same phase difference as soon as the CAP has propagated half a wavelength of the probe light in the material. The time for the CAP to propagate such a distance, *i.e.*, the period of the oscillating component, is given by the product

$$\frac{\lambda_{\text{probe}}}{2n} \times \frac{1}{v_{\text{ac}}}\,,$$

where n is the material refractive index at the vacuum probe wavelength λ_{probe}, and v_{ac} is the acoustic velocity. The frequency f_B of this oscillating component eventually reads

$$f_B = \frac{2nv_{\text{ac}}}{\lambda_{\text{probe}}}\,. \tag{2.1}$$

As a matter of fact, this frequency exactly matches the Brillouin shift in frequency that a photon may either loose (Stokes process) or gain (anti-Stokes process) during its inelastic interaction with an acoustic phonon, a phenomenon known as Brillouin scattering. This particle view complements the classical view of the same process described in the previous paragraph and explains why the technique is referred to as time-domain Brillouin scattering. Another way to understand this process is to consider the propagating CAP as a moving mirror on which the probe light reflects. This is akin to the principle of a Doppler radar. Because the Doppler shift affects both the probe light incident on the moving mirror and the probe light reflected back from it, the frequency change of the probe light scattered by the CAP, propagating at a velocity v_{ac}, is twice that which would have occured if the CAP had directly emitted the light. Consequently, there is a Doppler shift in frequency of the scattered light equal to $2v_{\text{ac}}f_i/c$, where f_i is the frequency of the incident probe light and c is the speed of light in the medium, resulting in the frequency f_B. The interference between the acoustically frequency-shifted scattered light and the reflected light at the surface, when the total light field intensity is monitored at the photodetector, will give rise to an oscillating component at the frequency f_B.

The TDBS-twin spectroscopy technique, known as Brillouin light scattering and here referred to as frequency-domain Brillouin scattering (FDBS) for clarity, relies on the same light-sound interaction process but monitors the frequency shift of the scattered light using a spectrometer. In FDBS, the Brillouin scattering occurs not with coherent acoustic phonons excited by a pump laser, but with thermal incoherent phonons naturally present in the material due to thermal agitation. While conducting FDBS experiment with an additional pump laser to coherently excite acoustic phonons is a possibility, the drawback for imaging or characterization purposes when working in the frequency domain with spectrometry is the loss of time resolution. This constitutes the main difference between the two techniques and has important implications in terms of achievable spatial resolution.

While one can argue that FDBS can achieve similar resolution as TDBS, it is not entirely true. In fact, both techniques have lateral resolution controlled by the probe beam focus but different origins for their axial resolution, which refers to the resolution along the direction of light propagation. In FDBS, the axial resolution is determined by the optical length over which the signal is gathered. This length is controlled by the optical technique used to focus the light and is smallest when confocal microscopy is used. On the other hand, in TDBS, the axial resolution is solely determined by the CAP length, which typically ranges from a few tens to hundreds of nanometers, *i.e.*, smaller than the axial resolution limit of optical methods (sub-optical resolution). This highlights the main advantage of TDBS over FDBS, as well as its main challenge. Achieving the expected axial resolution with TDBS is indeed challenging due to the need to process the oscillating component of the time signal with both good time and frequency resolution, particularly in cases of complex geometries, as will be seen later.

In terms of acquisition time and frequency resolution, however, FDBS currently outperforms TDBS. The superior frequency resolution of FDBS is a direct result of its spectrometric nature. Two main technologies are nowadays widely used in FDBS. The first involves a high-performance multipass scanning tandem Fabry–Pérot interferometer, providing high contrast at the cost of long acquisition times. The second technology, known as the virtually imaged phased array (VIPA), offers a much faster acquisition time (less than 0.1 ms) at the expense of lower contrast compared to the previous solution, but it enables achieving equivalent signal-to-noise ratio more rapidly. For 3D imaging purposes, the VIPA-based interferometer for FDBS may appear to offer a faster solution than the ASOPS-based setup for TDBS, which is true provided axial resolution is disregarded. In FDBS, the best axial resolution is achieved using the confocal microscopy technique, which however requires 3D scanning of the sample to obtain a complete 3D image. In contrast, TDBS only requires a 2D lateral scanning of the sample, as the third dimension (depth) is directly obtained from the time trace, and still provide an axial resolution more than twice better (*i.e.*, higher) than that of confocal Brillouin light microscopy. However, there is still room for improvement in acquisition time for TDBS.

Prior giving some hints regarding the on-going and future works, I now turn to summarize our recent applications and developments of the TDBS technique.

2.2 Characterization of elastic properties of materials in a diamond anvil cell

A field of research for which the (frequency-domain) Brillouin light scattering technique is well established as a lab tool for probing mechanical properties of transparent materials is high-pressure research. This research field is of fundamental importance for various branches of the natural sciences, including materials engineering, condensed-matter physics, physics of the Earth and planetology, as well as for predicting the consequences of earthquakes, meteorite shocks, tsunamis, and explosions. To replicate high pressure in the GPa range, the development of small footprint, safe high-pressure apparatus that can easily integrate with other laboratory equipment has been crucial in expanding the applications of high-pressure methods across multiple disciplines, such as physics, chemistry, or biology.

A prime example of such apparatus is the diamond anvil cell (DAC), which consists of two flat-culet diamond anvils facing each other, surrounding a micro-perforated metallic gasket to form an experimental chamber. The small volume of the chamber, combined with the exceptional hardness of diamonds, allows for achieving very/extremely high pressures, reaching up to hundreds of GPa. The use of diamonds in the DAC enables a wide range of experimental techniques that employ electromagnetic waves (such as optical and x-ray methods) to study materials under high pressure conditions.

Although extensively used in this field, the FDBS technique is constrained in terms of axial resolution due to the absence (as of yet) of a confocal microscopy configuration for long-working distance objectives, which are essential for focusing light through the diamond anvil within the highly-pressurized sample. Consequently, when employing FDBS to measure elastic properties of high-pressure phases of materials, it results in the averaging of properties along the entire optical path between the anvils. In cases where the materials is in a polycrystalline form, this averaging hinders the accurate determination of the single-crystal elastic properties of the tested materials. To overcome this limitation of FDBS, the TDBS technique comes into play. We have successfully applied TDBS, utilizing a setup with a mechanical delay line, to characterize high-pressure cubic H_2O ice [10] and solid argon [11].

2.2.1 Longitudinal sound velocities, elastic anisotropy, and phase transition of high-pressure cubic H_2O ice to 82 GPa

In Ref. [10], we have conducted comprehensive investigations into the elastic properties and phase transitions of cubic H_2O ice at high pressures ranging from 10 to 82 GPa. Our experimental approach employed the TDBS technique, which allowed us to precisely measure longitudinal sound velocities (v_L) in polycrystalline ice samples confined within a

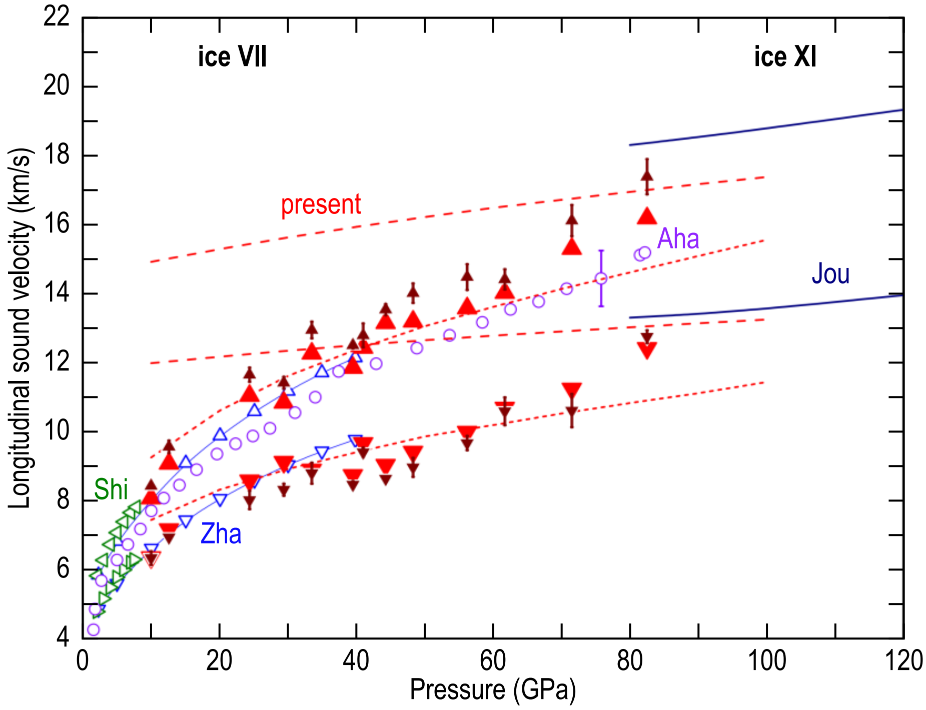

Figure 2.1: Experimental and theoretical $v_L(P)$ data for H_2O ice at high pressures as reported in our work. Symbols represent the experimental data between 10 and 82 GPa: Solid triangles pointing up and down are our experimental $v_L^{\max}(P)$ and $v_L^{\min}(P)$, respectively, obtained from time-frequency analysis with window sizes equal to one (smaller dark-red triangles with error bars) and two-and-half Brillouin oscillations (larger red triangles; error bars are smaller than the symbols). Our theoretical values of $v_{L\langle 111\rangle}(P)$ and $v_{L\langle 100\rangle}(P)$ for ice VII and ice X from ab initio calculations are represented by dotted and dashed lines, respectively. Some of the earlier experimental and theoretical results are shown for comparison. See Ref. [10] for complete references of these earlier results.

DAC (Fig. 2.1). These measurements were based on a time-frequency analysis of numerous time traces acquired at each tested pressure and utilized literature values for the H_2O ice refractive index in order to recover v_L from f_B [see eq. (2.1)]. The high axial resolution of TDBS revealed nuanced variations in v_L due to elastic anisotropy, enabling the accurate determination of the fastest and slowest sound velocities within a single crystal of cubic H_2O ice and facilitating the evaluation of existing equations of state. We extended the pressure range for measuring $v_{L\langle 111\rangle}(P)$ and $v_{L\langle 100\rangle}(P)$, achieving higher resolution and accuracy compared to earlier FDBS experiments. Notably, TDBS-derived sound velocities allowed us to assess the reliability of existing equations of state obtained via x-ray diffraction, and our data confirmed the mechanical stability criterion $(C_{11} - C_{12}) > 0$ for a specific equation of state in the entire pressure range.

Moreover, our investigations revealed significant elastic anisotropy and high hardness in cubic H_2O ice, shedding light on discrepancies observed in earlier reported equations of state and FDBS data. Ab initio calculations supported our experimental findings, indicating that ice X does not form at pressures below 80 GPa. TDBS, with its exceptional axial resolution, emerged as a robust method for transparent solids elastic property measurements even at pressures exceeding 100 GPa. In comparison, the TDBS technique demonstrates favorable attributes over FDBS, particularly in terms of enhanced axial resolution and accurate determination of sound velocities, enabling more reliable evaluation of material properties.

2.2.2 Elastic anisotropy and single-crystal moduli of solid argon up to 64 GPa

In the study published in Ref. [11], we have presented a comprehensive investigation into the elastic properties and anisotropy characterization of solid argon under high pressures, ranging from 12 to 64 GPa. Employing a combination of experimental and theoretical methods, we determined the single-crystal elastic moduli C_{ij}, shear modulus G, and Zener anisotropic ratio A for the face-centered cubic (fcc) structure of solid argon. Notably, the application of the TDBS technique enabled us to measure the maximal and minimal values of the product nV_L–a measure of elastic inhomogeneities–within compressed samples. This revealed remarkably strong deviations caused by elastic anisotropy, which were approximately twice as pronounced as those observed in classical FDBS experiments. By exploiting the unique axial resolution capability of the TDBS technique, we successfully determined the pressure dependence of longitudinal sound velocities $v_{L\langle 111\rangle}(P)$ and $v_{L\langle 100\rangle}(P)$ with precision never achieved before.

Our experimental results were corroborated by ab initio calculations, and we effectively ruled out the contribution of the hexagonal close-packed phase of argon to the observed elastic inhomogeneities. By analyzing the obtained data, we derived pressure-dependent shear moduli and validated our findings against existing theoretical models and previous FDBS measurements. The study also highlighted a significant elastic anisotropy in solid argon, reinforcing the demonstration of TDBS ability to characterize materials under extreme conditions.

In conclusion, our investigation demonstrated further the exceptional capabilities of TDBS in accurately determining elastic properties and elucidating anisotropic behaviors in transparent materials subjected to high pressures. The method high spatial resolution and sensitivity have enabled to uncover intricate details of solid argon behavior under compression, shedding light on its elastic moduli and anisotropy at unprecedented levels of precision.

2.3 Characterization of thin films of complex materials

Besides characterizing materials compressed to high pressures in a DAC, we have also been working, at atmospheric pressure conditions, on characterizing complex materials in thin film form (a few hundred nanometers thick) deposited on substrates. To address such characterization challenges, we have made use of a combination of various characterization techniques, including picosecond laser ultrasonics (with acoustic echo time-of-flight measurements and TDBS) performed at LAUM.

2.3.1 Evaluation of the structural phase transition in multiferroic thin films of $(Bi_{1-x}Pr_x)(Fe_{0.95}Mn_{0.05})O_3$

The work documented in Ref. [12] was achieved through fruitful collaborations between our team at LAUM and researchers from five other esteemed research laboratories across France, Russia, the USA, and China. This partnership allowed us to leverage collective expertise and diverse technical skills to achieve our research goals. By combining TDBS (measurements of refractive index and sound velocity product) with optical spectral reflectometry (optical thickness), scanning electron microscopy (thin film thickness), atomic force microscopy (thin film thickness, surface topography), and other sample characterization techniques, we investigated the impact of Praseodymium (Pr) substitution on the elasticity of mutiferroic $(Bi_{1-x}Pr_x)(Fe_{0.95}Mn_{0.05})O_3$ thin films (500-600 nm-thick). Through these comprehensive investigations involving ten films with varying Pr substitution concentrations deposited on two distinct substrates, Si (100) or $LaAlO_3$ (100), we unveiled a structural phase transition marked by intriguing elastic and optical anomalies. The observed minima in longitudinal sound velocity and maxima in the optical refractive index were found to correlate with the structural transition (Fig. 2.2). Importantly, our findings suggest that films with compositions near phase boundaries hold significant promise for multifunctional applications due to their exceptional ferromagnetic and ferroelectric properties.

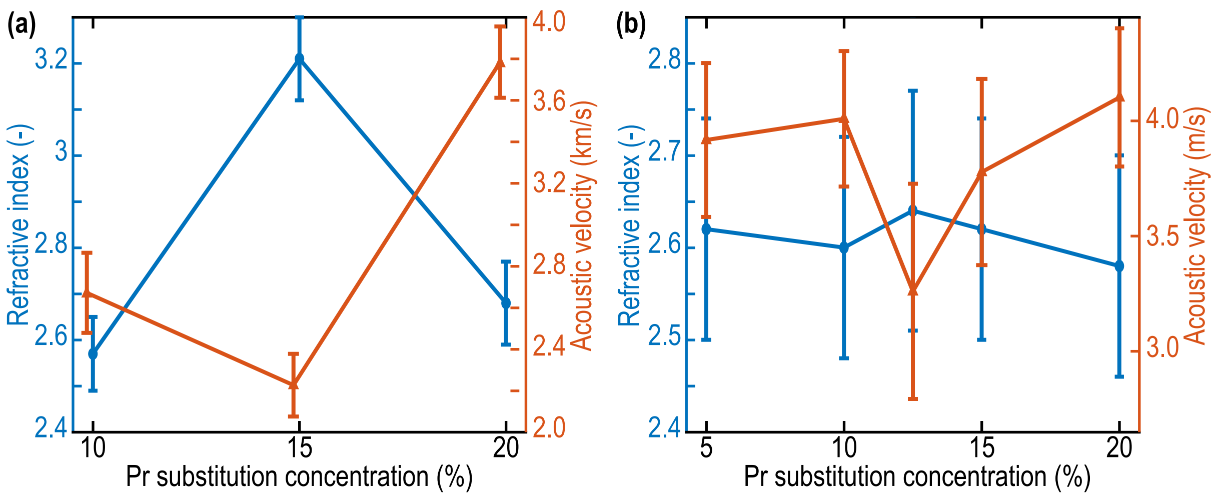

Figure 2.2: Dependence of the optical refractive index and longitudinal sound velocity on Pr substitution concentration in $(Bi_{1-x}Pr_x)(Fe_{0.95}Mn_{0.05})O_3$ films deposited on (a) Si (100) and (b) $LaAlO_3$ (100) substrates.

Moreover, these achievements pave the way for further advancements. The potential utilization of TDBS in combination with other techniques to investigate acousto-optical interaction efficiency variations across structural transitions indicates significant potential. This research direction could lead to the discovery of films with superior efficiency in acousto-optical modulation of light, holding substantial implications for the future of information and communication technologies.

2.3.2 Evaluation of optical and acoustical properties of $Ba_{1-x}Sr_xTiO_3$ thin film material library

Leveraging the versatility of our ASOPS-based setup, encompassing multiple probe light wavelengths, we undertook the exploration of a more complex structure: a material library of $Ba_{1-x}Sr_xTiO_3$ ($0 \leq x \leq 1$), realized as an ultra-thin film with a thickness of 250 nm. The genesis of this collaborative research effort was marked by our initiative to partner with the GREMAN laboratory in Tours, subsequently joined by a colleague from CEA to facilitate comprehensive characterization. The collaboration was driven by two main goals. Within our team, we sought novel systems showcasing gradients in elastic and optical properties, thereby extending the frontiers of TDBS capabilities. Concurrently, we recognized the potential synergy between our characterization technique and the expertise of the GREMAN team, renowned for their work on such graded materials. Following a year of joint efforts fueled by internal funding sources, this collaboration yielded the interesting outcomes documented in Ref. [13].

The film of $Ba_{1-x}Sr_xTiO_3$ was not dedicatedly prepared for its opto-acousto-optic evaluation by picosecond laser ultrasonics and exhibited significant lateral variations in thickness and surface roughness, making its accurate evaluation even more challenging. Addressing this challenge, this study eventually showcased a pioneering application of picosecond laser ultrasonics, based on a dedicated surface roughness modeling approach, for the comprehensive assessment of acoustical and optical properties in laterally compositionally graded epitaxial films. Through meticulous modeling, we effectively accounted for the intricate influence of surface roughness, thereby enhancing the accuracy and reliability of the measured acoustical and optical parameters.

To achieve the extraction of the sound velocity, $v_{\mathrm{ac}}(x)$, and refractive index, $n(x)$, at both green and UV optical wavelengths as a function of x, a systematic sequence of steps was followed. First, picosecond laser ultrasonics measurements were conducted along the compositional gradient. These measurements granted access to the product nv_{ac} through the precise estimation of the Brillouin frequency [Fig.2.3(a)], thanks to a fitting procedure detailed in Ref. [13]. Subsequently, the thickness of the film was determined with precision by scanning electron microscopy [Fig. 2.3(b)]. In the third phase, the time-of-flight of the acoustic pulse traversing the film [Fig. 2.3(c)] was derived.

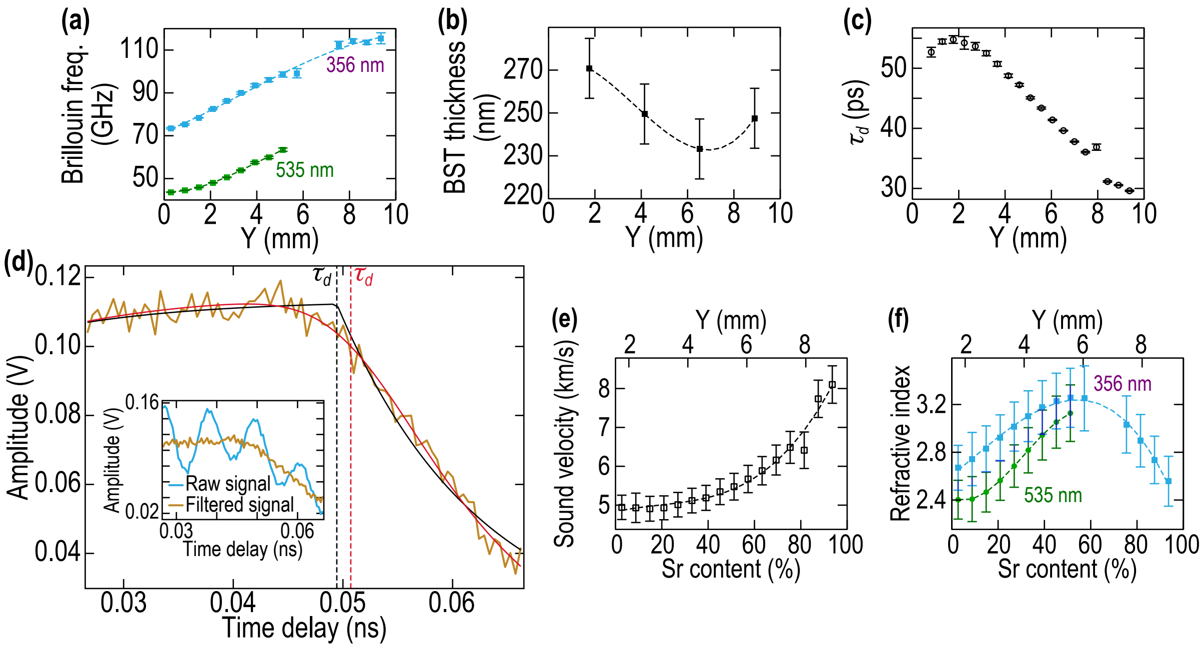

Figure 2.3: (a) Brillouin frequencies plotted against the position Y along the graded direction of the material library. The frequencies were estimated through a fitting procedure for both UV (top blue data points) and green (bottom green data points) probe light. (b) Variation in material library thickness observed as a function of Y. (c) Time-of-flight τ_d for acoustic wave propagation across the material library layer, determined via the fitting procedure illustrated in (d). Example showcasing the comparison between filtered signals (orange) and fit results with (red) and without (black) accounting for surface roughness. This example pertains to a signal with pronounced Brillouin oscillations before filtering (inset). The inclusion of surface roughness in the analysis prevents the underestimation of the time of flight τ_d. (e) Sound velocity within the material library, depicted as a function of Sr content (bottom x-axis) and Y (top x-axis). These values were estimated based on data from (b) and (c). (f) Refractive index within the material library, depicted as a function of Sr content (bottom x-axis) and Y (top x-axis). The refractive index values are derived from the data presented in (a) and (e). The top blue data points correspond to the refractive index at the UV probe wavelength (356 nm), while the bottom green data points correspond to the refractive index at the green probe wavelength (535 nm).

This was accomplished by fitting the experimentally revealed dynamics of acoustically-induced film thickness variations to a comprehensive theoretical model [Fig.2.3(d)]. Both previous steps allowed the determination of v_{ac} [Fig. 2.3(e)]. Finally, the refractive index n [Fig. 2.3(f)] was deduced from the previously measured Brillouin frequency, completing the comprehensive characterization process.

Our study made use of the power of combinatorial pulsed laser deposition technique in tandem with the ASOPS-based picosecond laser ultrasonics technique, resulting in a remarkable reduction in both deposition time and measurement duration. The combinatorial approach facilitates the simultaneous evaluation of acoustical and optical parameters across a continuous composition spread thin film, making easier the characterization process and suppressing the need for multiple samples with varying compositions. This strategy hence avoids variations of parameters from run to run. Our results showed that the fusion of advanced modeling techniques, combinatorial deposition, and ASOPS-based picosecond laser ultrasonics provides a robust and efficient/fast platform to explore and design novel nanophotonic and nanophononic structures with tailored functionalities.

2.4 Real-time monitoring of transient processes

Within this section, I will now shortly address the main achievements we have accomplished using the TDBS technique to monitor, in real time, transient processes such as structural phase transition of H_2O ice compressed in a DAC, light-induced modification in organo-silica thin films, and epoxy curing in the vicinity of a metal-epoxy interface.

2.4.1 *In situ* imaging of the dynamics of photo-induced structural phase transition

This section might have actually been titled "How to unveil structural phase transitions amid optical alignment challenges: hours of painstaking signal tuning, sudden loss, a coffee break to calm the nerves, and unexpected surprises", but that would have been a long title, wouldn't it? Yet, this title would have encapsulated the real narrative starring our postdoctoral fellow Maju Kuriakose, the driving force behind the findings detailed in Ref. [14]. Such is the charm of our research realm: picosecond laser ultrasonics experiments never fail to unveil their fair share of unexpected twists and turns.

More seriously, in Ref. [14], TDBS was successfully employed to dynamically induce a photo-induced structural phase transition in a sample featuring coexisting water ice phases VII and VI at a pressure of 2.15 GPa within a DAC at room temperature [see the sample geometry in Fig. 2.4(a)]. Real-time monitoring of the phase transition front propagation was achieved by tracking the Brillouin frequency change, which exhibited a low contrast (approximately 1%), as a function of the pump-probe time delay [see Figure 2.4(b)]. This time delay corresponds to the distance from the absorbing iron wedge embedded in the DAC, responsible for laser-based sound generation. This monitoring provided the opportunity to evaluate the velocity of the inter-phase boundary motion and the relative acoustic and optical parameters of the phases, as well as to identify their nature and speculate on the physical mechanisms initiating the phase transformation. Surprisingly, this transformation started from the water ice/diamond interface, rather than the laser-heated water ice/iron interface. The transformation front velocity was very small, about a few tens of pm/s, allowing evaluation using a setup based on a mechanical delay line. To track this slow dynamics, the signals were accumulated for 46 h (roughly 5 signals per minute) and analyzed every 10 minutes (by averaging 100 time traces) with an overlap of 50 time traces with adjacent time steps. Experiments revealed the deceleration of the

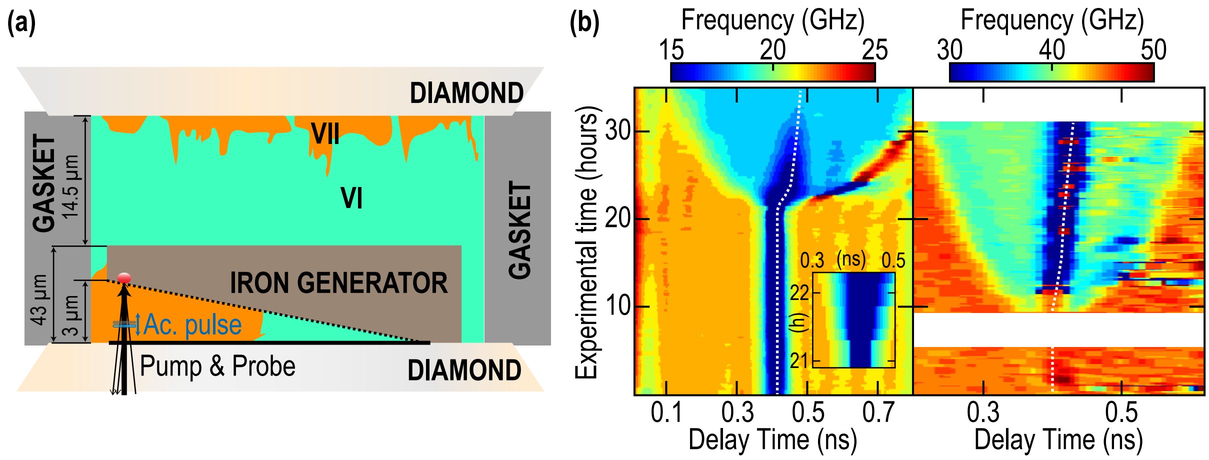

Figure 2.4: (a) Magnified cross-sectional side view of the DAC. The TDBS experiments revealed the coexistence of two phases in the sample: ice VI (light blue/green) and ice VII (orange). The opto-acoustic generator inside the sample chamber had a diameter of about 110 µm and was wedge-shaped at the bottom side. It touched the lower diamond anvil at its right end. The setup was used in two configurations: either with blue pump and red probe laser beams, or vice versa. (b) Brillouin frequency change with increasing exposure time of sample to laser radiation (vertical axis) plotted as a function of pump-probe time delay time (horizontal axis): (left) blue pump and red probe; (right) red pump and blue probe. The white dots drawn vertically at the center of each image serve as guides to identify the first reflection time of the acoustic pulse from the ice/diamond interface.

front as it propagated away from the diamond anvil, indicating that the ice transformation process was a result of the direct light interaction with the diamond, not with the water ice. Long story short, while the possibility of a recrystallization process has been ruled out by analyzing the Brillouin frequencies of both water ice phases, we concluded that the transformation of ice VII into ice VI near the ice/diamond interface could be induced by the photo-emission of electrons from diamond. Further details can be found in Ref. [14].

In these pioneering experiments, we showcased the possibility to conduct, using the TDBS technique, a one-dimensional real-time imaging of photo-induced structural phase transitions and their dynamics. While the observed process exhibited slow kinetics in this instance, this demonstration has paved the way for the practical use of TDBS as a tool to real-time monitoring of transient processes. For faster processes, an ASOPS-based experimental setup can be employed to effectively capture the dynamics, which is shortly discussed in the next subsections.

2.4.2 *In situ* imaging of a light-induced modification process in organo-silica films

In the findings detailed in Ref. [15], we explored porous organosilicate low-κ films produced using spin-on glass technology and thermally cured at 400°C for durations of one or two hours under both air and nitrogen atmospheres. This investigation enabled us to discern subtle distinctions among films cured within the critical phase, where the removal of porogen was nearly complete but matrix formation had not yet concluded. The acquired results also highlighted localized alterations in low-κ films arising from two-photon absorption. Leveraging the ASOPS-based setup, we were able to dynamically monitor the film modification process in real time via TDBS, conducting a mere 1,000 averages of the captured time traces. This approach provided a temporal resolution of 5 seconds, accounting for transfer time from the acquisition card to the computer hard disk. This revelation could bear significance for advancing active surface sites for area-selective atomic layer deposition processes, but, more importantly for the spreading use of TDBS, as it demonstrated TDBS ability to monitor *in situ* fast light-induced modifications.

2.4.3 Real-time monitoring of epoxy curing in the vicinity of a metal-epoxy interface

Utilizing the same ASOPS-based setup, albeit with lower pump and probe powers, we recently examined the temporal evolution of the curing process of a DGEBA-DETA epoxy mixture deposited onto Ti thin films on a sapphire substrate through TDBS monitoring. Two epoxy samples were investigated, one possessing a Ti film thickness of approximately $\sim$ 284 nm, and the other one having a Ti thickness of approximately $\sim$ 430nm. Each TDBS trace [Fig.2.5(a)] was obtained by averaging the signals 30,000 times (60,000 times for the second sample), resulting in a data acquisition time, and consequently a temporal resolution in the curing process, of 2 minutes (4 minutes for the second sample). In both instances, the creation of the CAPs occurred at the Ti/sapphire interface, with TDBS detection being carried out from the opposing side through the curing epoxy.

Both the Brillouin frequency [Fig.2.5(b)] and attenuation [Fig.2.5(c)] exhibited substantial fluctuations within the initial 5 hours of curing, aligning well with the reported temporal evolution of sound velocity via FDBS. As the epoxy underwent curing, the CAPs penetrated to greater depths from the interface, ranging from approximately 0.6 µm to 3 µm. Notably, the curing curves of Brillouin frequency and attenuation displayed slightly distinct behaviors between the two samples, potentially attributable to varying heating levels of the pump laser beam arising from the differing Ti layer thicknesses. After attaining saturation values, the Brillouin frequency remained largely unaltered for the epoxy sample with the larger Ti thickness (red colors). Additionally, we observed that attenuation manifested non-monotonic changes pre-saturation. The sample subject to lesser heating effects displayed an approximate 40% shift in Brillouin frequency, while attenuation showcased a more pronounced alteration, decreasing approximately 2.5 times between the uncured and cured states of the epoxy.

Our experimental findings also suggested that for the prospective application of TDBS in quantitatively non-destructive evaluations of metal-epoxy interfaces, the appropriate

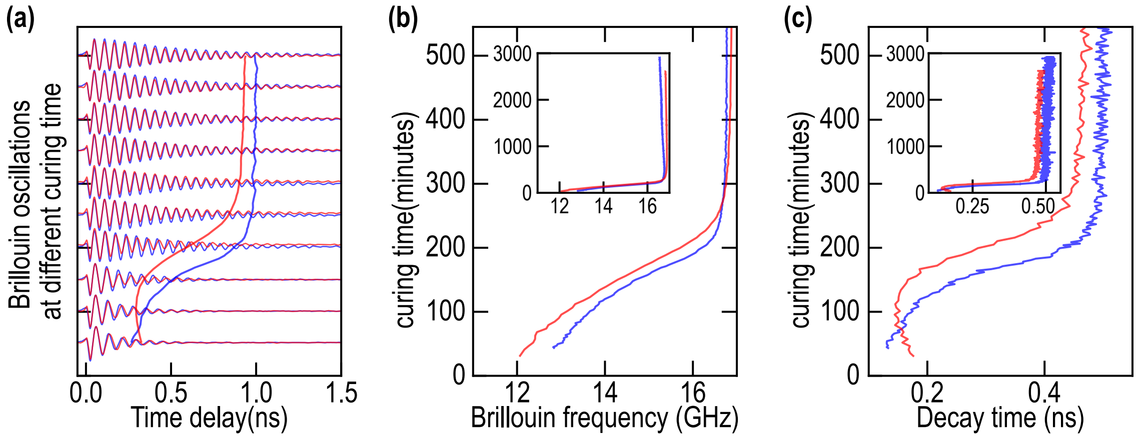

Figure 2.5: (a) Brillouin oscillations observed at various curing times, increasing from bottom-up, for the two samples. (b) Brillouin frequency and (c) decay time plotted against curing time in vertical axis. Both parameters were determined by fitting the decaying oscillations with a model signal represented as an exponentially decaying sinusoidal function of constant frequency. In (a)-(c), the blue and red colors correspond to the samples with $\sim$ 284 nm-thick and $\sim$ 430 nm-thick Ti layers, respectively.

selection of ultrafast optical pump/probe powers and wavelengths would play a pivotal role in preventing potential irreversible modifications of the epoxy. These outcomes, concomitantly with post-curing 3D TDBS imaging experiments, are currently being prepared for publication.

2.5 3D imaging of polycrystalline materials

Last but not least, this section is dedicated to presenting our efforts in demonstrating the intrinsic capabilities of the TDBS technique for achieving accurate spatially-resolved 3D images and evaluations of polycrystalline materials. The first significant advancement in this direction was made by my colleagues at LAUM in a work published in 2015 [16], where they unveiled micrometric-scale texture in a polycrystalline aggregate of H_2O ice at megabar pressures within a DAC via two-dimensional (2D) TDBS imaging.

Subsequently, in a paper published in 2016 [17], which marked my initial contribution to the field, we delved more specifically into this imaging technique, still in 2D as we utilized a setup with a mechanical delay line. In this study, we discussed the signal processing limitations affecting axial resolution when employing the conventional approach of short-time Fourier transform for time-frequency analysis, a technique from which images could be derived. Alongside this axial resolution discussion, we highlighted the potential of TDBS for achieving axial resolution limited solely by the CAP length. Furthermore, we presented 2D images of H_2O ice and argon compressed at high pressures in a DAC, with axial resolutions of approximately 250 nm and 350 nm, respectively.

In the following subsections, I will discuss our recent advancements in the 3D imaging of polycrystalline materials. These developments encompass experimental demonstrations utilizing an ASOPS-based setup, a prerequisite for such imaging, as well as a newly developed analytical theory aimed at addressing the growing complexity of the acquired signals caused by the inherent presence of inclined grain boundaries within polycrystalline materials.

2.5.1 First realization of 3D TDBS imaging of grain boundaries

After the installation of our ASOPS-based experimental setup in January/February 2018 and the subsequent initial tests, we embarked on our first 3D imaging experiment focusing on a grain boundary within a polycrystalline sample of cerium dioxide (CeO_2), also known as ceria. This sample was generously provided by our colleague, David H. Hurley, from the Idaho National Laboratory (USA), who was on his second visit to our facility and highly enthusiastic about our new experimental setup.[4]

Our collaborative efforts on this model fuel cell material led to three joint publications [18–20]. In our initial publication [18], based on experiments conducted at INL, we showcased TDBS proficiency in microstructure imaging, capturing mode- and depth-dependent acoustic velocities. This study marked the first demonstration of TDBS 3D capability through the analysis of a modest 4×5 points of the raster scan over an area of 40×50 µm^2 with step of 2 µm. Furthermore, we introduced the use of shear acoustic mode in TDBS imaging, enhancing contrast. Employing different probe light polarizations, our subsequent publication [19] exhibited the extraction of orientation information for individual crystallites by analyzing the influence of probe beam polarization on detected signal amplitude.

During David H. Hurley's visit to LAUM, we conducted a more finely resolved 2D lateral scan of the same sample over a 30×30 µm^2 area, achieving a lateral resolution of 0.469 µm with the ASOPS-based setup. Handling this substantial dataset for grain boundary reconstruction prompted the development of a two-step processing method, making use of the wavelet synchro-squeezed transform and the alphashape algorithm. Capitalizing on the detection of quasi-shear acoustic modes, we succeeded in identifying grains with improved contrast compared to the quasi-longitudinal acoustic mode, ultimately leading to 3D images of differently oriented grains from all acoustic modes (Fig. 2.6). Boundaries detected at the sample surface were verified by comparison with electron backscattering diffraction images. Remarkably, we identified a buried inclined in-

[4] Our collaboration had previously involved an attempt to detect grain boundaries during his initial visit, employing a setup based on a mechanical delay line. However, that endeavor pertained to a polycrystalline sample of tellurium dioxide (TeO_2), a more complex material due to its tetragonal crystal structure, introducing optical and elastic anisotropy. Regrettably, grain boundaries could not be detected in TeO_2, primarily attributed to limitations in lateral resolution caused by the setup's sluggishness and the presence of multiple Brillouin frequencies arising from optical and elastic anisotropy within individual grains.

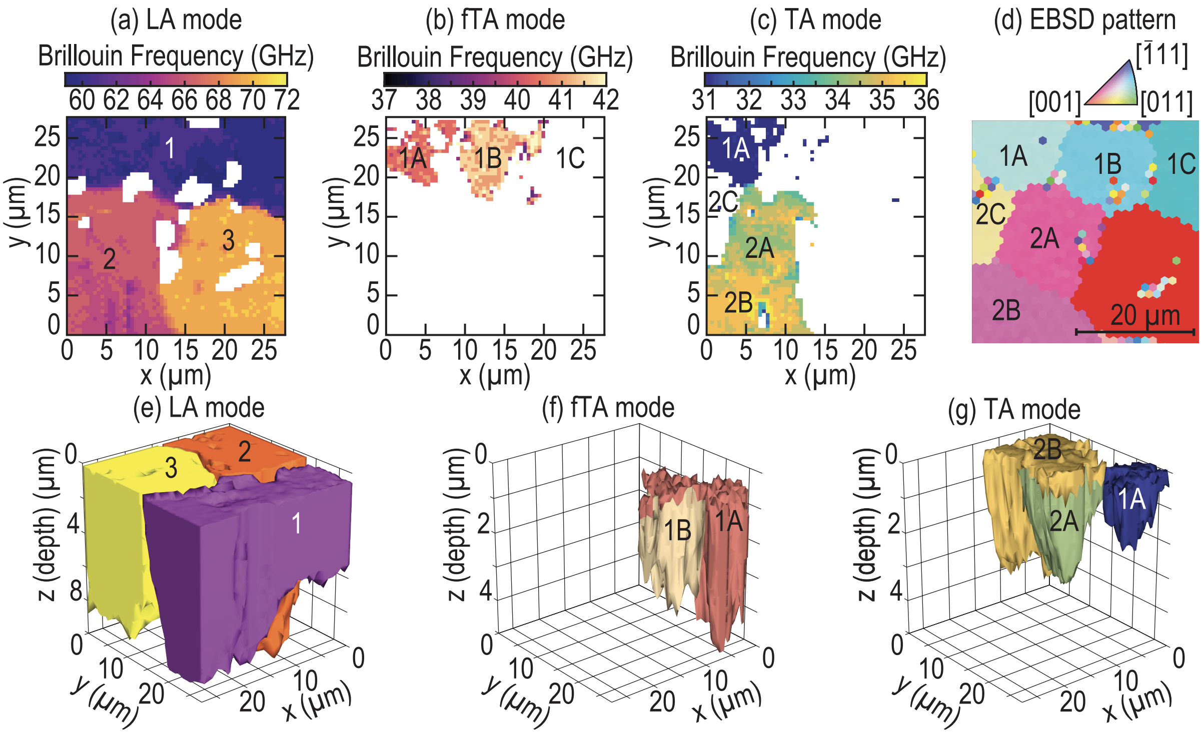

Figure 2.6: Imaging results: (a–c) upper view (free surface of the sample) of the first time segment processed by wavelet synchro-squeezed transform for three Brillouin frequency intervals, (d) electron backscattering diffraction (EBSD) image of the scanned area, and (e–g) 3D alphashape representations of grains based on the whole analysis of TDBS signals by wavelet synchro-squeezed transform for the same three frequency intervals as in (a–c). Segmented colors in (e–g) are related to the colors depicting Brillouin frequencies in (a–c). Good agreement is demonstrated between the surface TDBS reconstruction (a–c) and the EBSD image (d). The 3D alphashape results evidence the reconstitution of 3 grains (1/purple, 2/orange and 3/yellow) with the LA mode (e), 2 grains (1A/salmon-red and 1B/beige) with the fTA mode (f) and 3 grains (1A/blue, 2A/green and 2B/yellow) with the TA mode (g). TA modes allow to identify smaller grains from bigger homogeneous ones imaged by LA modes.

terface between differently oriented grains by monitoring changes in amplitude and phase of the signal associated with transverse and longitudinal waves, respectively. The estimated inclination angle of this interface highlighted TDBS sensitivity to image inclined boundaries, achieving an axial resolution of approximately 750 nm, constrained by the processing wavelet's duration.

2.5.2 First realization of 3D characterization of individual grains in a DAC by TDBS

To achieve 3D images of sample interiors, X-ray diffraction (XRD) techniques offer an alternative avenue through computed tomography (CT). However, these methods necessitate the acquisition of multiple projections achieved by rotating samples by 360 degrees

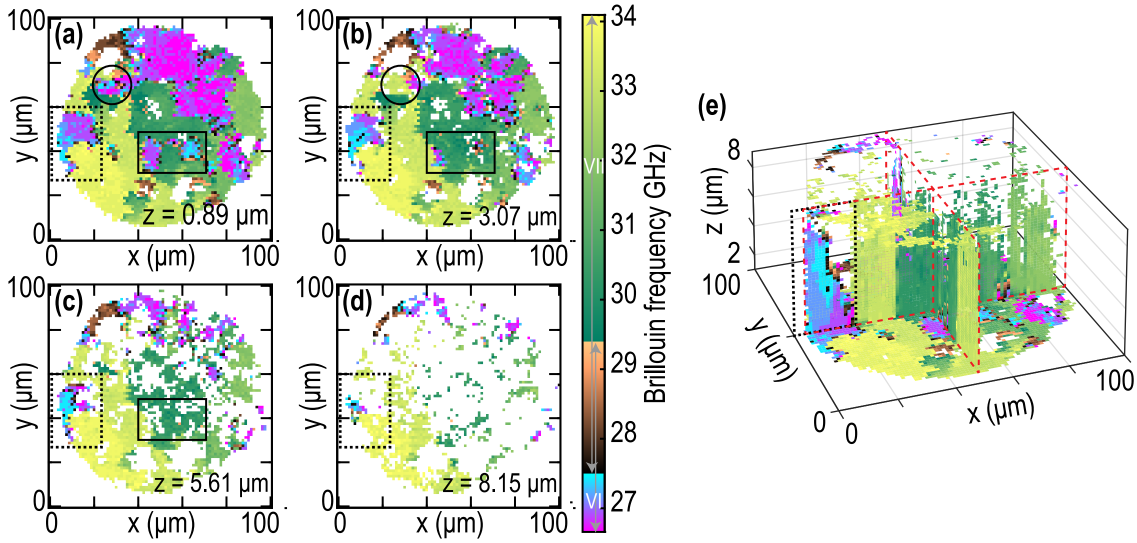

Figure 2.7: 3D TDBS imaging of the polycrystalline H_2O ice sample: (a)–(d) Slices are shown at particular positions along the z-axis indicated in the right-bottom corner of each slice, where $z = 0$ stand for the surface at which the CAPs are generated. The circles depict a volume zone composed of multiple small crystallites. The solid line rectangles depict the zones where crystallites of H_2O ice VI are seen between the optoacoustic transducer and the higher located crystallites of H_2O ice VII. The dotted line rectangles depict zones with oblique boundaries between several crystallites of H_2O ice VI. (e) 3D representation of the complete probed volume with the first and last slices, $z = 0.89$ µm and $z = 8.88$ µm (xy planes), and the middle slices (yz and xz planes) at $x = 50$ µm and $y = 50$ µm, respectively (red dashed rectangles). It is important to note that for the sake of visibility of 3D textures, the vertical z-axis is expanded by four times when compared with the x- and y-axes in (e). The color map is consistent across all parts, where blue tones represent H_2O ice VI, green tones represent H_2O ice VII, and copper tones correspond to the overlapping Brillouin frequency range of both phases. The phase identification was achieved by detecting or ruling out the presence of shear waves, as detailed in Ref. [21].

and, correspondingly, require unobstructed sample accessibility along at least two spatial directions. Nonetheless, XRD-based techniques are prone to generating artifacts coming from orientational texture and the considerable grain sizes of the examined materials. In contrast, the TDBS technique 3D images are immune to such artifacts. This is due to the direct collection of signals from each voxel, and the subsequent data treatment which is nearly free from assumptions. Furthermore, the TDBS approach facilitates 3D imaging of transparent polycrystalline samples within devices with limited access, as it only requires access along a single spatial direction and from one side only. These attributes lead us to believe that 3D images obtained using the ASOPS-based TDBS technique, featuring rapid acquisition times, could serve as a means of verifying 3D images obtained through XRD CT, particularly for samples situated within devices with limited access.

To illustrate the potential of TDBS in this context, we made efforts to demonstrate the feasibility of 3D characterization of individual grains within coexisting high-pressure H_2O

ice phases compressed in a DAC using TDBS. The experimental results and their thorough analysis were documented in Ref. [21] and the 3D TDBS image depicted in Fig. 2.7. These findings showcase TDBS's capacity to provide shape and relative coordinates of all discernible grains, regardless of their phase composition, within devices with limited access. In our experiments, this was achieved with a lateral resolution of 2.5 µm and an axial resolution of 1.2 µm. The method enabled the identification of each grain phase as well as its crystallographic orientation relative to a common coordinate system, in three dimensions, over a sample volume of about $100 \times 100 \times 10$ µm^3. Notably, our experiments marked the first observation of TDBS signals containing contributions from quasi-shear acoustic modes at high pressures within a DAC. We also provided examples of the fruitful application of TDBS imaging involving several acoustic modes simultaneously. We additionally revealed the potential to locate grain boundaries within a transparent polycrystal by identifying specific TDBS signals resulting from the simultaneous propagation of an acoustic pulse between two adjacent grains.

Collectively, we view our reported results as a significant stride toward the prospect of comprehensive 3D characterization of sample texture under extreme conditions such as high pressures and/or temperatures. Moreover, this approach offers insights into texture evolution under further compression, temperature variations, or other influencing factors. Such characterization holds particular relevance for diverse research disciplines engaged in extreme conditions. It holds the promise of *in situ* examination of mineral texture in the deep Earth, shedding light on its evolution under non-hydrostatic compression, with a level of detail currently unattainable by other techniques. This information could yield insights into the nature of seismic anisotropies observed in the Earth's mantle. Additionally, TDBS has the potential to quantitatively investigate kinetics of phase transitions and chemical reactions under high pressures and/or temperatures, as demonstrated in the previous section 2.4. It has also the potential to quantitatively investigate the relationships between the crystallographic orientations of crystallites in both the forming phase and the parent phase. Clearly, refining signal processing and the resolution of 3D TDBS imaging, enabling the recovery of quantitative shear sound velocities concurrently with longitudinal sound velocities within the entire imaged volume, will further expand the horizons of solid-state investigation under extreme conditions. This will be shortly discussed in the next chapters.

2.5.3 Theory for probe light and acoustic beams propagating at an angle and interface inclination evaluation

To address the increasing complexity of acquired signals arising from the inherent presence of inclined grain boundaries within polycrystalline materials, as observed in the previously presented 3D TDBS experimental results, we have recently developed an analytical theory.

This theory takes into account the propagation of probe light and acoustic Gaussian beams at an angle, incorporating the acousto-optic interaction at material interfaces [22].

The developed theory predicts the influence of the directivity pattern of acousto-optic interaction on TDBS signals when employing heterodyne detection of acoustically scattered light in the backward direction to the incident light. It reveals intricate relationships between various parameters, such as the carrier frequency, amplitude, and duration of acoustically induced “wave packets” in transient light reflectivity signals. Factors including the duration of the CAP, widths of both light and sound Gaussian beams, and their interaction angle are taken into account.

The theory elucidates the transient dynamics of these wave packets as they encounter material interfaces, describing how light scattering by the incident CAP transforms into scattering by the reflected and transmitted CAPs. Importantly, our theoretical framework highlights that the transient TDBS signals, accompanying CAP reflection from or transmission through an interface, are highly sensitive to the inclination angle of the interface. Consequently, these signals can serve as a valuable tool for assessing interface inclination through a single local TDBS measurement.

This perspective has been recently validated through experiments investigating the destruction of a single crystal plate of lithium niobate under non-hydrostatic pressure loading in a DAC [23]. In this case, 3D TDBS experiments revealed, among other findings, modifications of the single crystal plate with initially plane-parallel surfaces due to non-hydrostatic compression. The experiments also uncovered laterally inhomogeneous variations in plate thickness and the relative inclination of opposite surfaces. Our experimental observations, supported by an extension of the theoretical interpretations from Ref. [22], confirm that TDBS enables the evaluation of materials interface orientation and inclination locally, solely from single point measurements, thereby circumventing the need for interface profilometry.

This confirms the exciting possibilities for the expansion of TDBS imaging to analyze captivating processes and phenomena occurring during material deformation/destruction. For a more detailed exploration of the analytical model and its application for evaluating interface inclination, interested readers are referred to Refs. [22] and [23].

2.6 Concluding remarks

In this chapter, I have presented the research activities conducted at LAUM on the time-domain Brillouin scattering technique. Alongside discussing the published results since my arrival at LAUM, I’ve outlined the roadmap guiding our developments. Our goal is to showcase the wide-ranging benefits of applying this promising technique to evaluate various materials undergoing transient transformations in three dimensions and in real time. This encompasses acoustic, optical, and acousto-optical contrasts, combined with optical lateral and sub-optical axial resolution. I hope this summary has convincingly demonstrated the immense potential offered by TDBS to complement or even surpass more conventional imaging and evaluation methods for materials.

Despite its promising potential, TDBS is still in its early stages of development and requires further refinement to find broader applications across various scientific fields. A crucial aspect that can facilitate the widespread adoption of this technique is simplifying signal processing. Currently, processing signals demands a deep understanding of the technique. Offering an end-user-friendly processing method that comprehends sample complexity and addresses general inverse problems without the need for complex, time-consuming design of specific signal processing methods and analytical theories for each issue would undoubtedly advance TDBS utilization. This aspect forms a significant part of our current motivations, which I will discuss shortly in Chapter 4.

Chapter 3

Further research activities

In this penultimate chapter, I have chosen to briefly discuss other ongoing research activities in which I am actively involved. This section represents a potpourri of various investigations related to laser ultrasound, all currently in progress and being prepared for publication, or recently published. Importantly, these activities constitute a significant part of the foundation upon which the forthcoming perspectives in my research activities are built. Section 3.1 discusses a collaboration with CNRS researchers Thomas Dehoux and Maroun Abi Ghanem from Institut Lumière Matière (ILM, Lyon, France) focusing on bio-sourced materials. The subsequent sections deal with activities within the field of picosecond laser ultrasonics. Specifically, these sections entail the implementation of an an easily angle-adjustable fiber ultrafast Sagnac interferometer (Sec. 3.2) and the intriguing challenge of generating and detecting above-100 GHz surface acoustic waves (Sec. 3.3).

3.1 Bio-sourced materials presenting phononic properties

Beyond mere trendiness, the pursuit of utilizing and delving into the potentials offered by bio-sourced materials to replace non-renewable substances in our everyday devices, while also cultivating insights that will facilitate broader utilization of such materials in future generations of devices, can be regarded as crucial. This activity not only holds intrinsic significance but also holds the chance to discover exciting directions for the future. I cannot understand any better my colleagues at ILM who are dedicated to this matter on a daily base.

It all started for me at an international conference in December 2017, sparked by Maroun Abi Ghanem's impassioned yet intriguing presentation of recently obtained results[1] on the interaction of surface acoustic waves with micron-thick decellularized onion cell scaffolds. This event led us to engage in discussions about the potential interpretations of their findings. After conducting further experiments which sparked more discussions,

[1]At that time, Maroun Abi Ghanem was a postdoctoral fellow in Nicholas Boechler's team (University of California San Diego).

and performing analysis, we eventually documented this work, which was published in Ref. [24] in early January 2021 (Fig. 3.1). The samples were prepared by selecting fresh onion scales from different depths of an onion bulb, peeling off the outer half, dehydrating the samples, and placing them onto a 150 nm layer of gold coated onto a thick 1 mm glass substrate. Utilizing laser ultrasound to generate and detect broadband guided elastic waves at the gold-glass interface, we demonstrated the consistent occurrence of MHz phononic band gaps in these wild-type onion epidermises. The extinction ratios were comparable to those obtained in pillar-based locally resonant metamaterials, implying the potential for manipulating guided elastic waves using organic surfaces. This pioneering collaborative effort, combined with Maroun Abi Ghanem's recruitment at CNRS, prompted us to pursue ANR funding to further explore this intriguing avenue.

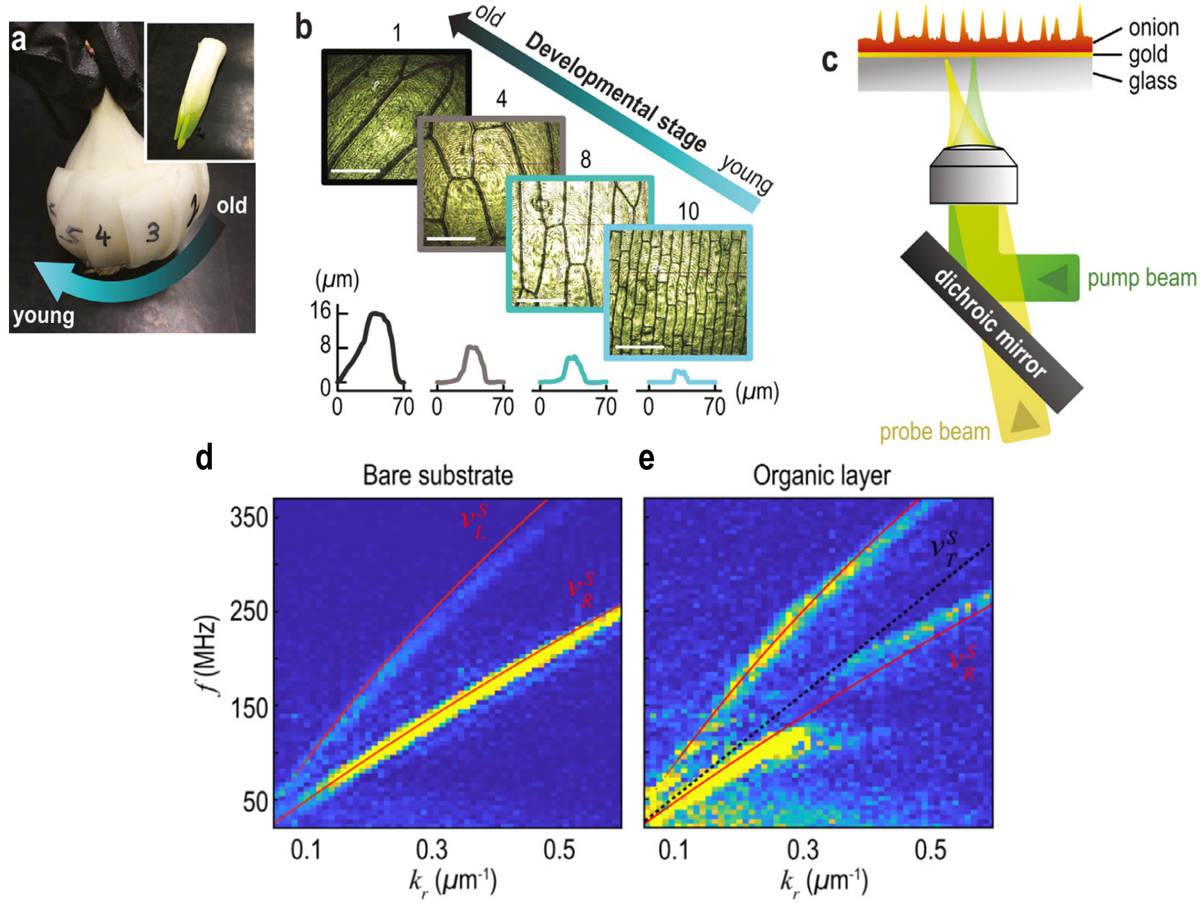

Figure 3.1: (a) Sample preparation. Epidermal layers were obtained from different depths of an onion bulb, going from the outermost (oldest) to the innermost scale (youngest), shown in the inset. (b) Optical microscope images of the peeled organic layers showing different morphogenetic profiles as a function of their developmental stage. The scale bar is 100 µm. The diagrams show profiles of the pillar-like structure of the cell wall as a function of the developmental stage (index denotes layer number). (c) Laser ultrasound setup. (d–e) Dispersion curves measured on (d) the bare substrate and (e) the organic layer. The red lines correspond to the dispersion of surface and longitudinal wave speeds calculated using a layered-half-space model. The black dashed line in (e) corresponds to the transverse wave speed in the bare substrate.

In both the aforementioned published work and our ANR project (biophoNonics), which delves deeper into this new realm, my role involves interpreting results by leveraging my expertise in analytical and numerical approaches related to guided elastic waves and anisotropic materials. This entails providing tools for simulating dispersion curves in anisotropic multilayer semi-infinite systems, accounting for cell elastic anisotropy, imperfect layer adhesion, and resonant boundary conditions due to the cell scaffolds. Additionally, explicit time-domain finite-element simulations (in collaboration with my colleague Cyril Desjouy and our postdoctoral fellow) of elastic wave propagation in such multilayer systems, while considering the same specificities, were conducted at LAUM to offer insights into the intricate yet not easily graspable detected signals. Within the scope of a master's student research internship (Nicolas Pajusco), we notably developed a `python` module enabling the rapid and accurate generation of dispersion curves in such anisotropic multilayer finite and semi-infinite systems using a spectral collocation method, which will soon be available on GitHub.

In addition to the biophotonics ANR project, our collaboration on biological materials now extends to our expertise at LAUM in picosecond laser ultrasonics. This includes two recent works where we contributed to the realization and analysis of surface acoustic waves using picosecond laser ultrasonics. Our results complement those obtained through transient grating spectroscopy for characterizing the phononic properties of two periodic biomaterials: natural nacre from Abalone shells (published in Ref. [25]) and the Mantis Shrimp's dactyl club (accepted for publication in Science [26]).

3.2 Application of ultrafast optical interferometry for opto-acousto-optical depth-profiling of materials

In Chapter 2, Section 2.1.3, we highlighted a key advantage of the TDBS technique over FDBS—its distinct axial resolution mechanism. The TDBS method theoretically offers sub-optical axial resolution, dictated solely by the length of the CAP, in contrast to FDBS. Nonetheless, realizing this anticipated axial resolution with TDBS presents challenges. In the applications discussed in Chapter 2, our reported results demonstrated that the axial resolution was determined by the processing technique employed for the oscillating component of the time signal. A trade-off should constantly be sought for achieving the best possible time and frequency resolutions simultaneously, as they are inversely related.

As part of our effort to experimentally demonstrate the ultimate axial resolution of TDBS imaging, a promising approach for overcoming the time-frequency resolution constraint involves the application of ultrafast optical interferometry as the detection method for TDBS signals, instead of the existing reflectivity change measurements. This idea, though not new to picosecond laser ultrasonics, had not been dedicatedly applied to TDBS until a recent PhD study in our team [27], supervised in collaboration with Osamu Matsuda (Hokkaido University). The utilization of ultrafast optical interferometry is known to enable the separation of variations in both amplitude and phase of the transient optical field reflectivity—a distinction not feasible when TDBS measurements rely on reflectometry. This approach holds the potential to unlock the finest axial resolution achievable by TDBS, free from limitations imposed by the probed acoustic wavelength determined by Brillouin scattering interactions, and instead governed solely by the characteristic length of the acoustic pulse. Furthermore, the signal processing barrier is circumvented, as the signal instantaneous frequency—directly proportional to the product of refractive index and acoustic velocity (nv_{ac})—can be theoretically derived directly from the measured variations of the phase with time.

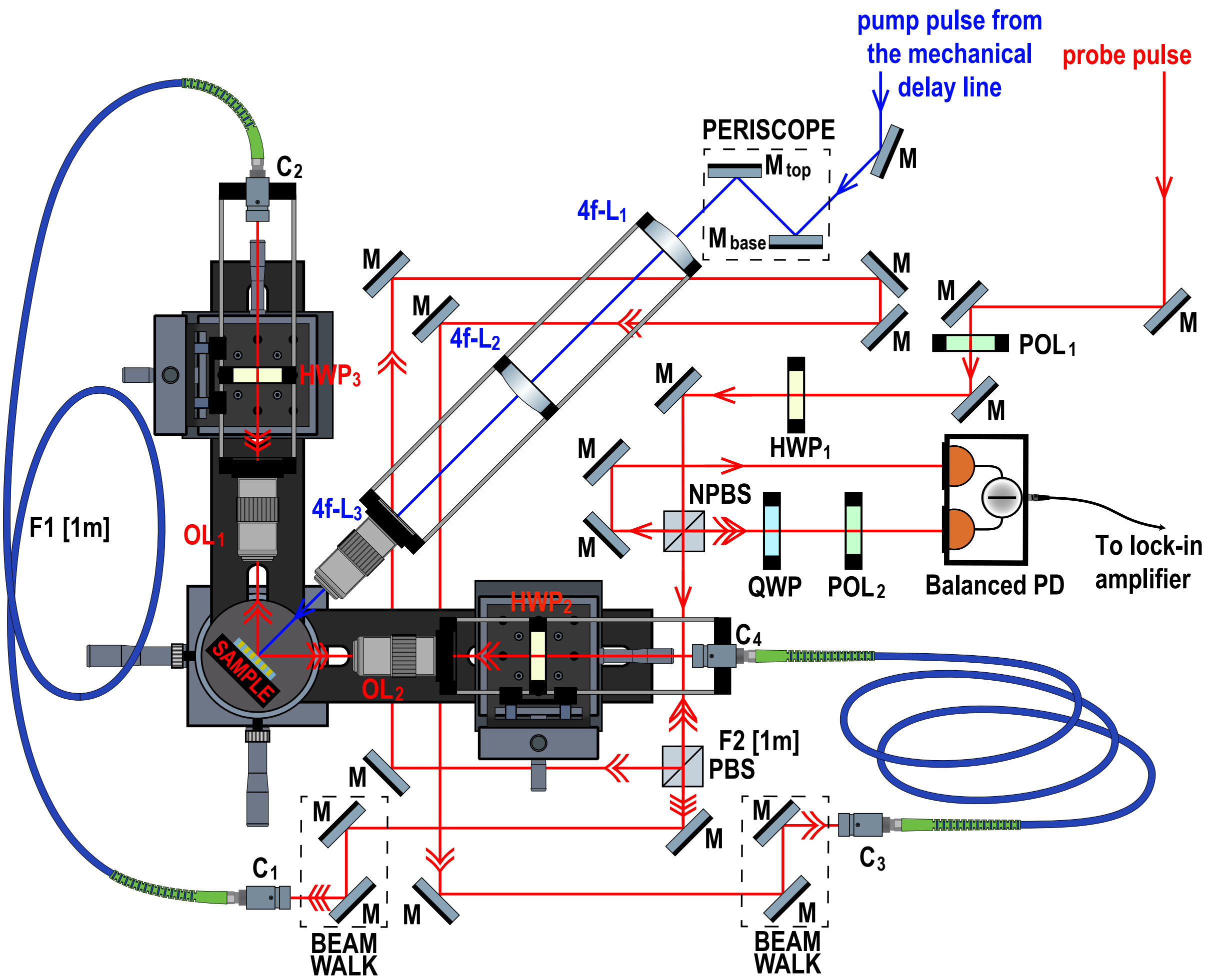

Figure 3.2: Scheme of the designed fiber-based ultrafast Sagnac interferometer with adjustable angles for ultrafast optical interferometry. The centers of rotation in both interferometer arms are precisely aligned, facilitated by a purpose-designed rotation mechanism. Perfect alignment of each element along the rail system for both paths of the probe light ensures that both probe focal spots maintain their positions upon variations in the incidence angle.

The comprehensive implementation of this concept is extensively detailed in the PhD thesis by Artem Husiev [27]. The thesis presents a meticulously designed ultrafast Sagnac interferometer, chosen for its stable and user-friendly common-path configuration (Fig. 3.2). This innovative setup incorporates optical fibers and enables measurements at varying angles of probe incidence, all precisely and continuously controlled. The key theoretical principle is based on the phase shift between acoustically-scattered light and heterodyning reflected light, specifically variations in the phase of the optical electric field reflectivity $\mathrm{d}r/r$. This phase shift, proportional to the optical thickness of the medium, encapsulates the information about the local n and v_{ac} at the CAP position, distinguishing the CAP from the stationary surfaces/interfaces reflecting the probe light. Consequently, the variation in phase shift over a small time interval $\mathrm{d}t$ depends on the local refractive index at the CAP position, $n(t)$, and the CAP displacement, $v_a(t)\mathrm{d}t$, which modulates the change in the geometrical thickness of the medium between the CAP and the stationary surfaces/interfaces. Thus, the pertinent details of $n(t)$ and $v_a(t)$ are contained within the time derivative, $\frac{\partial \varphi_r}{\partial t}(t)$, of the transient optical field reflectivity phase, $\varphi_r(t)$.

In scenarios where the CAP originates from the surface of a semi-infinite sample with depth-normal material inhomogeneity, the probe light undergoes scattering by the CAP and reflection at the surface, leading to the expression for the time derivative of the transient optical field reflectivity phase:

$$\frac{\partial \varphi_r}{\partial t}(t) = 2n(t)v_a(t)\cos[\theta(t)],, \tag{3.1}$$

where $\theta(t)$ represents the local angle between the CAP and probe light propagation directions within the medium. Hence, the determination of the local product $n(t)v_a(t)$ for a depth-inhomogeneous medium does not necessitate the discussion or definition of Brillouin oscillations or the Brillouin frequency. This product is discerned through the time derivative of the optical field transient reflectivity phase $\varphi_r(t)$, which can either be directly measured and then differentiated, or obtained experimentally by measuring the derivative directly. As the time derivative can theoretically be assessed between two sequential signal measurements, the experimental measurement of $\varphi_r(t)$ is anticipated to advance investigations into the ultimate axial resolution of TDBS imaging, a primary objective of Artem Husiev's PhD work. By virtue of the implemented interferometer, the interferometric measurements at two or more angles offer a means to independently quantify the local sound velocity and refractive index.

Throughout the course of this PhD, we delved into the anticipated performance of such an interferometric-based TDBS measurement from a theoretical standpoint. We also addressed potential limitations, particularly the reality that the detected signal encompasses more than just acoustic contributions. To address this, we proposed a signal processing methodology aimed at extracting the acoustic contributions from the experimental signals, thereby mitigating artifacts. Experimental trials on a homogeneous GaAs sample practically demonstrated the potential and constraints of the constructed interferometer. Notably, these experiments showcased a linear correlation between the transient reflectivity phase and depth in homogeneous samples. Our current efforts are focused on reporting deviations from this linear behavior in spatially inhomogeneous samples.

3.3 Picosecond surface acoustic waves

The latest addition to our ongoing activities in the realm of picosecond laser ultrasonics within our team at LAUM involves the ambitious pursuit of generating and detecting surface acoustic waves (SAWs) above 100 GHz. While I will not delve deeply into this topic here, as our results are yet to be published in peer-reviewed international journals (with the exception of the recent publication [28]), it is worth highlighting this significant effort led by my colleague Vitalyi E. Gusev, in which I am also actively involved. This topic will soon become a central focus of my work, as shortly elaborated upon in the forthcoming chapter.

In essence, the primary objective of this pursuit is to design optically controlled techniques for creating and detecting coherent SAWs with picosecond-scale periods and deeply sub-optical, nanometer-level wavelengths. These advanced SAWs hold immense potential for applications in nanometrology, nanoimaging, sensing, and manipulations across both fundamental and applied research domains. Our aim is to engineer ultrafast optoacoustic and acousto-optic processes at the nanoscale to enhance the performance of state-of-the-art 10-100 GHz coherent SAW transducers. Additionally, we aspire to extend the frequency band of these transducers by pioneering new methodologies, pushing the boundaries into the currently unattainable and unexplored (for SAWs) frequency range of 100 GHz - 1 THz. Our efforts thus far have led us to explore two primary approaches, with a third approach having taken center stage during my sabbatical year in 2023-2024, as briefly introduced in the subsequent chapter.

The first approach, subject of an intensive investigation supported by a postdoctoral Marie Skłodowska-Curie fellowship, centers on the development of transducers based on semiconductor and oxide superlattices that are cleaved perpendicular to their layer surfaces. These specialized samples, grown by our collaborators at C2N and GREMAN in France, as well as Nanjing University in China [28], yield mirror-like periodically nanostructured surfaces with an achievable pitch period theoretically as thin as a single atomic layer. By optically exciting such nanopatterned surfaces with periodic layers featuring optical, acoustical, and/or opto-acoustic contrasts, we aim to achieve the generation of SAWs within the targeted frequency range. The ballistic propagation and efficient detection of these high-frequency SAWs present key challenges we are actively addressing.

Another avenue of exploration, also supported by a dedicated postdoctoral fellow from the Institut d'Acoustique–Graduate School (Ecole Universitaire de Recherche, Le Mans

Université) in 2022, centers on engineering unconventional transducers for SAW generation and detection through the abrupt disruption of surface symmetry, taking advantage of features such as surface edges, steps, and material junctions. While we are yet to achieve SAW detection beyond 100 GHz with this method, we are currently preparing a manuscript detailing our success in generating and detecting SAWs above 20 GHz. This accomplishment was achieved by introducing a layered nanostructure approach that avoids resonance effects, thus enabling the generation and detection of wide-bandwidth GHz SAW pulses. This structure consists of opaque and transparent material layers with nanometric-scale thicknesses and closely matched acoustic impedances, ensuring minimal resonance and optimal conditions for broad-frequency SAW generation. Using a focused ion beam or precise polishing techniques, we cleaved the layered structure to create a plane with nanometric-scale surface structuring, designed for efficient optical generation and detection of nanometric SAW pulses. Materials with perovskite crystal structures were selected due to their favorable acoustic impedance properties and availability in both opaque and transparent forms, providing versatility for the study.

We envision that these high-frequency SAWs hold immense promise for revolutionizing nanoscale surface imaging and characterization. We find the prospect of evaluating their interactions with electrons, grain boundaries, and atomic-scale surface roughness to be of significant interest. This assessment is particularly sought after by researchers in the field, as it has the potential to enhance our understanding of heat transport mechanisms in nanostructures and few-layer materials. The increasing contribution of SAWs to heat transport highlights the importance of such an evaluation and aligns with the ongoing efforts of scientists dedicated to advancing this area of study. Furthermore, this work has the potential to catalyze the development of novel device categories with applications in charge carrier transfer, light manipulation, and spin control. In turn, these innovative devices could greatly enhance the landscape of future information and communication technologies.

CHAPTER

4

Concluding remarks and perspectives

As this manuscript comes to a close, this final chapter is dedicated to summarizing my research activities so far and presenting some perspectives on the work I have participated in during my time at LAUM. Before proceeding to the dedicated sections on conclusions and perspectives, I would like to acknowledge once again that none of the work I have presented would have been possible without the pioneering studies upon which it is built, my colleagues at LAUM and IMMM, and our national and international collaborators. The value of collaborative efforts with confident, clear-minded, enthusiastic, kind, and exceptionally skilled colleagues is the greatest lesson I have learned in my career so far. I am deeply indebted to all of them for shaping the researcher I am today.

4.1 Concluding remarks

Since my recruitment at Le Mans Université, my research activities have spanned several scientific themes discussed in the preceding chapters: zero-group velocity (ZGV) Lamb modes for non-destructive testing and evaluation of plates, time-domain Brillouin scattering (TDBS) for material characterization and imaging, and other activities involving surface acoustic waves (SAWs).

Our work on ZGV modes has demonstrated their capacity for damage localization, fatigue lifetime prediction, and qualitative—and potentially quantitative—assessment of cumulative fatigue damage. Experimental results showcased the ability of ZGV modes to monitor these parameters effectively. Moreover, attenuation measurements of these modes allowed us to differentiate local bond conditions (e.g., intact, with cohesion defects, or adhesion defects), proving the imaging capability of this technique. In this context, we developed a multi-layer semi-analytical model (in Python) to address electromagnetic, thermal, and elastodynamic problems sequentially. This model, freely available to the scientific and industrial community (https://doi.org/10.5281/zenodo.4301720), accounts for laser optical penetration, thermal conduction and convection, and diverse coupling conditions. Additionally, we have demonstrated the feasibility of monitoring the frequency of ZGV modes for damage localization, fatigue lifetime prediction, and damage-level assessment during fatigue processes. We also conducted an initial experimental investigation into the interaction of ZGV modes with a crack in a brittle material. The unpublished results from this study paved the way for further research aimed at understanding how resonances and guided elastic waves behave at a crack in a plate, viewed through the lens of edge waves. This work is carried out in collaboration with Sylvain Mézil and Claire Prada at the Langevin Institute in Paris.

I have extensively explored TDBS for the characterization and imaging of materials, particularly under high pressures in diamond anvil cells (DACs), including polycrystals and phase transitions. These studies have spanned various materials, such as thin films (ferroelectrics, low-dielectric constant materials), polycrystals, nanolayers, and polymers. Using longitudinal and transverse bulk acoustic waves, we employed TDBS to investigate transparent and opaque materials alike, achieving high-frequency (>10 GHz) characterization. A significant result of this work was the demonstration that TDBS offers superior axial resolution compared to conventional frequency-domain Brillouin light scattering, enhancing the characterization of elastic properties. Furthermore, this technique enabled 3D imaging of the complex textures of polycrystals and real-time monitoring of transient processes such as polymerization and phase transitions. To support these findings, we developed a comprehensive 3D analytical theory to interpret experimental results obtained in complex geometries, including non-collinear propagation of acoustic and probing electromagnetic waves. Despite the demonstrated performance of TDBS as a technique for imaging and characterizing transparent materials with unprecedented axial resolution, its adoption across various scientific fields remains limited, particularly among end-users who are not specialists in the technique. To broaden its accessibility, it is essential to develop and provide the scientific community with user-friendly signal processing methods. While ASOPS-based experimental setups are already commercially available as picosecond laser ultrasonics microscopes, the key lies in offering processing tools that account for sample complexity and general inverse problems, eliminating the need for the intricate, time-consuming signal processing and analytical theories currently required for each specific case. This overarching goal drives our ongoing TDBS research activities, alongside experimental setup enhancements such as those discussed in the previous chapter.

I have also contributed to studies of SAW propagation in bio-based materials exhibiting phononic properties, which I will continue through the fruitful collaboration with Thomas Dehoux and Maroun Abi Ghanem. This research activity with SAWs has also extended the analysis of guided elastic waves in single carbon fibers within the frame of an industrial contract with Safran (2022–2023). Finally, I also contributed to the overall goal in our team to achieve picosecond SAWs (>100 GHz) generation and detection. Concerning that latter matter, we have explored so far two approaches making use of a layered nanostructure approach to generate quasi-monochromatic or broadband GHz SAW pulses.

We demonstrated an unconventional method using cleaved superlattices (SLs) to generate and detect ∼40 GHz quasi-monochromatic SAWs. We have also recently achieved broadband SAW pulse generation at ∼30 GHz and detected reflections using a 100-nm wide absorbing layer between transparent media. Nonetheless, efficiently generating and detecting broadband picosecond SAW pulses with spectra spanning up to hundreds of GHz, without requiring complex nanosurface preparation, remains an unresolved challenge.

Through these diverse research activities, I have had the privilege of contributing to advancements in both fundamental understanding and applied methodologies. Addressing the challenges highlighted in the previous paragraphs will be central to my research in the coming years, with the overarching goal of broadly demonstrating and disseminating the benefits of using (picosecond) laser ultrasonics techniques for imaging, non-destructive evaluation, and material characterization.

4.2 Perspectives

In terms of perspectives, while not aiming to be exhaustive—since this manuscript is ultimately public—I have chosen to shortly highlight aspects of my current work at LAUM that, I hope, will capture the essence of the future main directions I intend to pursue alongside my colleagues at LAUM and our collaborators.

4.2.1 Non-destructive evaluation of a crack with edge guided waves

In collaboration with Sylvain Mézil and Claire Prada, we are currently investigating the potential of evaluating cracks using edge guided waves, as part of Juliette Alcaraz's PhD at LAUM, which is co-supervised by them and our team. A crack can be modeled as an imperfect interface between two plates, which can be experimentally simulated by bringing two plates into side contact under controllable pressure. Preliminary experiments were performed on such a simulated crack by contacting and then compressing two cleaved silicon plates (see Fig. 4.1). A scan with superimposed laser generation and detection was conducted across the crack in each configuration. In all cases, the crack was successfully localized and identified through spectral analysis of the measured resonances. Far from the crack location, resonances associated with ZGV Lamb modes were consistently observed. When the plates were just in contact, at the crack location, spectral analysis [Fig. 4.1(b)] revealed resonances at frequencies similar to, but slightly higher than, the edge resonances of a free-edge plate [Fig. 4.1(a)]. Under compression [Fig. 4.1(c)], multiple resonances appeared around two frequencies flanking the resonance observed in the just-contacting case. These intriguing observations have sparked our ongoing investigations.

To better understand the results depicted in Fig. 4.1(c), we first revisited free-edge plates made of black glass and silicon to experimentally obtain the dispersion curves of

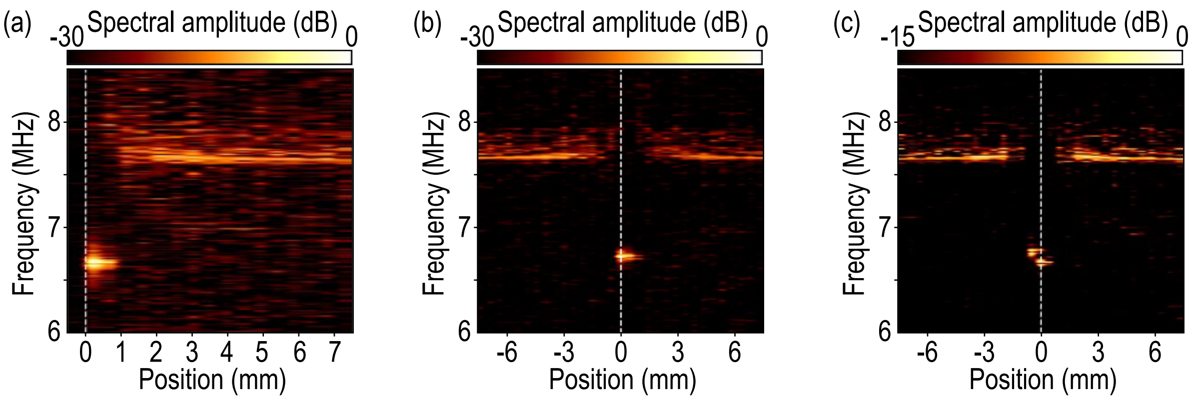

Figure 4.1: Spectral amplitude map (in dB) as a function of position from the free edge (a) or the simulated crack position (b-c) and frequency: (a) free-edge plate, (b) just-contacting plates, (c) compressed plates.

propagating edge waves for comparison with theoretical expectations, with the goal of achieving, for the first time, the experimental observation of higher-order propagating edge waves. Thus far, the comparison shows very good agreement in the case of the isotropic black glass plate. For the anisotropic silicon plate, theoretical dispersion curves are currently being prepared by Maria V. Wilde from the Department of Mathematical Theory of Elasticity and Biomechanics at Saratov State University, Russia. The next steps will involve analyzing, both experimentally and theoretically, the case of two plates brought into contact under different controllable pressures. In this scenario, we anticipate the coupling of edge waves propagating along both edges, the emergence of interface waves, and potentially nonlinear phenomena, which may explain the results shown in Fig. 4.1(c).

4.2.2 4D imaging by time-domain Brillouin scattering

As discussed in Chapter 2, we have demonstrated the capability of TDBS for 3D imaging, notably of coexisting grains of high-pressure water ice phases VI and VII [21]. With improvements to our ASOPS-based experimental setup, including an auto-balancing procedure for the photodetector that enables faster acquisition of large-area images by reducing the number of averages needed for a good signal-to-noise ratio, we are now extending these capabilities to 4D imaging. This work is part of Nicolas Pajusco's PhD research. By performing 3D imaging at successive time intervals during a single variation of non-hydrostatic high-pressure load, or after the sample reaches a steady state at different non-hydrostatic high-pressure loads, we aim to track grain movements and explore the 4D imaging potential of TDBS—capturing spatial (3D) and temporal or pressure-related (1D) changes. This allows us to investigate pressure- and stress-induced real-time relaxation, phase transitions, and crystallite formation or reorientation.

To generate significant non-hydrostatic stress, the water ice sample was loaded into a membrane diamond anvil cell (DAC) without a pressure-transmitting medium. A 20 nm-thick Au layer was deposited on the bottom diamond anvil to serve as an opto-acoustic transducer (OAT), launching coherent acoustic phonons (CAPs) into the water ice layer squeezed between the OAT and the top diamond anvil. We conducted two types of 4D imaging. First, starting from a single crystal of ice VI at 1.5 GPa (mean pressure), the mean pressure in the DAC was increased stepwise to ∼7.5 GPa and then decreased to ∼2.2 GPa. After each pressure step, a 3D TDBS image of the water ice sample was recorded, allowing us to track, in three spatial dimensions, the appearance and movements of water ice VI and VII grains in response to load changes.

Second, at ∼2.2 GPa—the expected pressure threshold for the transition from ice VII to ice VI—3D TDBS imaging of the water ice sample was continuously performed for over a month. By combining local pressure measurements using the ruby fluorescence scale with TDBS 4D capabilities, we accessed the kinetics of the transformation from a polycrystal solely composed of water ice VII to one solely composed of water ice VI. These results highlight the advantages of TDBS in high-pressure experiments over frequency-domain Brillouin light scattering techniques, which lack the axial resolution required for such investigations.

We are currently analyzing the extensive dataset collected during these two types of 4D imaging. For instance, in the time-resolved 3D imaging of the phase transition, we aim to extract the mass ratios of the two water ice phases at each imaging time point. Using these mass ratios, the known time intervals between images, and assuming that the transformation proceeds similarly in sample volumes beyond the imaging depth (limited by CAP attenuation, reflection at grain boundaries, or probe light coherence time), we aim to determine the transformation rate constant in the Arrhenius equation.

4.2.3 Neural network-assisted imaging and characterization by time-domain Brillouin scattering

Concurrently with the previously discussed demonstration of the 4D imaging capabilities of TDBS, Nicolas Pajusco's PhD work also aims to explore the potential of neural networks for solving the ill-posed inverse problem of imaging and characterizing complex materials, such as polycrystals, in 3D, by TDBS. Our approach involves the use of a specific type of neural network, known as physics-informed neural networks (PINNs), where the functional minimized by the network incorporates a mathematical model of the physics underlying the generation and detection of TDBS signals. While this is an ambitious goal, we have begun by first developing a forward numerical model.

The model is based on the finite element method applied to the weak form of the 2D Helmholtz equation, which governs the out-of-plane electric field component and accounts for coupling through the photoelastic effect with a static CAP. The CAP propagation is simulated with a discontinuous Galerkin method. To simulate a TDBS signal, the model for the electric field is solved for numerous time steps corresponding to the CAP propagation within the geometry. The numerical model is currently being evaluated and compared with analytical calculations and experimental results, such as those discussed in Refs. [22] and [23], as well as in the simpler geometry of a liquid water wedge.

This validation process has revealed challenges for the recently developed 3D analytical theory, all of which are under active investigation. These efforts are crucial steps before further pursuing the initial PINN concept.

4.2.4 Picosecond acoustics with interferometry-based detection

Besides the use of the ultrafast Sagnac interferometer discussed in the previous chapter—specifically for depth profiling by time-domain Brillouin scattering in inhomogeneous materials—another interesting application of this device, which offers easy adjustment of the incident angle, is its use for detecting SAWs via Brillouin scattering.

The schematic representation of the reflectometric configuration is shown in Fig. 4.2, where an acoustic pulse propagating at the free surface of a sample is illustrated, with the probe light incident at an angle θ_i. In the case of light backscattered by the SAW (Fig. 4.2), the ability to vary the probe incidence angle is crucial due to the momentum conservation law. Unlike Brillouin scattering by bulk waves, where refraction modifies the light wavevector projection, no such refraction occurs for surface waves. Instead, the SAW can be conceptualized as a moving Bragg mirror, introducing a Doppler shift to the scattered light. Regardless of the interpretation, momentum conservation implies:

$$q_{SAW} = 2k_i \sin\left(\theta_i\right) \ , \tag{4.1}$$

where q_{SAW} is the SAW wavenumber, and $k_i \approx k_s$ are the incident (i) and scattered (s) probe light wavenumbers, which are nearly equal since the Brillouin shift is small relative to the light frequency.

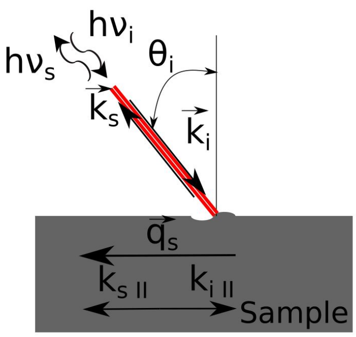

Figure 4.2: Schematic representation of the light backscattering configuration for a SAW pulse.

The second fiber arm of the Sagnac interferometer can advantageously provide a probe beam incident from the right, reflecting off the surface at an angle θ_i. This reflected light will be collinear with the light backscattered by the SAW, enabling heterodyning of the scattered light.

Future experimental steps will require strengthening the theoretical framework for SAW detection and resolving the phase-matching problem. For instance, consider a SAW wavepacket with a central frequency of 7.4 GHz (acoustic wavelength of 405.4 nm) propagating along the surface with a phase velocity of 3 nm/ps. If the probe light has a wavenumber of $k_i = 7.76$ rad/µm ($\lambda_{probe} = 810$ nm), the optimal incidence angle for efficient SAW detection via Brillouin scattering, according to Eq. (4.1), is $\theta_i = 87.4°$.

Following initial experiments with SAWs in the GHz frequency range, where Brillouin scattering is expected, higher-frequency SAWs could also be investigated. However, a theoretical basis for their (interferometric) detection would need to be developed.

4.2.5 Nanometric surface acoustic waves for non-invasive mechanical surface nanoscopy

Last but not least among our current interests is the investigation of the generation and detection of very high-frequency SAWs, as discussed in chapter 3. While numerous techniques exist for nanoscale measurements in material science, some—such as nanoindentation (destructive) or those requiring specialized environments like vacuum or cryogenic temperatures—are less practical. Common table-top techniques for mechanical surface characterization at the nanoscale, beyond the optical diffraction limit, include Scanning Probe Microscopy (SPM) and near-field methods. SAW-assisted SPM techniques utilize the atomic force microscope (AFM) tip to detect MHz SAWs by exploiting the nonlinear force-distance curve as a demodulator. This approach is not limited to SAWs and can involve bulk ultrasound, enabling the detection of nanoscale features with ultrasonic waves (<100 MHz) corresponding to wavelengths in the micrometer range.

Higher frequency waves, generated by ultrashort laser pulses, offer improved spatial resolution. Recent advancements have focused on near-field monitoring techniques, such as plasmonic nanofocusing probes, which, however, demand complex nanosurface preparations for the tip. During my sabbatical year in 2023–2024, I have been studying the fundamentals of near-field optics and AFM, as well as gaining hands-on experience with scattering-type scanning near-field optical microscopy (s-SNOM)—an AFM integrated with optical capabilities. My goal is to develop tip-assisted near-field optical and contact methods for the generation and detection of SAW pulses at nanometric wavelengths, with the aim of creating a non-invasive mechanical surface nanoscopy technique. This is a highly challenging undertaking, with a tough road ahead, but one I am eager to pursue and resolve.

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PART II

Curriculum Vitæ

Chapter 5

Curriculum Vitæ and Summary of Activities

(up to date as of December 2024)

Samuel Raetz

Maître de Conférences, normal class (6$^{\text{th}}$ grade), at Le Mans Université

Laboratoire d'Acoustique de l'Université du Mans (LAUM)

UMR 6613, Institut d'Acoustique–Graduate School (IA-GS), CNRS, Le Mans Université

Avenue Olivier Messiaen, 72085 LE MANS Cedex 9, France

5.1 Presentation

5.1.1 Personal Information

Samuel Raetz

273 Chemin de la Bossifière, 72510 REQUEIL

37 years old, born on November 10, 1986, in Lavelanet (09, Ariège)

French nationality, Married

5.1.2 Contact Details, Website, and Professional Networks

Phone : +33 (0)7 54 82 02 10

Email : samuel.raetz@univ-lemans.fr

Personal page and networks : Personal Page, Research Gate, ORCID, Google Scholar

5.1.3 Curriculum Vitæ

Degrees

2012 **PhD** in Mechanics (with Highest Honors)

Development of numerical methods for imaging optoacoustic sources in solid media

Supervised by Bertrand Audoin and co-supervised by Thomas Dehoux

University of Bordeaux

2009	**Engineering Degree** in Mathematics and Mechanics (with High Distinction) ENSEIRB-MATMECA, Bordeaux
2009	**Research Master's Degree** in Mechanics and Engineering (with High Distinction) University of Bordeaux

Professional Career

2014-Present	**Maître de Conférences at Le Mans Université (Faculty of Science and Technology)**, Research area "Opto-Acoustics, Laser Ultrasonics", team "Acoustics and Mechanics of Materials", **Laboratoire d'Acoustique de l'Université du Mans** (LAUM), Le Mans Université, UMR CNRS 6613.
2013-2014	**Postdoctoral Researcher**, Research axis "Laser Ultrasonics and Guided Elastic Waves", unconventional imaging and detection, **Institut Langevin**, ESPCI ParisTech, CNRS, PSL Université Paris, Sorbonne Université.
2012-2013	**Temporary Teaching and Research Assistant (50%)**, Laser Ultrasonics Group, Department of Physical Acoustics, **Institut de Mécanique et d'Ingénierie de Bordeaux** (I2M), University of Bordeaux, UMR CNRS 5295.
2009-2012	**PhD Student**, Laser Ultrasonics Group, Department of Physical Acoustics, **Institut de Mécanique et d'Ingénierie de Bordeaux** (I2M), University of Bordeaux, UMR CNRS 5295.

Awards and Distinctions

2024	**Junior Prize**[1] awarded by the International Photoacoustic and Photothermal Association (IPPA). This prize is given every two years to an outstanding nominated candidate under the age of 40 and approved by the Selection Committee.

[1]Citation: "*for original contributions to non-destructive testing through laser ultrasonics, including insights into non-axisymmetric optoacoustic generation in low-absorbing materials and evaluation of adhesive bonding and fatigued materials using zero-group velocity Lamb modes; for significant advancements in refining the experimental setup, innovating signal processing techniques, and supporting theoretical analyses of three-dimensional imaging of complex transparent materials using time-domain Brillouin scattering, facilitating remarkable applications of the technique, particularly in assessing and tracking the evolution of complex materials, notably*

2022 **Outstanding Reviewer Award 2022** for the journal "Measurement Science and Technology", IOP Publishing, recognizing my invaluable service in maintaining the quality and integrity of the journal's publications.

2011 **R. W. B. Stephens Prize**, awarded for the best oral presentations at the "International Congress on Ultrasonics 2011", held in Gdansk, Poland, from September 5 to 8, 2011 (ICU'2011).

Miscellaneous

2021 **Doctoral and Research Supervision Bonus (PEDR)**.

Responsibilities

2024-Present	**Elected Member of the Board of Governors of Le Mans Université**.
2023-Present	**Alternate Member of the National Council of Universities (CNU), Section 60**.
2023-Present	**Scientific Lead of the Physical, Underwater, and Ultrasonic Acoustics Group** (GAPSUS) of the French Acoustical Society (SFA).
2022-Present	**Co-Coordinator of the Wave Physics & Acoustics track** of the Master's Program in Acoustics at the Department of Acoustics, Le Mans Université (since September).
2022-Present	**Coordinator of the Laser Ultrasonics Research Group** at the Laboratoire d'Acoustique de l'Université du Mans (LAUM).
2020-Present	**Coordinator of the Communication Group** at the Department of Acoustics, Le Mans Université (except during the 2023-2024 academic year).
2017-2022	**Elected Member of the Council of the Laboratoire d'Acoustique de l'Université du Mans (LAUM)**.
2017-2023	**Appointed Member of the Doctoral School Council of Le Mans Université**.
2015-2022	**Laser Safety Officer** at LAUM.

under high pressure conditions in diamond anvil cells; and for dedicated involvement in various activities and services benefiting the photoacoustic and photothermal communities."

2010-2012 **Elected Doctoral Representative on the Council of the Doctoral School of Physical and Engineering Sciences at the University of Bordeaux** (ED No. 209).

5.2 Activity Report

5.2.1 Teaching Activities at Le Mans Université since September 2014

Since my arrival in Le Mans, at both undergraduate and master's levels, I have taught (italicized) or currently teach the following subjects (the lectures/tutorials (TD) and practical work (TP) under my responsibility are highlighted in blue) at the Faculty of Science and Technology, where I am assigned:

- geometrical optics (21h lectures/TD, 12h TP) in **L1** Acoustics and Vibrations (AV),
- fundamental equation and acoustic propagation in 1D (12h TP) in **L2** AV,
- numerical simulation I (27h TP, Python) in **L2** AV,
- numerical simulation II (21h TP, Python) in **L2** AV,
- plane wave in 3D (12h TP) in **L3** AV,
- elements of radiation (30h lectures/TD, 12h TP) in **L3** AV,
- electromagnetism and optics (9h TD) in **L3** AV,
- introduction to wave propagation in isotropic solids (12h TP) in **L3** AV,
- apprentice mentoring (typically 2 per year) in **LP** AV,
- Python basics for instrumentation (1h TD, 3h TP) in **M1** Wave Physics & Acoustics (WPA),
- elastic waves in solids (16h lectures, 4h TD) in **M1** WPA,
- student projects (about 3 per year) in **L1, L3, M1, and/or M2**,
- *optoacoustics and applications to NDT (10h lectures) in **M2** Acoustics research,*
- *Acoustics project (13.5h TD, 40h TP) in **L1** Science for Engineering (SPI, 2012-2017 accreditation),*
- *Matelec project (12h math tutorials, 12h MATLAB TP) in **L1** SPI,*
- *analog signal processing (16h TP) in **L2** SPI,*
- *3D acoustics: spherical waves (13h lectures/TD) in **L3** SPI.*

In total, I have created around ten new practical works (geometrical optics, acoustics (L2/L3 AV), numerical simulation, Python basics for instrumentation [in collaboration with two research engineers from LAUM]). For each course I manage, I create an online learning space on the UMTICE platform to provide students with my handouts (geometrical optics), slides (all), problem sets or homework assignments (undergraduate level), and past exam papers (undergraduate level).
My teaching workload, expressed in tutorial-hour equivalents (h eqTD), is distributed as shown in the table below, with the last row indicating the percentage of these hours dedicated to practical work (TP).

Year	14-15	15-16	16-17	17-18	18-19	19-20	20-21	21-22	22-23	23-24	24-25
h eqTD	128	160.5	205.5	220	217	248	250	234	219.16		186
% h TP	37.5	58	56	44	62	54	50	47	38		35

I benefited from a reduced teaching load granted to new lecturers in 2014-15 and 2015-16. The increase in teaching workload was due to the need to accommodate the rising enrollment in our programs in recent years, with a decrease observed since 2023. For the 2023-2024 academic year, I benefited from 6 months of research leave (CRCT) granted by the CNU and 6 months of delegation to CNRS.

As part of the EUR Institut d'Acoustique – Graduate School (IA-GS), a project funded under the PIA3 Investment for the Future Program in 2017, a new international master's track, "Wave Physics & Acoustics (WPA)", was launched in September 2019 (headed by Vincent Tournat, CNRS Senior Researcher). This track aims to immerse students in research by integrating them into a research project with a team at LAUM from the first semester of M1, which they can continue in M2, with the same format during the first semester (parallel to their courses) and a full-time internship in the second semester. Compared to a traditional master's program in France, WPA students have only lectures, with fewer hours but denser content (3 modules of 40h per semester). Each module is assessed through mini-projects. Thanks to this organization, students can invest 50% of their time over two years in their research projects. Since the track's inception, I have hosted one student per master's year in the "Laser Ultrasonics" research group at LAUM: one M1 student in 2019-20 and one M1 and one M2 student each year since, except in 2023-2024, when I chose not to host an M1 student due to a lack of available supervision time. This supervision is not included in the h eqTD as we collectively decided it falls under research activities. Since September 2022, I have been **co-coordinator of this WPA track** with Guillaume Pénelet (Professor), who took on sole responsibility during my CRCT/CNRS delegation year (2023-2024), except for the recruitment process, which we managed together.

From 2020 to 2022, I also participated in an Erasmus+ Knowledge Alliance project, “ASKNOW” (2020-2022), led by Manuel Melon (Professor, Le Mans Université), involving 8 European partners: KFB (Poland), KU Leuven (Belgium), RWTH Aachen (Germany), UNIZG-FER (Croatia), Le Mans Université (France), Head Acoustics (Germany), Jazzy Innovation (Poland), and Kahle Acoustics (Belgium). The ASKNOW project aimed to develop 5 interactive online courses to integrate into the ACOUCOU platform (https://acoucou.org) developed in a previous project. Together with J.-P. Dalmont (Professor, Le Mans Université) and A. Novak (Research Engineer, LAUM), I contributed (approximately 40%) to the creation of the course on the fundamentals of acoustics (30 lessons comprising three sections: lecture, interactive content, and assessment).

5.2.2 Collective and Administrative Activities

Le Mans Université

2023-present	**Elected Member of the Board of Directors (CA) of Le Mans Université**. Following the promotion of Jean-Philippe Melchior, the delegate of the FSU list for College B on the Board of Directors (a member of the CA following the 2021 elections), I accepted, being third on this list, to occupy the vacant seat starting in December 2023. To date, I have participated in 10 CAs and have delegated my voting rights for 8 meetings (including 6 between January and July 2024, during my CRCT/CNRS delegation period).
2022-present	**Coordinator of the Laser Ultrasonics Research Operation** (OR-UL) at the Laboratoire d’Acoustique de l’Université du Mans (since March 2022). This role primarily involves organizing and leading OR-UL meetings every three weeks to facilitate exchanges among the members about ongoing activities through two oral presentations per meeting, discussing scientific directions and equipment needs, and contributing to the training of students involved in the OR, both in terms of research organization and scientific exchanges.

2022-present	**Co-Coordinator of the Wave Physics & Acoustics Track** of the Master's in Acoustics at the Department of Acoustics, Le Mans Université. This role was officially established in September 2022, but I began collaborating with Guillaume Pénelet **as early as May 2022** to oversee recruitment, streamline the organization of the two years of the program, and redesign the workload distribution for students in collaboration with colleagues. We drafted the syllabus and formalized a skills matrix implemented in 2022. After this intensive period, during which we each spent an average of 2 full days weekly from May to mid-July 2022, the coordination activities now require 1 to 2 hours weekly on average (preparing timetables, organizing Advanced Lectures, weekly student meetings, evaluating applications, managing excellence scholarships, handling daily affairs, updating materials for recruitment announcements, and presenting the program). Guillaume Pénelet and I share responsibility for the two years of the Master's program. Tasks are divided equally based on our availability. For example, Guillaume manages semester-specific scheduling issues for one semester, and I handle the other. During 2023-2024, I only assisted Guillaume in reviewing applications (e.g., Études en France [approximately 15 applications handled on a rolling basis] or MonMaster [about 50 applications in 2023-2024]).
2021	**Member of the Jury for the Thesis Awar Competition** at Le Mans Université. My contribution involved reviewing the 14 submitted applications (across all disciplines), evaluating them, and participating in discussions to allocate the awards.
2020-present	**Coordinator of the Communication Group** at the Department of Acoustics, Le Mans Université (DAUM), responsible for internal communication within the department and the university, as well as external communication targeting future users (student fairs, open days, etc.) and the general public (scientific outreach leveraging DAUM's teaching and student projects). I was replaced in this role by Bruno Gazengel (Professor, Le Mans Université) for the 2023-2024 academic year.
2017-2022	**Elected Member of the LAUM Council** (Laboratoire d'Acoustique de l'Université du Mans). The LAUM council consists of 15 members (including 4 appointed) and meets approximately once a month.

2017-2023 **Appointed Member of the Doctoral College Council** at Le Mans Université. This council, which meets monthly, includes representatives from doctoral schools, research units, and doctoral students (34 members).

2015-2022 **Laser Safety Officer** at LAUM. This support role for the lab's director and the prevention officer involved assessing laser risks at LAUM, proposing preventive measures, raising awareness among newcomers about laser safety, and allocating 5% of my workload to this mission, as stipulated in the mission letter.

French Acoustical Society (SFA)

2023-present **Head of the Specialized Group** for physical acoustics, underwater acoustics, and ultrasonics (**GAPSUS**) of the **SFA**. The board consists of 16 elected members in 2023 and 2024. The board meets once a month to discuss the scientific activities of physical acoustics in France, participate in organizing the AFPAC (Anglo-French Physical Acoustics Conference) every two years in France, organize physical acoustics sessions at the CFA (French Acoustics Congress), and prepare proposals for candidates supported by GAPSUS for SFA awards and medals. I prepare the agenda and chair the GAPSUS board meetings. I also represent GAPSUS at the SFA Board of Directors (CA), which meets for a full day every two months.

2019-2022 **Secretary of the GAPSUS Specialized Group (SFA)**. The board included 18 elected members in 2019 and 2020, and 14 elected members in 2021 and 2022. The board met two to three times per year to discuss the scientific activities of physical acoustics in France, participate in organizing the AFPAC (Anglo-French Physical Acoustics Conference) every two years in France, organize physical acoustics sessions at the CFA (French Acoustics Congress), and prepare proposals for candidates supported by GAPSUS for SFA awards and medals. I wrote the minutes of the meetings.

National Council of Universities (CNU)

2023-present	**Substitute Member of the National Council of Universities (CNU), Section 60 (Mechanics, Acoustics)**. I participated in the February 2024 meeting of Section 60 regarding the **Qualification and CRCT Procedure** (2024 session, 3 days), where I reviewed 21 qualification dossiers (requiring one to two hours of expertise per dossier).

Organization of National/International Conferences, Summer Schools

2024-present	**Co-organizer of the sessions "Laser Ultrasonics, Sound-Light Interaction"** (with Dr. Thomas Dehoux) **and "Development and Design of Experiments in Physical, Underwater and Ultrasonic Acoustics"** (with Adeline Bernard) **at the 17th French Acoustics Congress**, April 27-30, 2025, Paris, France (https://cfa2025.fr/).
2023-present	**Head of the Organization of the International Summer School "Son et Lumière 2025"**, which will be held in August 18-29, 2025, at the Banyuls-sur-Mer Oceanological Observatory (https://sel2025.sciencesconf.org/). Since June 2024, after researching and booking a location, as well as preparing and submitting the thematic school application to CNRS, Alexey V. Scherbakov (TU Dortmund) joined me in leading the school, and we have been working together to prepare for this 9th edition. We are supported in this endeavor by an organizing committee of 6 people and a scientific committee of 21 international colleagues.
2023	**Co-organizer** (with Dr. Chenyin Ni and Dr. Haiyang Li) **of the session "Laser Ultrasonics Applications in Industry and Aeronautics" at the International Congress on Ultrasonics** (2023 ICU BEIJING), September 18-21, 2023, Beijing, China (https://www.2021icu.org.cn/Home/Default). The session featured 7 presentations. Along with C. Ni and H. Li, we managed the session publicity (mailing), received, reviewed, and sorted the accepted abstracts to propose the session program to the organizing committee.

2023 **Member of the Scientific Program Committee and Local Organizing Committee of the 17th International Conference on Phonon Scattering in Condensed Matter** (Phonons2023), July 2-7, 2023, Paris, France (https://phonons2023.sciencesconf.org/). My participation in the scientific program involved suggesting names and taking part in decisions about plenary and invited presentations and offering feedback on session programs. In the local organizing committee, I assisted with receiving and setting up materials, catering, and room arrangements for the poster sessions and coordinated with the caterer for breaks.

2021-2022 **Co-organizer** (with Dr. Thomas Dehoux) **of the session "Laser Ultrasonics, Sound-Light Interaction" at the 16th French Acoustics Congress**, April 11-15, 2022, Marseille, France (https://cfa2022.sciencesconf.org/resource/page/id/7). The session included 21 presentations divided into 4 sub-sessions. Together with Th. Dehoux, we managed the session publicity (mailing, individual solicitations), received, reviewed, and sorted the 21 accepted abstracts to create thematic sub-sessions. I chaired one of the sub-sessions.

2019-2020 **Co-organizer** (with Prof. Andrey Akimov) **of the session "Laser Ultrasonics" at the e-Forum Acusticum 2020**, December 7-11, 2020, Lyon, France. The session featured 28 presentations across 5 sub-sessions. Together with Prof. Akimov, we managed the session publicity (mailing, individual solicitations), received, reviewed, and sorted the 28 accepted abstracts to create thematic sub-sessions. I chaired all 5 sub-sessions.

2018-2019 **Co-organizer** (with Prof. Andrey Akimov) **of the session "NanoPhoNonics and Acoustic Metamaterials" at the METANANO 2019 conference**, July 8-12, 2019, Saint Petersburg, Russia (https://metanano.itmo.ru/2019/). The session included 21 presentations split into 4 sub-sessions. Together with Prof. Akimov, we managed the session publicity (mailing, individual solicitations), received, reviewed, and sorted the 21 accepted abstracts to create thematic sub-sessions. I chaired one of the sub-sessions.

2016-2017 **Member of the Local Organizing Committee of the French-speaking Colloquium CMOI-FLUVISU 2017**, March 20-24, 2017, Le Mans, France (https://cmoi-fluvisu.sciencesconf.org/resource/page/id/5.html). My role in the organization included advising on finances (based on my experience as treasurer for the 2016 French Acoustics Congress) and assisting with the logistics of the congress during the week before, the week of, and the weekend after the congress.

2015-2016 **Treasurer of the 13$^{\text{th}}$ French Acoustics Congress joint with the VIbrations, SHocks and Noise colloquium**, April 11-15, 2016, Le Mans, France. A total of 590 abstracts were submitted, and 776 people registered. The preparation of this event took up two full months of my work time.

2011-2012 **Organizer of the Annual "Doctoral School Day"** for the Doctoral School of Physical Sciences and Engineering at the University of Bordeaux (ED n°209).

2010 **Member of the Technical Team for the 2$^{\text{nd}}$ International Symposium on Laser-Ultrasonics (LU2010)**.

Thesis Jury

2024 **Reviewer** (External Examiner) for the thesis defended on June 6, 2024, by Ms. Menting YAO, conducted at the Engineering Research Department of The University of Nottingham (Nottingham, UK) under the supervision of Prof. Matt CLARK and co-supervised by Dr. Richard SMITH, Dr. Fernando PEREZ-COTA, and Dr. Rafael FUENTES DOMINGUEZ, titled "Development of GHz optoacoustic lenses for sub-optical resolution imaging."

2023 **Examiner** for the thesis defended on June 2, 2023, by Mr. Victor GAY-OUX, conducted at I2M (Université de Bordeaux) under the supervision of Prof. Michel CASTAINGS and co-supervised by Dr. Mathieu RÉNIER, titled "Transmission of a bounded ultrasonic beam with normal incidence through an immersed plate. Application to localized non-destructive testing of bonded assemblies."

2022 **Examiner** for the thesis defended on November 3, 2022, by Mr. Peilong YUAN, conducted at the Laboratory for Soft Matter and Biophysics (KU Leuven, Belgium) under the supervision of Prof. Christ GLORIEUX, titled "Laser ultrasonic testing methods for crack detection and plastic deformation monitoring under dynamic loading stress."

2021 **Examiner** for the thesis defended on April 27, 2021, by Ms. Louise LE RIDANT, conducted at I2M (Université de Bordeaux) under the supervision of Prof. Bertrand AUDOIN and co-supervised by Dr. Marie-Fraise PONGE, titled "Optimization of a multilayer opto-acoustic transducer for picosecond acoustics."

2021 **Examiner** for the thesis defended on March 31, 2021, by Mr. Omar ORTIZ CABELLO, conducted at C2N (Université Paris-Saclay) under the supervision of Dr. Daniel LANZILLOTTI-KIMURA and Dr. Pascale SENELLART, titled "Coherent acoustic-phonon dynamics in GaAs/AlAs heterostructures."

2016 **Examiner** for the thesis defended on March 25, 2016, by Mr. Eleftherios ANAGNOSTOPOULOS, conducted at I2M (Université de Bordeaux) under the supervision of Prof. Bertrand AUDOIN and co-supervised by Dr. Damien SÉGUR, titled "Modeling of laser-generated ultrasounds: Application to the inspection of aeronautical and composite components."

Selection Committees

2022 **Expert Member** of the jury for the external CNRS competition No. 25, IR2 body, BAP C for a position at the Langevin Institute (Paris).

2022 **External Member** of the selection committee for position No. 63 MCF 0066 at the Université Polytechnique Hauts-de-France (IEMN-DOAE, INSA Hauts-de-France; Valenciennes).

2021 **Local Member** of the selection committee for position No. 60 MCF 4291 at Le Mans Université (LAUM, UFR Sciences, ENSIM; Le Mans).

2019 **External Member** of the selection committee for position No. 63 MCF 0162 at the Université Polytechnique Hauts-de-France (IEMN-DOAE, ISTV; Valenciennes).

Reviewer Activity for International Peer-Reviewed Journals (85 for 25 journals)

- Acta Acustica (1)
- AIP Advances (1)
- Applied Optics (5)
- Applied Physics Letters (9)
- Mech. Syst. Sig. Proc. (1)
- Nano Letters (1)
- Optics & Laser Technology (1)
- Optics Letters (2)

- Applied Physics Reviews (3)
- Applied Sciences (2)
- Electronics (2)
- IEEE T-UFFC (2)
- Int. J. Thermophys. (7)
- Journal of Applied Physics (10)
- J. Phys. D: Appl. Phys. (2)
- Light: Science & Applications (3)
- Meas. Sci. Technol. (3)
- Photoacoustics (14)
- Review of Scientific Instruments (2)
- Scientific Reports (1)
- Sensors (4)
- Smart Materials and Structures (1)
- Int. J. Acoust. Vib. (1)
- J. Acoust. Soc. Am. (2)
- Ultrasonics (7)

Reviewer Activity for Research Funding Agencies

2023-2024 **Evaluator for the Agence Nationale de la Recherche (ANR)** (1 project per year at step 2). This evaluation activity consists of critically assessing the scientific excellence and quality of project construction, in accordance with international principles of competitive project selection. The final evaluation of a project is justified based on its quality and scientific ambition, its organization, the potential for its realization by the consortium, its scientific impact, and its potential outcomes.

2023 **External Evaluator for the Nottingham Research Fellowship program at the University of Nottingham.** This evaluation activity consists of assessing the value of the proposed research in the project as an expert in the field. This statement should not exceed 2 pages and should comment on the quality, impact, and opportunity of the proposal.

5.2.3 Research Activities

Keywords: laser ultrasonics, picosecond acoustics, guided elastic waves, time-domain Brillouin scattering, imaging, non-destructive testing and evaluation, signal processing

Summary of my Scientific Output and Communications

My scientific output and communications, detailed below and accessible for publications on my ORCID page (https://orcid.org/0000-0003-3683-8764), are summarized as follows.

- Publication of **35 articles in peer-reviewed international journals**, including **7 articles in a special issue** of a peer-reviewed international journal.
- Submission of **6 articles to an open-access preprint archive** (arXiv), all subsequently published in peer-reviewed international journals.
- **Participation in international conferences**, including those listed below to which I regularly contribute, for a total of **112 communications**, **including 7 invited talks** that I presented in sessions I did not organize (3 in plenary sessions):
  - Anglo-French Physical Acoustics Conference – AFPAC (2011, 2013, 2017, and all since 2019) ;
  - International Conference on Photoacoustic and Photothermal Phenomena – ICPPP (since 2015) ;
  - International Congress on Ultrasonics – ICU (since 2011) ;
  - International Symposium on Laser Ultrasonics – LU (since 2010) ;
- **Participation in national conferences**, for a total of **19 communications**. The national conference to which I regularly contribute and participate in each edition is the French Acoustics Congress (CFA) organized by the SFA.
- **Invited seminar speaker**, in France and Spain, for a total of **3 seminars**:
  - Institut de Mécanique et d'Ingénierie de Bordeaux (Université de Bordeaux, France, April 13, 2023)
  - CIC nanoGUNE (Donostia-San Sebastian, Spain, November 6, 2023)
  - Institut Langevin (ESPCI Paris – PSL – CNRS, France, November 14, 2023)
- **Course in summer school**: "Fundamentals of time-domain Brillouin scattering and its applications to depth-profiling and 3D imaging of materials" (3h), at the summer school "Optomechanics & Nanophotonics" (April 17-28, 2023, École de Physique des Houches, France).

Overview of my Research Activities prior to Le Mans Université

Before my recruitment at Le Mans Université, during my PhD, my ATER position, and my postdoctoral research, I worked on the topics described below.

- During my PhD research on laser ultrasonics, under the supervision of Prof. Bertrand Audoin and Dr. Thomas Dehoux, I studied theoretically, numerically, and experimentally the **generation of ultrasound by a non-axisymmetric thermoelastic**

source. We demonstrated the asymmetry of the **directivity patterns** of compressional and shear acoustic waves generated by such sources in optically low-absorbing media, allowing for the selection of preferred directions for acoustic wave generation through the refraction direction [A1-A3]. In my thesis manuscript, I also studied methods based on the *in silico* backpropagation of the measured normal displacement to **image optoacoustic sources in 2D and 3D geometries**, showing that the reconstruction of the depth dimension of the source is rendered impossible by the particular characteristics of cylindrical waves in the 2D case, while a good localization of the source is possible, along with the retrieval of an asymmetric signature reflecting the asymmetry of a 2D oblique volume source. In the case of a 3D volume source with axial symmetry, it was shown that the largest dimension of the acoustic source is reconstructed more accurately at the expense of the other dimension of the source, with a detailed discussion of the artifacts [Thesis manuscript].

- As a postdoctoral researcher (CNRS fellowship) at the Institut Langevin, ESPCI ParisTech, CNRS, PSL Research University, Paris, France, under the supervision of Dr. Claire Prada, I experimentally and theoretically studied the **non-destructive evaluation** of the glass/polymer coupling in a laminated glass **using Lamb modes with zero group velocity (ZGV) generated and detected by lasers** (in collaboration with Saint-Gobain Recherche, Aubervilliers, France). In this work, I studied **the effect of a non-axisymmetric thermoelastic source on the generation of ZGV Lamb modes** in optically low-absorbing plates [A4]. I also participated in the **study of interfacial stiffnesses of a three-layer structure** using ZGV Lamb modes [A5] and utilized experimental results from **Lamb waves generated and detected by a multi-element probe** to contribute to applications of a signal processing method based on the spatial Laplace transform for the recovery of complex wavenumbers [A7].

Overview of my Research Activities at Le Mans Université

Since my recruitment at Le Mans Université, I have worked on the following scientific topics:

- **Picosecond acoustics for the characterization and imaging of materials under high pressures** in diamond anvil cells (polycrystals, phase change) ([A6, A9, A10, A13, A23, A31, A32], *ANR blanc LUDACism 2012-2016*, *ANR PRCE I2T2M 2018-2023*) and, at atmospheric pressure, **in thin films** (ferroelectrics ([A12, A19, A21], *LMAc scientific project "OPTOA" 2015-2019*, *ANR PRC Up-Down 2018-2021*), material library [A27], low dielectric constant materials [A29]), **polycrystals** ([A14, A17, A24], *LMAc scientific project "OPACOP", 2018-2021*), **nanocoatings** [A15], **semiconductors** [A34] or **polymers** ([IC61, IC63, IC71,

IC78, IC83, IC94, IC97, IC98, IC105, IC108], *ANR PRCE I2T2M 2018-2023*), using primarily longitudinal or transverse bulk elastic waves in transparent materials (**time-domain Brillouin scattering**) or opaque materials (echo detection), but also very high-frequency vibrations ($>$ 10 GHz) in coatings;

- **Non-destructive and contactless testing** of composite materials [A8] or metals [A22], bonded structures [A16, A25, A26], ductile fatigued materials [A11, A18] or cracked brittle materials ([IC49, IC57, IC92, IC109], *PAPD Université Bretagne-Loire 2017*) using, in particular, **guided elastic waves** (zero-group velocity Lamb modes, in particular);

- More recently, the propagation of surface elastic waves (Rayleigh, Sezawa) in **bio-sourced materials with phononic properties** ([A20, A35], *ANR PRC "bio-phoNonics", 2021-2024*), guided elastic waves in a single carbon fiber (*industrial contract, Safran, 2022-2023*), and **picosecond surface acoustic waves (>100 GHz) generated and detected by laser (fs)** ([A33, IC66, IC67, IC70, IC79, IC84, IC86, IC91, IC98, IC99, IC100, IC102, IC111], *IA-GS postdoctoral fellowships 2019-2021, 2022, MSCA postdoctoral fellowship STSAW 2022-2024*).

Research on time-domain Brillouin scattering has notably demonstrated that this method has better axial resolution than the more conventional Brillouin light scattering technique in the frequency domain, allowing for a better characterization of elastic properties [A10, A13], and that it is capable of providing 3D imaging of the complex texture of polycrystals [A6, A14, A17, A23, A24] and real-time monitoring of transient processes such as polymerization or phase transition [A9, A29]. We have developed the necessary 3D analytical theory to understand and exploit experimental results [A31] obtained in the case of complex geometry, leading to a propagation direction of the acoustic wave that is non-collinear with the electromagnetic wave probing this acoustic wave [A32].
In the context of non-destructive testing using zero-group velocity Lamb modes, the experimental results reported in [A11] demonstrated the ability of this method to (i) locate damages, (ii) predict fatigue life, and (iii) qualitatively, and potentially quantitatively, assess the cumulative damage levels during the fatigue process. The study of the attenuation of these modes showed the possibility of a quantitative measurement that differentiates the local bonding state (good, with cohesion defects, with adhesion defects), demonstrating the imaging capability of the technique [A16]. A multi-layer semi-analytical model was also developed (in Python) and made available to the scientific and industrial community (`https://doi.org/10.5281/zenodo.4301720`), solving the electromagnetic, thermal, and elastodynamic problems sequentially, accounting for the optical penetration of the generation laser, thermal conduction and convection, as well as various optical, thermal, and mechanical coupling conditions [A25]. We also demonstrated the possibility of monitoring the frequency of a zero-group velocity Lamb mode to (i) locate damages

[A8], (ii) predict fatigue life, and (iii) qualitatively and potentially quantitatively assess the cumulative damage levels during the fatigue process [A11, A18].

Research Contracts

Responsibility in Public Scientific Contracts

2025-2029 **Responsible for the institution** in the ANR PRC "ALICE" project (AAPG2024), **735 k€** (LAUM : 42.5 k€), duration: 4 years

Quantitative imaging of the thermodynamics of phases in cells using Brillouin microscopy

Partners: ILM [UMR 5306 CNRS UCBL] (leader), I2BC [UMR 9198 CEA CNRS Univ. Paris-Saclay], Laboratoire Navier [UMR 8205 ENPC Univ. Gustave Eiffel CNRS], L2C [UMR 5221 CNRS Univ. Montpellier], LAUM [UMR 6613 CNRS LMU]

2021-2024 **Responsible for the institution** in the ANR PRC "biophoNonics" project (AAPG2020), **502 k€** (LAUM : 120 k€), duration: 3 years

Bio-sourced phononic materials

Partners: ILM [UMR 5306 CNRS UCBL] (leader), Laboratoire RDP [UMR 5667 CNRS ENS Lyon UCBL INRAE INRIA], IRPHIL [UR 4187 Univ. Lyon 3], LAUM [UMR 6613 CNRS LMU]

2018-2021 **Principal Investigator/Coordinator** of the Pari Scientifique "OPACOP 2018" (RFI Le Mans Acoustique, Région Pays de la Loire), **100 k€**, duration: 3 years

Development of opto-acousto-optic techniques for three-dimensional imaging of materials with nanoacoustic shear waves

Partners: LAUM [UMR 6613 CNRS LMU] (leader), IMMM [UMR 6283 CNRS LMU], LSPM [UPR 3407 CNRS], D.H. Hurley (USA), O. Matsuda (Japan)

2017 **Project leader** for a project awarded by the "Post-Doctoral Attractiveness Program" (PAPD) of Université Bretagne Loire (2017 campaign, Industry Department), **60 k€**, duration: 1 year

Non-destructive testing of cracked and fatigued materials by linear and nonlinear resonance of zero-group velocity Lamb modes

Participation in Public/Private Scientific Contracts

2022-2024 **Participant** in the work carried out within the framework of a **Marie Skłodowska-Curie Actions (MSCA) postdoctoral fellowship**: *Sub-THz surface acoustic waves (STSAW)*

2022-2023 **Industrial contract with SAFRAN**: *Characterization of the elastic properties of a single carbon fiber using surface elastic waves in picosecond acoustics.*

2022 **Participant** in the work carried out within the framework of a **postdoctoral fellowship funded by the Institut d'Acoustique – Graduate School (IA-GS)**: *All-optical generation and detection of sub-THz surface acoustic waves*

2019-2021 **Participant** in the work carried out within the framework of a **postdoctoral fellowship funded by the IA-GS**: *Opto-acoustic and acousto-optic transducers for above 100 GHz surface acoustic waves*

2018-2022 **Task leader** in the ANR PRCE "I2T2M" project, **391 k€** (LAUM : 250 k€), duration: 4 years

In situ three-dimensional opto-acousto-optical imaging of material transformations at the nanometric scale

Partners: LAUM [UMR 6613 CNRS LMU] (leader), IMMM [UMR 6283 CNRS LMU], LSPM [UPR 3407 CNRS], SAFRAN, NETA

2018-2021 **Participant** in the ANR PRC "UpDown" project, **475 k€** (LAUM : 92 k€), duration: 3 years

Ultra-fast photostriction in domains, walls, and ferroelectric nanostructures

Partners: IMMM [UMR 6283 CNRS LMU] (leader), LAUM [UMR 6613 CNRS LMU], SPMS [UMR 8580 CNRS CentraleSupélec], C2N [UMR 9001 CNRS Univ. Paris-Saclay]

2018-2019 **Participant** in the CNRS Instrumentation aux limites "BioPAN" project, **29 k€**

Partners: IMMM [UMR 6283 CNRS UCBL] (leader), LAUM [UMR 6613 CNRS LMU]

2018-2019 **Participant** in the work carried out within the framework of a **post-doctoral fellowship funded by the Acoustic Hub®** (Région Pays de la Loire project): *Opto-acousto-optical quantitative imaging of material elasticity and grain boundaries*

2017-2020 **Co-supervisor of the CIFRE thesis of Romain Hodé, co-financed by SAFRAN** and the National Association for Research and Technology (ANRT): *Development of laser ultrasonic methods for the nondestructive evaluation of bonded aeronautical assemblies*

2015-2019 **Participant** in the Pari Scientifique "OPTOA 2015" (RFI Le Mans Acoustique, Région Pays de la Loire), **150 k€**, duration: 3 years

GHz ferroelectric optically controlled acoustic transducers

Partners: LAUM [UMR 6613 CNRS LMU] (50% lead), IMMM [UMR 6283 CNRS LMU] (50% lead), A. Lomonosov (Russia), O. Matsuda (Japan)

2014-2016 **Participant** in the ANR blanc "LUDACism" project, **480 k€** (LAUM : 157.6 k€), duration: 4 years

Laser ultrasonics in diamond anvil cell for investigation of simple molecular compounds at ultrahigh pressures

Partners: LAUM [UMR 6613 CNRS LMU] (leader), IMMM [UMR 6283 CNRS LMU], LSPM [UPR 3407 CNRS]

National and International Scientific Collaborations since September 2014

The Ultrasonics Lasers research group I joined upon arriving at Le Mans is definitively focused on scientific collaborations. I have thus benefited from numerous ongoing collaborations and helped develop new ones through my previous work and/or professional relationships. It is also clear that the strategic projects underway at LAUM when I arrived, such as the RFI LMAc and Acoustic Hub®(see international collaborations), complemented the dynamism of the permanent members of the research group in terms of collaborations. The lists of national and international collaborators below briefly present the collaborations I have been involved in or helped initiate (highlighted in blue) for LAUM, as well as a list of joint projects and publications (conference presentations, although present, are not listed here).

Collaborations at Le Mans Université

- IMMM, UMR 6283 CNRS, Le Mans Université

- Pascal Ruello, Gwenaëlle Vaudel, Vincent Juvé. Joint projects and publications: ANR *Up-Down*, Pari Scientifique LMAc "OPTOA 2015", Instrumentation aux Limites "BioPAN", [A12, A19, A21, App3, App5], 10 conference presentations
- Alain Bulou. Joint projects and publications: ANR *I2T2M*, Pari Scientifique LMAc "OPACOP 2018", [A6, A9, A10, A12, A13, A21, A23, A31, App4, App5], 32 conference presentations
- Erwan Nicol. Joint project: ANR *I2T2M*, 7 oral conference presentations
- Nicolas Delorme. Joint project and publication: Pari Scientifique LMAc "OPTOA 2015", Instrumentation aux Limites "BioPAN", [A12, A33], 8 oral conference presentations

- LAUM, UMR 6613 CNRS, Le Mans Université
  - Vitali Goussev (V. E. Gusev), Nikolay Chigarev, Vincent Tournat. Joint projects and publications: ANR *I2T2M*, ANR *UpDown*, ANR *LUDACism*, Pari Scientifique "OPACOP 2018" and "OPTOA 2015", CIFRE ANRT Scholarship, PAPD 2017, Instrumentation aux Limites "BioPAN", post-doc Acoustic Hub®, post-doc MSCA, post-docs IA-GS, [A6, A8-A19, A21-A34, App3-App6], 111 conference presentations
  - Jean-Philippe Groby, Aroune Duclos, Alan Geslain. Joint publications: [A7, A22], 2 oral conference presentations

National Collaborations

- Andreas Zerr (LSPM, UPR 3407 CNRS, Université Paris 13). Joint projects and publications: ANR *LUDACism*, ANR *I2T2M*, Pari Scientifique "OPACOP 2018", [A6, A9, A10, A13, A23, A30, A31, App4], 36 conference presentations
- Mathieu Ducousso (SAFRAN Group, Magny-les-Hameaux). Joint projects and publications: CIFRE ANRT Scholarship, ANR *I2T2M*, [A16, A25, A26], 16 oral conference presentations
- Thomas Dehoux, Maroun Abi Ghanem (ILM, UMR 5306 CNRS, Université Lyon 1). Joint projects and publications: ANR *biophoNonics*, [App2, A7, A20, A35], 6 oral conference presentations in this context
- Jérôme Wolfman, Béatrice Negulescu (Laboratoire GREMAN, UMR CNRS 7347, University of Tours, INSA CVL). Joint project and publication: ***submission of an ANR project on very high-frequency surface acoustic waves (with cleaved super-lattices)*** (with C2N) to the 2022 and 2023 call for projects (not

accepted), samples for postdoctoral research by S. Sandeep and R. Delalande, [A27], 5 oral conference presentations

- Sylvain Mézil, Claire Prada (Langevin Institute, ESPCI ParisTech, CNRS, PSL University Paris, Sorbonne University). Project: PhD thesis CDE by J. Alcaraz, 2 oral conference presentations in this context; joint publications before my recruitment at Le Mans University [App1, A4, A5, A7], 4 oral conference presentations
- Daniel Lanzillotti Kimura, Aristide Lemaître, Martina Morassi (C2N, UMR 9001 CNRS, University Paris-Saclay). Joint projects: ***submission of an ANR project on very high-frequency surface acoustic waves (with cleaved super-lattices)*** (with GREMAN) to the 2022 and 2023 call for projects (not accepted), samples for the postdoctoral research (MSCA-STSAW) of C. Li, 3 oral conference presentations

International Collaborations

- Prof. Osamu Matsuda, Faculty of Engineering, Hokkaido University (Japan), **invited since 2014** as part of the **Acoustic Hub® (Pays de la Loire Region project) and then IA-GS** (except in 2020-2021 due to COVID, visit rescheduled to December 2022) and co-supervisor of the PhD of Artem Husiev. No articles where I am a co-author yet, but several articles where my LAUM colleagues are co-authors. 2 oral conference presentations.
- Dr. David H. Hurley, Director, Center for Thermal Energy Transport under Irradiation Materials Science and Engineering Department, Idaho National Laboratory (USA), invited as part of the **Acoustic Hub® (Pays de la Loire Region project) in 2017 and 2018**. Joint publications: [A14, A17, A24, A32, App6], 9 oral conference presentations
- Prof. Andrey Akimov, Department of Physics and Astronomy, University of Nottingham (UK), invited as part of the **Acoustic Hub® (Pays de la Loire Region project) in 2019**. Joint publication: [A7], 2 oral conference presentations, co-organization of 2 sessions at 2 international conferences (Metanano 2019 and FA 2020)
- Asst. Prof. Xiaodong Xu, Department of Science & Engineering, Nanjing University (China), whom I invited as part of the **incoming international mobility program** of Le Mans University **from 13/01/2020 to 14/02/2020** and again from 04/07/2024 to 24/07/2024 (canceled due to visa processing delays). The travel expenses (travel, accommodation, meals) for the invited researcher were covered by this program.
- Prof. Mikhail R. Baklanov, European Centre for Knowledge and Technology Transfer (EUROTEX) (Belgium), provided us with samples **in 2022** of materials with

low dielectric constant for characterization and use as inhomogeneous samples in the context of Brillouin interferometry (following the PhD of A. Husiev). Joint publication: [A29], 2 oral conference presentations.

- Dr. Rainer Hillenbrand, CIC nanoGUNE (San Sebastián, Basque Country, Spain), with whom the collaboration started in **2022**, and has been intensifying in 2023-2024 as part of my CRCT and my CNRS delegation.

For information, the Acoustic Hub® program (AAP Connect Talent, Pays de la Loire Region), led by Vincent Tournat (CNRS Research Director at LAUM), aimed to give recognized international researchers the opportunity to collaborate and develop research projects in the field of acoustics at Le Mans, by funding repeated research visits (from 1 to 3 months) over the years. For the most promising and strategic projects, additional funding (PhD or postdoctoral scholarships) could be obtained to strengthen the collaboration, which was the case for our research theme (with Osamu Matsuda). This program was extended as part of the Institut d'Acoustique-Graduate School (IA-GS).

Supervision of young researchers

Supervision of postdoctoral researchers

	LAST NAME	First Name	Title [project, communications]	Start-End	Supervision	Co-supervisors
1	KURIAKOSE	Maju	Picosecond laser ultrasound for imaging compressed polycrystalline materials [ANR LUDACism, A6, A9, A10, A13]	15/01/2015 - 15/01/2016	30%	V. Goussev 30% N. Chigarev 40%
2	KURIAKOSE	Maju	Laser excitation of surface acoustic waves in ferroelectrics [OPTOA2015, A12]	10/01/2016 - 10/01/2017	20%	P. Ruello 20% V. Goussev 20% N. Chigarev 40%
3	DE LIMA SAVI	Elton	Opto-acousto-optical quantitative imaging of material elasticity and grain boundaries [Acoustic Hub, A12, A15, A23, A24]	26/02/2018 - 25/02/2019	40%	D. Hurley 30% V. Goussev 30%
4	LI	Haiyang	Sensitive non-destructive testing of cracked and fatigued materials by linear and nonlinear resonance of zero group velocity Lamb modes [PAPD, IC49, IC57, IC109]	01/03/2018 - 28/02/2019	70%	V. Goussev 30%
5	SANDEEP	Sathyan	In situ 3D opto-acousto-optical imaging of materials transformations at nanoscale [ANR I2T2M + IA-GS, A23, A27, A29, A31, A35]	01/10/2019 - 24/09/2023	30%	V. Goussev 20% N. Chigarev 30% E. Nicol 20%

6	LI	Changxiu	Opto-acoustic and acousto-optic transducers for above 100 GHz surface acoustic waves [IA-GS, A33]	04/11/2019 - 03/05/2021	35%	V. Goussev 30% N. Chigarev 35%
7	LE RIDANT	Louise	Modélisation de la propagation d'ondes élastiques guidées dans des matériaux phononiques bio-sourcés anisotropes et problèmes inverses [ANR biophoNonics, FC17, IC85]	06/09/2021 - 05/09/2023	100%	
8	DELALANDE	Ronan	All-optical generation and detection of sub-THz surface acoustic waves [IA-GS, IC86, IC102, IC111]	01/01/2022 - 31/12/2022	35%	V. Goussev 30% N. Chigarev 35%
9	LI	Changxiu	Sub-THz surface acoustic waves [MSCA-STSAW, A33]	01/02/2022 - 31/01/2024	30%	V. Goussev 40% N. Chigarev 30%

Supervision of PhD Students

The supervision rates below are the official rates imposed by the doctoral school of Engineering Sciences and Systems (SIS) No. 602, which I am part of. It sets a minimum of 40% for the PhD supervisor's rate and 30% for the other two supervisors when there are three. Apart from Nikolay Chigarev, a research engineer at Le Mans University, and Sylvain Mézil, a research fellow at CNRS (Langevin Institute, Paris), the other co-supervisors are University Professors (V. Goussev, R. Fablet) or CNRS Research Directors (V. Tournat), as I do not yet have the habilitation to supervise research.

In practice, I have been at the forefront of the daily supervision of Guqi Yan and Romain Hodé, whom I trained in experimental laser ultrasonics methods, guided wave theory (particularly for zero group velocity Lamb modes), and for whom I was the primary reviewer of articles, the thesis manuscript, and the first to review, correct, and rehearse presentation slides and posters for conferences.

My involvement in Théo Thréard's PhD was similar, except for the initial article reviews and thesis manuscript, which were handled by V. Goussev (I was the second reviewer), and the training in picosecond acoustic experimental methods provided by N. Chigarev and later by S. Sandeep (postdoctoral researcher). For Théo, I was in charge of signal processing, which was central to the imaging method used in his thesis.

For Artem Husiev's thesis, N. Chigarev and O. Matsuda provided training in picosecond acoustic experimental methods. O. Matsuda and I oversaw the reflection on the experimental setup at the heart of Artem's thesis, and I was responsible for managing the equipment purchases. As with Guqi and Romain, I was the primary reviewer of the article (currently being finalized), the thesis manuscript, and the first to review, correct, and rehearse presentation slides and posters for conferences.

For Juliette Alcaraz's thesis, I provided her with training in experimental laser ultrasonics methods and the theory of guided elastic waves in plates or on plate edges. Sylvain

Mézil follows and participates in the supervision through weekly progress meetings and discussions on a dedicated collaborative communication platform (Slack) for Juliette's thesis. Both of us ensured the first review and rehearsal of Juliette's slides for the French Acoustics Congress and the International Congress on Ultrasonics, as well as her poster for the Sound and Light Summer School. Vitali Goussev supervises this thesis work by attending monthly progress meetings.
Finally, for Nicolas Pajusco's thesis, my involvement is similar to that of Guqi Yan and Romain Hodé's theses. In this thesis supervision, Ronan Fablet (PR, Lab-STICC, Brest) will contribute his expertise in physics-informed neural networks when the groundwork is ready. With Vitali Goussev, we provide training in laser generation theory and laser detection via Brillouin scattering of acoustic waves in transparent inhomogeneous elastic media, which is essential for the success of this thesis. The experimental training was provided by S. Sandeep during the first year of the thesis.

	LAST NAME	**First Name**	**Title [contract/project, communications]**	**Start-End**	**Super-vision**	**Co-supervisors**
1	YAN	Guqi	Zero-group-velocity Lamb modes in laser ultrasonics: fatigue monitoring and material characterization [CDE[2], A11, A18, A22]	01/10/2015 - 20/11/2018	30%	V. Tournat 40% V. Goussev 30%
2	HODÉ	Romain	Development of laser ultrasonic methods for the nondestructive evaluation of bonded aeronautical assemblies [CIFRE Safran, A16, A25, A26]	01/10/2017 - 24/11/2020	30%	V. Tournat 40% V. Goussev 30% M. Ducousso (SAFRAN)
3	THRÉARD	Théo	Development of opto-acousto-optical methods for three-dimensional material imaging with nano-acoustic longitudinal and transversal waves [OPACOP2018, A23, A24, A27, A31, A32, A33, App6]	01/10/2018 - 06/12/2021	30%	V. Goussev 40% N. Chigarev 30%
4	HUSIEV	Artem	Application of ultrafast optical interferometry for opto-acousto-optical depth-profiling of materials [Acoustic Hub, A29, IC58, IC66-67, IC69, IC74]	01/11/2017 - 14/12/2022 Extension COVID	30%	V. Goussev 40% N. Chigarev 30% O. Matsuda (Japon)
5	ALCARAZ	Juliette	Sensitive non-destructive testing of cracked materials by linear and nonlinear resonances in plates: zero-group-velocity Lamb modes and edge resonances [CDE, A34, FC16, FC18, IC92, IC109]	01/10/2021 -	30 %	V. Goussev 40% S. Mézil 30% (Institut Langevin)
6	PAJUSCO	Nicolas	4D quantitative opto-acousto-optical imaging of transient material transformations [CDE, A31, IC83, IC87-88, IC93-96, IC99, IC101, IC103-105, IC107-108, IC110, IC112]	19/09/2022 -	30%	V. Goussev 40% R. Fablet 30% (Lab-STICC)

[2] Contrat doctoral d'établissement

Supervision of internships for second-year Master's Research students

	LAST NAME	First Name	Title [communications]	Start-End	Super-vision	Co-supervisors
1	HUSIEV	Artem	Optoacoustic nanoscopy [FC9]	01/03/2017 - 30/09/2017	60%	N. Delorme 5% N. Chigarev 25% V. Goussev 10%
2	HODÉ	Romain	Characterization of aeronautical bonding by synthesis of laser ultrasound wavefronts	01/04/2017 - 30/09/2017	90%	M. Ducousso 5% V. Tournat 5%
3	MÉTEYER	Erwan	Laser ultrasonics in micro-structures made by 3D printing [FC9, IC35, IC38]	01/10/2017 - 15/07/2018	70%	V. Tournat 30%
4	VOROBIOV	Anton	Optoacoustic nanoscopy	01/03/2019 - 26/07/2019	30%	A. Husiev 60% N. Chigarev 10%
5	DOVERI	Élise	Focused laser ultrasound in materials for bonding studies and acoustic imaging	01/10/2019 - 24/07/2020	100%	
6	ALCARAZ	Juliette	Sensitive non-destructive testing of cracked materials by linear and nonlinear resonances in plates: zero-group-velocity Lamb modes and edge resonances [IC57]	08/09/2020 - 23/07/2021	100%	
7	PAJUSCO	Nicolas	Guided elastic waves in a multi-layer anisotropic system with finite and semi-infinite cross-section: analytical and numerical approaches [IC78]	06/09/2021 - 15/07/2022	100%	
8	GOUTIER	Lou-Anne	Experimental demonstration of collinear interaction in time-domain Brillouin scattering	06/09/2022 - 15/07/2023	70%	N. Chigarev 15% V. Goussev 15%
9	LI	Ruosong	Time-domain Brillouin scattering and the application in time-domain Brillouin scattering imaging of ultrafast optical interferometry	06/09/2023 - 01/02/2024	80%	S. Sathyan 20%

5.3 Scientific Production

My name is in bold, the names of our master's students, PhD students, and post-doctoral researchers are underlined. The name of the presenter of conference presentations is marked with an asterisk (*). The large number of authors for some articles and conference presentations is due to the collaborations between laboratories and the desire to include study and research engineers as authors instead of just acknowledging them in the acknowledgments section. The years are highlighted in green, notable facts are highlighted in blue. The 7 articles in special issues all followed a standard peer review process and are identified by a gray highlight for the article number. The quartile (Scopus) of each journal at the time of publication is highlighted in orange.

5.3.1 Articles in peer-reviewed international journals

[A35] N. A. Alderete, S. Sathyan, **S. Raetz**, J. Margueritat, M. Asgari, N. Boechler, M. A. Ghanem, H. D. Espinosa, *Characterization of the phononic landscape of natural nacre from abalone shells*, **Small**, 2407959 (2024). [Q1] [https://doi.org/10.1002/smll.202407959]

[A34] N. Chigarev, K. Strzalkowski, V. Gusev, P. Sedzicki, J. Alcaraz, **S. Raetz**, J. Zakrzewski, *Evaluation of the sound velocities of cadmium zinc telluride crystals by time-domain Brillouin scattering technique*, **Applied Physics A 130(10)**, 732 (2024). [Q2] [https://doi.org/10.1007/s00339-024-07898-6]

[A33] C. Li, N. Chigarev, T. Thréard, K. Zhang, N. Delorme, V. Tournat, **S. Raetz**, H. Lu, V. E. Gusev, *Optically Controlled Nano-Transducers Based on Cleaved Superlattices for Monitoring Gigahertz Surface Acoustic Vibrations*, **ACS Nano 18(13)**, 9331-9343 (2024). [Q1] [https://doi.org/10.1021/acsnano.3c07576]

[A32] V. E. Gusev, T. Thréard, D. H. Hurley, **S. Raetz**, *Time-domain Brillouin scattering theory for probe light and acoustic beams propagating at an angle and acousto-optic interaction at material interfaces*, **Photoacoustics 33**, 100563 (2023). [Q1] [https://doi.org/10.1016/j.pacs.2023.100563]

[A31] S. Sandeep, **S. Raetz**, N. Chigarev, N. Pajusco, T. Thréard, M. Edely, A. Bulou, A. Zerr, V. E. Gusev, *Time-domain Brillouin scattering for evaluation of materials interface inclination: Application to photoacoustic imaging of crystal destruction upon non-hydrostatic compression*, **Photoacoustics 33**, 100547 (2023). [Q1] [https://doi.org/10.1016/j.pacs.2023.100563]

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[App1] S. Mezil, F. Bruno, **S. Raetz**, J. Laurent, D. Royer, and C. Prada. (2015) *Investigation of interfacial stiffnesses of a tri-layer using Zero-Group Velocity Lamb modes.* **arXiv cond-mat.mtrl-sci**, 1511.04367. [https://arxiv.org/abs/1511.04367] Cette prépublication est parue dans J. Acoust. Soc. Am. : https://doi.org/10.1121/1.4934958 [A5].

5.3.3 Communications at International Conferences

In the list of communications at international conferences presented below, I would like to clarify that all the conferences I participate in are selective conferences, some of which are highly selective, with the possibility to submit proceedings in almost all of them (those listed previously all offer this option). With the exception of communication [IC1], which was accompanied by an article published in a special edition dedicated to the LU2010 conference of the Journal of Physics (IOP), I have never submitted other conference proceedings for the main reason that some journals consider these articles as a prior publication of results, which could diminish the originality of the manuscript submitted, where they are typically preliminary or less developed results.

[IC112] N. Pajusco*, **S. Raetz**, N. Chigarev, **S. Sandeep**, M. Edely, A. Bulou, A. Zerr, V. Gusev, *4D time-domain Brillouin scattering of water ice phase transition under non-hydrostatic load*, **AFPAC 2025**, January 2025, Chédigny, France. (Oral)

[IC111] R. Delalande, S.M. Kukhtaruk, J. Wolfman, N. Chigarev, B. Negulescu, G. Liu, **S. Raetz***, V.E. Gusev, *Nanometric surface acoustic wave pulses generated and detected by laser*, **AFPAC 2025**, January 2025, Chédigny, France. (Oral)

[IC110] N. Pajusco, **S. Raetz***, N. Chigarev, S. Sandeep, M. Edely, A. Bulou, A. Zerr, V. Gusev, *4D time-domain Brillouin scattering imaging of the evolution of non-hydrostatically loaded water ice*, **LU 2024**, October 2024, Nanjing, China. (Oral)

[IC109] R. Hodé, G. Yan, J. Alcaraz, N. Chigarev, H. Li, J. Blondeau, N. Cuvillier, C. Prada, S. Mezil, V. Gusev, M. Ducousso, V. Tournat, **S. Raetz***, *Laser-ultrasonics-based zero-group-velocity (ZGV) Lamb modes for inspecting damaged and adhesively-bonded materials*, **LU 2024**, October 2024, Nanjing, China. (Oral, **invité**)

[IC108] V. Gusev*, T. Thréard, N. Pajusco, S. Sandeep, N. Chigarev, **S. Raetz**, *Emerging theoretical models in applications of picosecond laser ultrasonics for imaging inhomogeneous materials*, **LU 2024**, October 2024, Nanjing, China. (**Keynote**)

[IC107] N. Pajusco, **S. Raetz**, N. Chigarev, S. Sandeep, M. Edely, A. Bulou, A. Zerr*, V. E. Gusev, *3D-imaging of texturing of a polycrystalline ice VII sample upon non-hydrostatic squeezing in a DAC*, **61st EHPRG Meeting**, September 2024, Thessaloniki, Greece. (Poster)

[IC106] **S. Raetz***, *Probing and imaging complex structures from macro- to sub-microscale via laser ultrasonics*, **International Conference on Photoacoustic and Photothermal Phenomena (ICPPP22)**, July 2024, Coimbra, Portugal. (Oral, **invité**, **IPPA Junior Prize**)

[IC105] S. Sandeep, N. Pajusco, E. Nicol, N. Delorme, N. Chigarev, V. Tournat, N. Cuvillier, J. Delozanne, M. Ducousso, **S. Raetz***, V. E. Gusev, *Analysis of rough metal/epoxy interfaces using time-domain Brillouin scattering*, **International Conference on Photoacoustic and Photothermal Phenomena (ICPPP22)**, July 2024, Coimbra, Portugal. (Oral)

[IC104] N. Pajusco*, **S. Raetz**, N. Chigarev, S. Sandeep, M. Edely, A. Bulou, A. Zerr, V. E. Gusev, *4D imaging of water ice under high-pressure non-hydrostatic load by time-domain Brillouin scattering*, **International Conference on Photoacoustic and Photothermal Phenomena (ICPPP22)**, July 2024, Coimbra, Portugal. (Oral)

[IC103] N. Pajusco*, N. Chigarev, **S. Raetz**, V. E. Gusev, *Time-domain Brillouin scattering measurement in thin layer of water with non-parallel interfaces*, **International Conference on Photoacoustic and Photothermal Phenomena (ICPPP22)**, July 2024, Coimbra, Portugal. (Poster)

[IC102] R. Delalande, S. M. Kukhtaruk, J. Wolfman, N. Chigarev, B. Negulescu, G. Liu, **S. Raetz**, V. E. Gusev*, *All-optical monitoring of surface acoustic wave pulses of nanometers lengths*, **International Conference on Photoacoustic and Photothermal Phenomena (ICPPP22)**, July 2024, Coimbra, Portugal. (Oral)

[IC101] V. Gusev*, N. Chigarev, S. Sandeep, T. Thréard, A. Bulou, E. Nicol, N. Pajusco, V. Tournat, N. Cuvillier, D. Hurley, M. Ducousso, A. Zerr, **S. Raetz**, *Advances in Applications of Picosecond Acoustic Interferometry for Nanoscale Imaging*, **20th**

World Conference on Non-Destructive Testing (WCNDT), May 2024, Incheon, Korea. (**Keynote**)

[IC100] C. Li*, A. Lemaitre, N. Chigarev, M. Morassi, **S. Raetz**, A. Maznev, D. Lanzillotti-Kimura, V. E. Gusev, *Optical monitoring of sub-THz Rayleigh and THz surface skimming acoustic waves on cleaved superlattices*, **Ultrafast Phenomena and Nanophotonics XXVIII (PW24O SPIE OPTO)**, January 2024, San Francisco, United States of America. (Poster)

[IC99] S. Sandeep, **S. Raetz***, N. Chigarev, N. Pajusco, T. Thréard, M. Edely, A. Bulou, A. Zerr, V. E. Gusev, *Imaging of crystal degradation upon non-hydrostatic compression via time-domain Brillouin scattering*, **AFPAC 2024**, January 2024, Loch Lomond, United Kingdom. (Oral)

[IC98] **S. Raetz***, V. E. Gusev, N. Chigarev, S. Sathyan, *Time-domain Brillouin scattering: A tool for elastic property characterization, three-dimensional imaging, and real-time monitoring of transient processes*, **Optics + Ultrasound VI (O+U VI)**, October 2023, Nottingham, United Kingdom. (**Keynote**)

[IC97] **S. Raetz***, V. E. Gusev, N. Chigarev, S. Sathyan, *Recent advances on time-domain Brillouin scattering for three-dimensional imaging and transient process monitoring*, **Workshop on Progress in Photoacoustic & Photothermal Phenomena**, September 2023, Erice, Sicily. (Oral, **invité**)

[IC96] N. Pajusco*, **S. Raetz**, V. E. Gusev, *Numerical simulation of time-domain Brillouin scattering detection based on finite element and discontinuous Galerkin methods: case of inclined interface*, **Workshop on Progress in Photoacoustic & Photothermal Phenomena**, September 2023, Erice, Sicily. (Oral)

[IC95] N. Pajusco*, **S. Raetz**, N. Chigarev, V. E. Gusev, *Qualitative comparison between experimental and numerical simulations for time-domain Brillouin scattering: the case of an inclined interface*, **Workshop on Progress in Photoacoustic & Photothermal Phenomena**, September 2023, Erice, Sicily. (Poster)

[IC94] S. Sandeep, **S. Raetz***, E. Nicol, M. Edely, N. Pajusco, N. Chigarev, N. Cuvillier, J. Delozanne, M. Ducousso, V. Tournat, V. E. Gusev, *Time-domain Brillouin scattering imaging of mechanical metal/epoxy interfaces*, **ICU 2023**, September 2023, Beijing, China. (Oral)

[IC93] S. Sandeep, N. Pajusco, **S. Raetz***, N. Chigarev, T. Thréard, M. Edely, A. Bulou, A. Zerr, V. E. Gusev, *Time-domain Brillouin scattering for evaluation of materials interface inclination*, **ICU 2023**, September 2023, Beijing, China. (Oral)

[IC92] J. Alcaraz*, **S. Raetz**, S. Mezil, C. Prada, N. Chigarev, V. E. Gusev, *Edge waves monitored by laser ultrasonics for the detection of a crack*, **ICU 2023**, September 2023, Beijing, China. (Oral)

[IC91] C. Li*, A. Lemaitre, N. Chigarev, M. Morassi, **S. Raetz**, D. Lanzillotti-Kimura, V. E. Gusev, *Rayleigh and Bulk Surface Skimming Acoustic Waves Propagation on Superlattices*, **ICU 2023**, September 2023, Beijing, China. (Oral)

[IC90] V. E. Gusev*, **S. Raetz**, N. Chigarev, S. Sathyan, *Recent Advances in Applications of Picosecond Acoustic Interferometry for Imaging of Polycrystalline Materials and Material Modifications*, **ICU 2023**, September 2023, Beijing, China. (Keynote)

[IC89] Ch. Li, Ph. Djemia, N. Chigarev, S. Sodki, Y. Roussigné, **S. Raetz**, V. E. Gusev, A. Zerr*, *Elastic moduli, anisotropy and refractive index of γ-Ge_3N_4 via laser ultrasonics, Brillouin light scattering, and first-principles calculations* **The Joint 28th AIRAPT and 60th EHPRG International Conference 2023**, July 2023, Edinburgh, UK. (Poster)

[IC88] **S. Raetz***, S. Sandeep, N. Chigarev, N. Pajusco, T. Thréard, M. Edely, A. Bulou, A. Zerr, V. E. Gusev, *Time-domain Brillouin scattering for evaluation of materials interface inclination: Theory*, **XVII International conference on phonon scattering in condensed matter (Phonons 2023)**, July 2023, Paris, France. (Poster)

[IC87] **S. Raetz***, S. Sandeep, N. Chigarev, N. Pajusco, T. Thréard, M. Edely, A. Bulou, A. Zerr, V. E. Gusev, *Time-domain Brillouin scattering for evaluation of materials interface inclination: Application to imaging of crystal degradation upon non-hydrostatic compression*, **XVII International conference on phonon scattering in condensed matter (Phonons 2023)**, July 2023, Paris, France. (Oral)

[IC86] R. Delalande*, J. Wolfman, G. Liu, B. Negulescu, N. Chigarev, **S. Raetz**, V. Gusev, *Optical monitoring of surface acoustic pulse on nanostructured perovskites*, **XVII International conference on phonon scattering in condensed matter (Phonons 2023)**, July 2023, Paris, France. (Poster)

[IC85] M. Abi Ghanem*, M. Robin, L. Le Ridant, S. Dieudonné, O. Hamant, **S. Raetz**, T. Dehoux, *Plant-based MHz-GHz phononic materials*, **XVII International conference on phonon scattering in condensed matter (Phonons 2023)**, July 2023, Paris, France. (Poster)

[IC84] C. Li*, A. Lemaitre, N. Chigarev, M. Morassi, **S. Raetz**, D. Lanzillotti-Kimura, V. E. Gusev, *Sub-THz Rayleigh and bulk surface skimming acoustic modes on super-*

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SCIENCES
DE L'INGENIERIE
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Le Mans Université

Titre : Imagerie, évaluation et caractérisation non destructives de matériaux et structures complexes par ultrasons laser

Mot clés : Ultrasons laser, ondes élastiques guidées, acoustique picoseconde, diffusion Brillouin dans le domaine temporel, imagerie, évaluation et caractérisation non destructives.

Résumé : Ce manuscrit présente un aperçu de mes activités de recherche au Laboratoire d'Acoustique de l'Université du Mans (LAUM, UMR CNRS 6613) depuis 2014, axées sur le développement des techniques d'ultrasons laser. Mes travaux portent sur l'étude des modes de Lamb à vitesse de groupe nulle (ZGV) pour l'évaluation non destructive de la fatigue des matériaux et des collages adhésifs, ainsi que sur l'application de la diffusion Brillouin dans le domaine temporel (TDBS) pour la caractérisation des propriétés élastiques, l'imagerie tridimensionnelle et le suivi en temps réel, en particulier dans des environnements sous haute pression. Bien que la technique TDBS offre une résolution axiale supérieure à celle de la diffusion Brillouin dans le domaine fréquentiel, son adoption à plus large échelle nécessite le développement d'outils conviviaux pour traiter des problèmes de traitement du signal complexes. Parallèlement, mes recherches explorent les ondes acoustiques de surface (SAWs), avec des efforts en cours pour atteindre une génération et une détection efficaces de SAWs hautes fréquences. Ces activités visent à contribuer à établir les ultrasons laser comme une technique polyvalente et accessible pour l'imagerie et la caractérisation des matériaux.

Title: Imaging, non-destructive evaluation and characterization of complex materials and structures by laser ultrasound

Keywords: Laser ultrasonics, guided elastic waves, picosecond acoustics, time-domain Brillouin scattering, imaging, non-destructive evaluation and characterization.

Abstract: This manuscript provides an overview of my research activities at the Laboratoire d'Acoustique de l'Université du Mans (LAUM, UMR CNRS 6613) since 2014, focusing on the advancement of laser ultrasonics techniques. My work encompasses the study of zero-group velocity (ZGV) Lamb modes for non-destructive evaluation of material fatigue and adhesive bonding, as well as the application of time-domain Brillouin scattering (TDBS) for elastic property characterization, three-dimensional imaging, and real-time monitoring, particularly in high-pressure environments. While TDBS offers superior axial resolution compared to frequency-domain Brillouin light scattering, its wider adoption depends on the development of user-friendly tools to manage complex signal processing challenges. In parallel, my research investigates surface acoustic waves (SAWs), with ongoing efforts directed toward the efficient generation and detection of high-frequency SAWs. These activities aim to help establish laser ultrasonics as a versatile and accessible technique for imaging and material characterization.