

Needle-Based Ultrasonic Diagnostic Tool

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A thesis presented for the degree of
Master of Philosophy

Faculty of Engineering
The University of Nottingham

2018



The University of
Nottingham

UNITED KINGDOM • CHINA • MALAYSIA

Acknowledgements

I would like to express my special thanks of gratitude to Dr. Richard Smith for taking part in the supervision of my project and for his support, guidance, hours of one-to-one discussion and patience. I would like to acknowledge Prof. Matt Clark for offering me this opportunity to be involved in this project. I would also like to thank Dr. Serhiy Korposh, and Dr. Kevin Webb for their constant academic support in last couple of months towards the completion of my degree. Thank to Dr. Fernando Perez Cota for his advice that was essential in early part of my project. I would, in particular, like to mention Prof. Steve Morgan, who lent a helping hand during a difficult time.

In addition, I would like to take this opportunity to thank Dr. David Thomas, Matt Orton, Donna Astill-Shipman, and James Ross for providing me un-paralleled level of support and help during my hard time toward the completion of my degree. I would like to acknowledge the members of Photonic and Optic Groups of the Faculty of Engineering for sharing their time. Finally, I would like to thank my family and friends for their unremitting encouragement and caring throughout my study.

Achievements

I attended the following national and international conferences to present my work:

- IOP Anglo-French Physical Acoustic Conference AFPAC2016 London- 2016 (oral presentation)
- International Symposium on Laser-Ultrasonics and Advanced Sensing (LU2016) Lynz-2016 (oral presentation)
- Optical and UltraSound Imaging – OPUS France- 2016 (oral presentation)
- One day optics meeting sponsored by IOP- institute of Physics Biomimetic The University of Nottingham- 2016 (oral presentation)
- Lunchtime seminar talk in Photonics and Optics Group 2016 (oral presentation)

I achieved the following success during my study:

- £ 300 RECENDT award for the best paper from LU2016 conference (second place)
- £ 516.65 grant from Graduate School Travel Prize -The University of Nottingham
- First place postgraduate poster competition, Advanced Optics Group, University Of Nottingham (2016)

I completed the following courses during my study in the University Of Nottingham:

- Introduction to the literature review process for engineers
- Latex for researchers: an introduction
- Further latex for researchers: developing your skills in latex
- Introduction to Matlab for engineers
- Introduction to C programming for engineers

Abstract

Designing extremely small sized photoacoustic transducers can be very important in medical applications to perform *in vivo* imaging or to measure elastic properties of tissue that is hard to access externally, for example deep-seated tissue or tissue that is in very small and narrow cavities. The medical applications range from *in vivo* biopsy, in order to prevent of unnecessary biopsies, to the monitoring of cancer therapy, to investigation of the progression of disease *in vivo* as well as microsurgical guidance. Recently, a three-layered thin-film Fabry-Perot Interferometer (FPI) based structure has been used as a transducer to generate acoustic waves and launch them into cells under study. Brillouin oscillations in the cells were measured in the gigahertz (GHz) range which allowed high resolution imaging of living cells. The approach used in this study demonstrated the potential of providing the new insights in cell biology and it offers advantages over other mechanical imaging techniques in terms of speed, signal amplitude, and resolution. In this thesis, a method for extending this technique is described, where small FPI transducer (sub-mm) is fabricated on the tip of a single mode fibre to produce a wide range of mechanical frequencies with the focus on biomedical applications. The FPI comprises of an optical cavity or spacer sandwiched between two parallel partially reflecting mirrors. In the current study, a novel pump-probe technique is used for the generation and detection of acoustic waves via the same transducer, while two separate transducers are currently used in other studies; one for generation and one for detection of acoustic waves.

In this study, the Brillouin frequencies of the transducers are generated and measured in reflection using a single optical fibre. An opto-thermo-mechanically coupled model was used to predict the behaviour of the transducer when interacting with the pump and probe laser beams. This allows the optimisation of the design parameters for a particular operating frequency range. Different fabrication methods are used based on the utilised materials in the transducers. Sputter coater method is used for deposition of metallic materials such as gold or Indium Tin Oxide (ITO) where spin coating and dip coating methods are used for deposition of polymeric materials such as Polystyrene (PS) or Cellulose Acetate (CA). The aim is to produce acoustic waves in different frequency regions from hundreds of megahertz (MHz) to several GHz,

based on the applications. Several approaches are demonstrated to perform this either by using different materials, or/and increasing the spacer thickness.

In this study it was demonstrated that by increasing the thickness of Indium Tin Oxide (ITO) spacer in a FPI-based transducer fabricated on the tip of a single mode fibre from about 150nm to 560nm the frequency decreased from ~ 9.8 GHz to ~ 4.9 GHz, respectively. Also, by replacing material from ITO to Polystyrene (PS) in spacer as well as increasing its dimension the frequency was reduced from 550 MHz to 430 MHz for the 560nm and 931nm PS devices, respectively.

A tuneable absorbing nanoscale thin-film system was developed experimentally and numerically, comprising of an absorptive dye mixed in dielectric coatings on reflecting surfaces. In this part of study, it was demonstrated for the first time the manipulation of light absorption to tailor the transducer for an available wavelength for the pump laser pulse in order to generate acoustic waves. Specifically, the Cellulose Acetate thin films mixed with Aluminum Phthalocyanine Chloride (APC) absorptive dye with the maximum absorption at 748nm coated on an opaque gold substrate. It was demonstrated that the absorption behaviour of dielectric thin films can be flexibly tuned by adjusting the proper system parameters such as dye solution concentrations and the thickness of dielectric thin film layer coating. From this, to the best of my knowledge, it has been the first time that novel hybrid devices were proposed as the new configurations of FPI-based transducers for the generation and detection of acoustic waves. In the hybrid devices, the absorption can take place within a thin absorptive layer either before or after the metallic layers in the FPI structure for the generation of acoustic waves where the absorption normally takes place within the metallic layers.

The use of sputter assisted spin-coating fabrication method was proposed and demonstrated for the first time the possibility of the fabrication of a multi-layered structure using this method. In this method, different layers from the same dye-CA polymeric solution were deposited layer after layer by coating a protective layer such as ITO between each layer. This method was proposed in order to increase and control the thickness of the transducers more accurately. This method also offered more

surface smoothness, where a surface roughness of several nanometres can make significant difference in the optical and mechanical frequency responses of the transducers.

In addition, the used of sputter assisted dip-coating method was proposed for the fabrication of the transducers on the tip of optical fibres using the same procedure. Compared with the repeated dip-coating process used in single coating solutions, this method shows more advantages by virtue of the synergistic effect of the deposited layers. The multi-layered dip-coating differs from the layer-by-layer assembly where monomers from a solution are fabricated layer-by-layer.

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Chapter 1

Motivations

1.1 Introduction

In many circumstances in applications ranging from medical field to research, a very small inspection device is needed to observe or monitor deep-seated tissue or narrow spaces in the body that are hard to access externally. The use of different technologies such as spectroscopy or ultrasound detection makes it possible to produce extremely small transducers, smaller than a grain of salt, which can be mounted at the end of a needle. Ultrasound has become a key technique in numerous areas in medicine including diagnosis, treatment, and monitoring of disease progression as well as therapies [1]. Developing needle-based ultrasound will potentially find many areas of application, including *in vivo* biopsy, monitoring response to cancer therapy, microsurgical guidance, and small animal investigation.

Cancer is a serious medical condition that must ordinarily be diagnosed through the removal of a small piece of suspicious tissue via a biopsy, typically via a needle or minor operation. The sample is in turn sent to a pathologist for further examination. These procedures are invasive, costly and time consuming for both patients and healthcare specialists. A pathologist must fix, slice, stain, and examine the sample microscopically [5]. The simplest types of biopsies are minimally invasive, but depending on the location of the lesion accessing the suspected cancer can require significant or repeated invasive procedures. The development of small sensors to provide information in conjunction with the biopsy needle is being pursued to detect and evaluate the deep-seated soft tissue and prevent unnecessary further biopsies or invasive treatments [3].

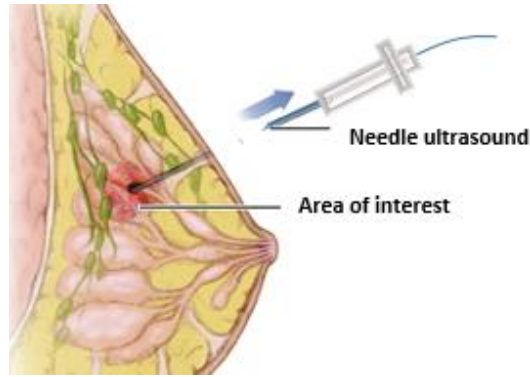


Figure 1: Inspection of cancerous tissue in breast cancer using needle-based ultrasound tool the figure has been adapted and modified from www.myhealth.alberta.ca [8].

For example, introducing a small-sized ultrasound transducer into a needle during breast cancer screening may allow the physician to make more informed decision about the location and malignancy of the samples (Figure 1). This approach could help to prevent unnecessary removal of a healthy tissue [6].

A needle based sensor may also be useful for the detection of changes in morphology and structure of deep-seated tissue to detect the cancerous tissues [7]. The collected information may be presented in real time. Experimentally, this may allow physicians, surgeons, and researchers to study and evaluate the progression of diseases *in vivo*. Scientists believe that the use of angiogenesis inhibitors, which block the development of new blood vessels, may provide a therapeutic effect. By "choking" the tumour of its nutrients and oxygen, its growth and spread to other body parts could be halted. There is still much work which remains to be done in this area, and one can immediately see the role of a needle-based ultrasound probe [8]. Another possible application is the localization of surgical guidance, such as the localization of the tip of epidural needle in epidural space where precision is the key factor for minimizing the risks of misguiding the needle [9].

Currently, various techniques such as endoscopic procedures are used to take biopsies from a target organ. Such procedures do have complications such as bleeding risk, infection, perforation, which may be fatal [11].

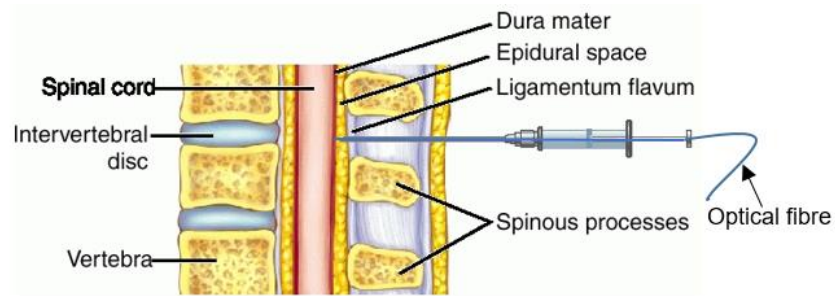


Figure 2: Localization of epidural space that can be embedded with high frequency ultrasound. The figure has been adapted and modified from www.medical-dictionary.thefreedictionary.com [10].

Implementation of a small-sized sensor into such endoscopic techniques would allow smaller instruments to be used, which would result in the procedure being more comfortable for the patients being less invasive and having less fatal complications. In addition to the above, due to the size and nature of such a new technique, it may allow certain group of patients who would currently not be considered as suitable for such procedures due to their co-morbidities and inability to tolerate current invasive procedures, to have such procedures carried out on them. This would fit perfectly into the current momentum of minimally invasive procedures [1].

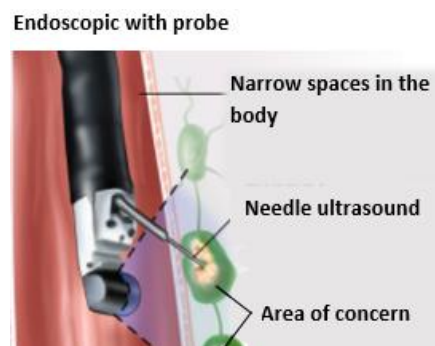


Figure 3: ultrasound-guided endoscopy to obtain sub-surface information in a narrow space in the body. The figure has been taken and modified from www.cancer.gov website [12].

The sensor also may allow the evaluation and study of the tissue in areas such as joints or abdominal cavities or access to target locations such as the epidural space. A needle-based ultrasound probe could be designed through which endoscopic fibres are passed, and to which forceps are attached. The identical access ports for

endoscopic biopsies could be used to access to the cavities for the needle based sensors (Figure 3) where more information can be obtained from the sub-surface of the region of interest. More information can be also gathered at the same time by the proposed endoscopic biopsy procedures. This may allow the physicians to make more accurate and safe decision about the location and malignancy of tissue samples. This may prevent the removal of healthy tissue by mistake or unnecessary further procedures [1].

The sensing element of needle ultrasound diagnostic tool requires very small-sized transducers as an alternative to broadband and larger size of piezoelectric transducers. The lateral dimensions must be on the sub-millimetre scale in order to work with acoustic waves at MHz frequencies. The fundamental advantage of this type of acoustic detection is its high sensitivity and the significantly smaller structure of the sensors in comparison with piezoelectric receivers [13].

1.2 Aim and goals

The aim of this project is to develop a multi-functional needle with diagnostic abilities. Development of this tool will be based on optical fibre technologies with the overall ambition of demonstrating applications in different biomedical and industrial settings. In the current work, it was proposed a technique using plate transducers which have been already designed for nano-scale live-cell imaging [49] in order to produce sub-mm ultrasonic probes for deployment in needles. This project deals with fabricating a small size high sensitivity FPI on the tip of a single mode optical fibre. The aim is to produce a tuneable transducers for a wide range of frequencies from several GHz to hundreds MHz for different applications. The transducer developed in this study operates in a manner similar to a FPI. This technique is used typically for a pure detection but here the device will be used for both the generation and detection of acoustic waves. The thickness of the layer and the choice of materials in the device are determined by the two optical wavelengths of our laser system. In this study, a pump-probe pico-second laser ultrasound system is used to generate and detect ultrasound in different frequency regions. The challenges for developing this early prototype for needle ultrasonic applications will be also discussed.

The aims of this study are to:

- Fabricate a micro scale transducer on the tip of a single mode fibre
- To generate and detect the acoustic wave via the same transducer
- Deliver pump and probe laser beams into the same single mode fibre
- Produce a wide range of frequencies from several GHz to hundreds of MHz for different applications

However, the direction of the study had to be changed due to difficulties in delivering of the pump (415 nm) and probe (780nm) laser beams through the same single mode fibre with 730nm cut off wavelength. With the initial laser system, the generation of acoustic waves at the excitation wavelength (415nm) and the detection of acoustic waves at the wavelength of the probe (780nm) using a single mode fibre was challenging due to high attenuation. The delivery of two laser beams from two different regions (i.e. visible and Near-Infrared) in a single mode waveguide caused a creation of grating structures in the pure fused silica core of a single mode fibre that is not typically suitable to transmit UV light. Gratings can be damaged by the absorption of UV light in the pure silica fibre core which occurs during the excitation of the transducer. Transmitting even $\sim 1\text{mW}$ pulsed laser beam at 415nm through the single mode fibre caused transmission degradation after just about one hour due to the absorption of UV light in the pure silica fibre core which occurred during the excitation of the transducer. These contribute to a high loss of power of the laser beams (attenuation). In addition, a modal dispersion limits the delay time between two pump and probe pulses [14].

Because of the outlined issues, and in order to bring the wavelength of pump and probe closer to each other, it was proposed to tailor the transducer for an available wavelength for the pump laser beam. For this, the absorption properties of the materials was tailored by adding an absorptive dye with a maximum absorption at a particular wavelength for the pump laser beam. Therefore, the generation and detection of the acoustic wave can be conducted via the same single mode optical fibre. A tuneable absorbing nanoscale thin-film system, comprising of dye molecules mixed in dielectric coatings on reflecting surfaces are developed experimentally and

numerically. In this part of the study, the manipulation of light absorption to tailor the transducer for an available wavelength for pump laser beam is demonstrated for the first time in order to generate acoustic waves. Specifically, Cellulose Acetate (CA) thin films mixed with absorptive Aluminium Phthalocyanine Chloride (APC) dye are used with a maximum absorption at 728nm coated on thin film gold substrate. It is also demonstrated that the absorption behaviours of dielectric thin films can be flexibly tuned by adjusting the system parameters such as dye solution concentrations and the thickness of dielectric thin film layer coating.

The aims of this part of the study are:

- Selection of a polymer and absorptive dye with maximum absorption at ~740nm
- Compatibility of the dye with the polymer
- Thickness analysis of poly thin films using different concentrations
- Effect of dye concentration on the properties of dye-polymer thin films
- Determination of thickness dependent optical constants of dye-polymer thin films
- Fabrication of higher order devices on the tip of single mode fibre with high uniformity
- Generation of acoustic waves via optical cavity

1.3 The structure of thesis

The motivations behind the study and research aims were described in the current Chapter, and the rest of this thesis will be presented as follows. A detailed background of the solutions for proposed problems and objectives will be discussed in Chapter 2. The detailed modelling of the proposed transducers as well as optimal design parameters are discussed in Chapter 3. In Chapter 4, the materials and method applied for the fabrication of the transducers and measurements are presented. Chapter 5 presents the results from the optical and mechanical measurements of the transducers where the models are validated. The results will be discussed in the same Chapter. The future development of the transducer using a new fabrication method

and hybrid devices are discussed in Chapter 6. Finally, a brief conclusion and applications of the proposed transducer of this work are drawn in Chapter 7.

Chapter 2

Background

2.1 High-frequency ultrasound

Sound waves with the frequencies from 20 KHz up to tens of GHz are defined as ultrasound [15]. Ultrasonic imaging is a tool that is considered as one of the most important diagnostic devices in the medical field and the associated clinical applications are still expanding. In 1955, Douglass Howry developed “brightness mode” or B-mode imaging instrumentation where the 2-dimensional B-scan image was formed by combination of A-scans (the intensity of reflected ultrasound signal as a function of depth) [16]. The operating frequencies of conventional ultrasonic imaging systems are between 2 MHz and 15 MHz where the frequencies above these ranges can provide images with high resolution qualities. For instance, high-frequency ultrasound with operating frequency above 20 MHz can provide high resolution ($<100\text{ }\mu\text{m}$) images in clinical and biomedical applications such as ophthalmology [17], intravascular imaging [18], and dermatology [19]. John Julian Wild and John Reid in collaboration with scientist from the National Cancer Institute in the United States extended the concept of sound navigation and ranging (SONAR) [20][21]. They applied ultrasound in MHz operating range to visualize human soft tissue.

In ultrasonic systems, the transducer is the key element for the generation and detection of acoustic waves. A simple piezoelectric transducer consists of an active element such as Piezoelectric Zirconate Titanate (PZT) sandwiched between two electrodes. Acoustic waves are generated by vibration of the transducer’s elements due to the application of a voltage pulse across the elements. The operating frequency of the transducer is a fundamental parameter for producing ultrasound images. The resolution of images increase as the operating frequency of the transducer is increased. However, the penetration depth is reduced as the frequency is increased [1].

The clinical applications of ultrasound are carefully chosen based on the operating frequency of the transducers. Medical applications such as cardiology, ophthalmology,

and peripheral vascular imaging [22] are performed between 1 MHz and 10 MHz, whereas the imaging of structures of ocular tissue [23] and skin [24] are performed between 10 MHz and 40 MHz with a resolution of $100\mu\text{m}$ [1]. Microscopic resolution can be achieved by increasing the frequency from 40MHz to 100 MHz with a penetration depth between 4-8mm and a resolution of $50\mu\text{m}$ [24]. In 1995, Pavlin and his group developed a new method of imaging using high frequency ultrasound to investigate anterior segment structures of glaucomatous eyes [25] and anterior segment tumours [26]. In the same time, another group studied muscular and elastic arteries *in vitro* using high frequency intravascular imaging system (i.e. 45 MHz) [22]. Figure 4 presents the needle probe used in their study.

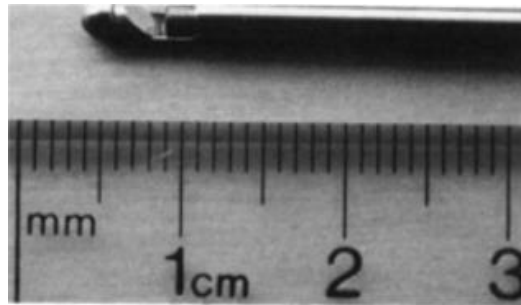


Figure 4: The photograph of needle probe (Lockwood et al., 1999 [22]) intravascular imaging system. The outer diameter of the needle is 2.8mm.

Higher frequency skin imaging can provide more information in regards to skin disorders such as psoriasis and eczema or basic studies about skin aging and the effects of irritants. A study was reported on skin imaging *in vivo* using a real-time ultrasound backscattered microscope (UBM) operating within the 40-100 MHz range [24]. Another group used the same ultrasound technique (i.e. UBM) at a frequency of 50 MHz to image immature articular cartilage [27]. In addition, cells and bacteria as biological specimens can be investigated using high frequency ultrasound which provides micron resolution images [28][29].

In 1995, a comprehensive study associated with high-frequency ultrasound was at conducted by Chandraratna [30]. They used transducers operating frequencies of 100, 200, 400, and 600 MHz. They investigated the lowest frequency required for cellular imaging of myocardium specimens. It has been reported that for visualization

of cellular details such as fibrosis, cell fallout and round cell infiltration in the myocardium could be identified with transducers operating at 600 to 200 MHz. At these frequencies the pathological changes can be detected, whereas cellular details could not be adequately identified with a transducer operating at 100 MHz [30].

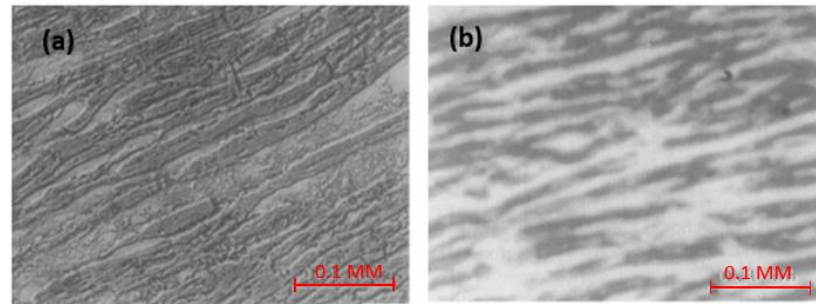


Figure 5: Acoustic microscopic image of normal myocardium obtained with (a) 600 MHz and (b) 200 MHz transducers. The images have been taken from Chandraratna et al., AM Heart journal, in 1995 [30].

As can be seen from Figure 5, the details of myocytes cells such as myofibrils can be visualized using a 600 MHz transducer Figure 5(a), where the cell details cannot be identified in the image obtained by a 200 MHz transducer Figure 5(b). They concluded that a transducer operating at the frequency of 200 MHz possibly has *in vivo* application using this technique. A similar study was conducted by another group using very high frequency ultrasound operating at 1000 MHz and 600 MHz using myocardial specimens with a dimension of 5 μ m [31]. They demonstrated that the myocardial cells can be assessed in these frequencies. However, they reported that these frequencies are not suitable for *in vivo* applications due to poor penetration [31].

Another study conducted by scientist from Ryerson University showed a high resolution ultrasound and photoacoustic images of stained neutrophils, lymphocytes and monocytes from blood smear [32]. In their study, a 1000 MHz ultrasound transducer was used to provide a micrometre resolution. Photoacoustic images were produced using a 532nm pulsed laser beam. They claimed that this technique has potential application in diagnosis of cell abnormalities.

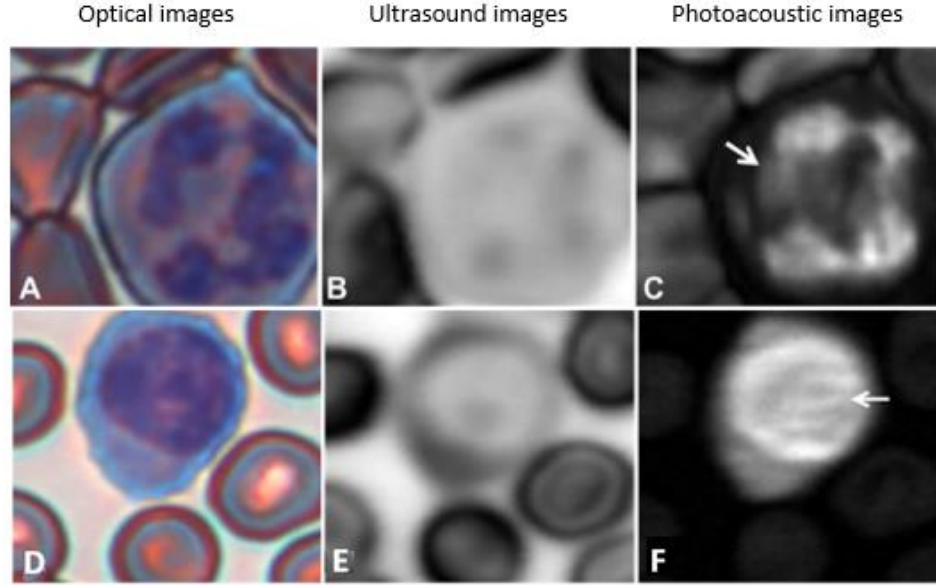


Figure 6: Optical, ultrasound and photoacoustic images (Strohma et al., 2016 [32]) of stained neutrophils (A, B, and C) and monocytes, (D, E, and F). The scale bar is 5 μm .

The Figure 6 shows optical, ultrasound and photoacoustic images of neutrophils (A, B, and C) and monocytes (D, E, and F). The fine structures are difficult to observe in the ultrasound and photoacoustic images. Two of the limitations in ultrasound imaging are a high attenuation at GHz range and poor penetration depth [32]. This study will focus on the fabrication of ultrasonic transducers with nanoscale dimensions to produce frequencies between hundreds of MHz ($>200\text{MHz}$) up to several GHz.

2.2 Needle-based Laser ultrasound

The concept of high frequency needle-based ultrasound was introduced by Eggleton and his group [2]. They published a patent by which a system composed of an ultrasound transducer positioned within a needle was described. The transducer directs its acoustical beams radially into the tissue and was designed to operate in the pulse echo mode [3]. In this mode, the ultrasound waves are transmitted into the tissue of interest by the transducer. The sound waves, which are reflected back from the tissue, are in turn detected by the same transducer. However, they never published any further details in this regards [4].

Laser ultrasound (LU) is a well-established technique to generate and detect ultrasound by using light as an alternative to piezoelectric transducers [33]. This technique offers several clear advantages such as couplant-less, wire-less operation, non-contact measurement and high sensitivity due to its small size (sub mm) of the transducer [49]. Optical generation and detection of acoustic waves was introduced in 1960s [34]. All-optical ultrasound transducers (AOUST) based on optical fibre are used as transmitting-receiving transducers due to their very small size. The miniaturization of these type of transducers facilitates insertion of the transducers in the region of interest in the human body where it is difficult to access.

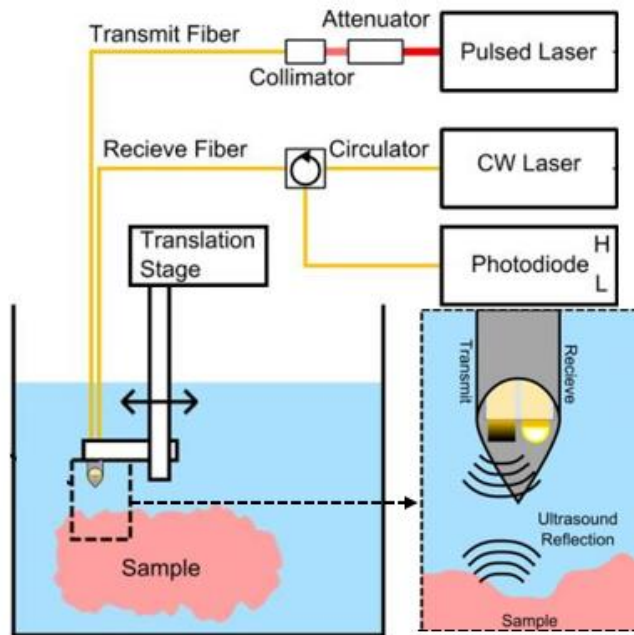


Figure 7: imaging system setup used by Paul Beard et al., (2015) [35] consisting of an optical fibre for laser generation of ultrasound and an adjacent fibre with a FPI fabricated at the tip for detection of ultrasound within tubular metal housing.

Colchester from University College London (UCL) developed an AOUST probe for vascular imaging [35]. In this study an LGUS transmitter and an FPI fabricated at the end of adjacent optical fibre as a receiver were located within tubular metal housing with inner diameter of <0.84 mm. An optical fibre with a core of silica glass (diameter: $200\mu\text{m}$) and ultrasound bandwidth of ~ 20 MHz was used to detect ultrasound. The Figure 7 shows the imaging setup in the mentioned study. The transmitter developed using carbon nanotubes polydimethylsiloxane (CNTs-PDMS) composite materials. This

group imaged *ex vivo* a vascular tissue from swine that corresponded to histology [36].

Laser ultrasound relies on the temperature rise produced by the absorption of a focused short light pulse on an opaque material to excite acoustic waves which have frequencies in the MHz region ($\geq 100\text{nm}$). The width of the optical pulse can be as short as tens of femtoseconds leading to the generation of ultrasound in the GHz range. Ultrashort laser pulses are an excellent and the only way to generate and detect acoustic waves with such high frequencies [37]. A detector based on Fabry-Perot Interferometer is one type of the optical detection of ultrasound (ODUS) technique which facilitates detection of broadband ultrasound from hundreds of MHz to several GHz [38][39]. Fabry-Perot fibre-optic ultrasonic hydrophones have been well studied by scientists from UCL. The hydrophones are mainly used for the detection of acoustic waves [39].

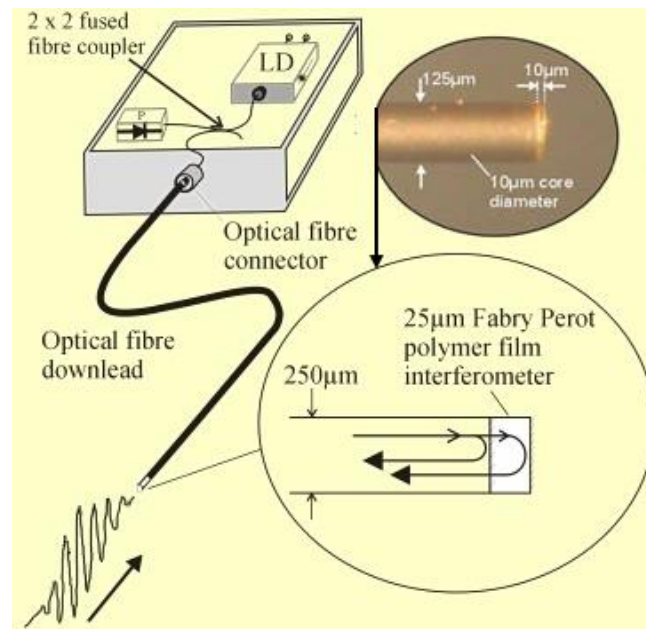


Figure 8: Schematic and photograph of the fibre optic hydrophone system used by Morris et al. [46].

In 1996, Paul Beard and Tim Mills developed a low-finesse FPI fabricated at the end of a multi-mode optical fibre. They claimed that the performance of the sensor is comparable with a piezoelectric hydrophone and it can be considered as an alternative

to piezoelectric devices for the detection and measurement of ultrasound [40]. They investigated the performance of the FPI as a miniaturized optical fibre ultrasonic hydrophone in several publications [41][42][43][44][45]. They investigated different parameters using FPI sensors such as temperature and acoustic pressure [46]. However, the FPI fibre-optic ultrasonic hydrophones are mainly used for characterizing ultrasound fields and providing up to 50 MHz acoustic bandwidth with 10 μ m element size [46].

As an incident acoustic wave strikes the surface of a FPI-based ultrasonic hydrophone, its thickness is modulated. This leads to the light reflection from its two sides which ultimately cause an optical phase modulation. The intensity of reflected light is then modulated and measured by a photodiode [46].

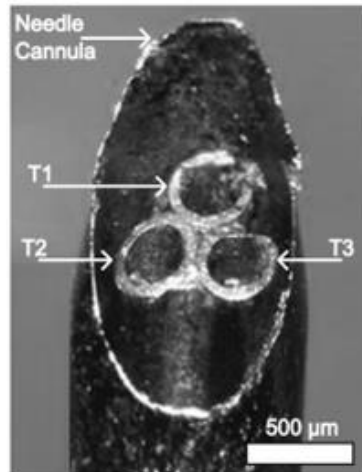


Figure 9: Needle probe (Colchester et al., [47]) comprise of three 24 gauge hypotubes: positioned in a 16 gauge injection needle: T1 is for photoacoustic sensing, T2 for receiving ultrasound, and T3 is for transmitting ultrasound for pulse-echo sensing.

The needle probe shown in Figure 9 has been developed by a group of scientists from UCL [47]. The needle comprised three separated optical fibres to generate and detect acoustic waves which have been accommodated within a 16-gauge injection needle. The distal end of an optical fibre is coated by an optically absorbing material (i.e. polydimethylsiloxane (PDMS)) for generation of acoustic waves and detection of ultrasound is performed by a single-mode fibre (T2) which has been fabricated with

a high-finesse FP cavity at the tip. A multi-mode fibre with core diameter of $200\mu\text{m}$ (T1) is positioned in the needle for photoacoustic sensing which was not used in this experiment. The needle probe developed in this study is a real-time needle guidance with photoacoustic and laser-generated ultrasound probes [47]. The acoustic waves are generated and detected by separated optical fibres within a needle. The bandwidth of the sensing ultrasound transducer was about 20 MHz. The B-mode image of human fingertips was provided by the needle.

2.3 Laser ultrasound as generator as well as detector

In the current study, the FPI method is used for both generation and detection of ultrasound via the same transducer. In this technique, the electrons are excited through irradiation of the transducer by a pump laser pulse (150fs) and their energy is transferred into lattice vibration during the diffusion of electrons. A thermal expansion and shrinkage occurs in thin film, leading to generation of ultrasonic plane waves which propagate in the direction of thin film thickness [48].

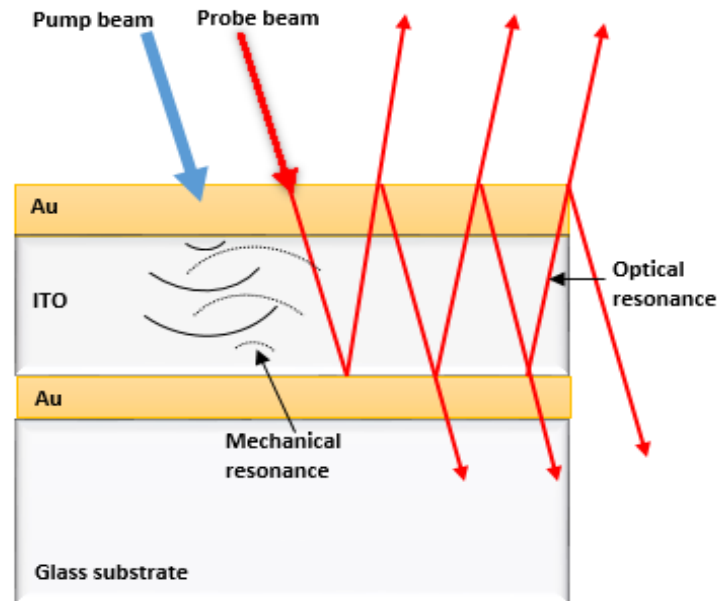


Figure 10: Metallic reflector layer-dielectric-Metallic reflector configuration of three layer FPI transducer. The transducer is excited with 415nm of pump pulse and 780nm of probe pulses which produce mechanical resonance and optical resonance, respectively.

The generated acoustic waves are reflected from two parallel mirrors in FPI and causing the generation of mechanical resonance as shown in Figure 10. The round trip time is too short (picosecond orders) and it is difficult to be measured by conventional methods. Light can propagate about 0.3mm within 1ps, while it allows only 5nm propagation distance for ultrasound [49]. Thus, using optical techniques, we can study the nanometre-order phenomena via millimetre-order measurements. This is the essence of the picosecond laser ultrasound technique.

In this study, the pump and probe technique is used to generate and detect acoustic waves in hundreds of MHz range and above and this method does not require the use of fast electronics. The pump pulse generates the coherent phonon pulses which propagate in the thickness direction and repeat reflections between the film surface and the film-substrate interface. Here another time-delayed short laser pulse is used to probe the fast events (see section 2.3.1). Laser ultrasound based on the pump-probe technique is commonly referred to as picosecond laser ultrasound (PLU) [50], where the detected signals are typically in the picosecond time scale. In this section, the mechanisms of generation and detection of ultrasound by laser light pulses will be discussed.

2.3.1 Optical generation of ultrasound

As previously mentioned, ultrasound can be generated optically by the absorption of light energy from the pump laser pulses in the first metal layer in FPI transducer. The absorption of the short pulse of light produces heat which causes the material to expand rapidly (Figure 11(a)) generating stress that launches a strain pulse (Figure 11(b)). The strain distribution and temperature rise induced by a laser pulse can be calculated through a series of equations [52][53] where light is absorbed by a material that is thick enough for generation and propagation of ultrasound until it is fully attenuated [49].

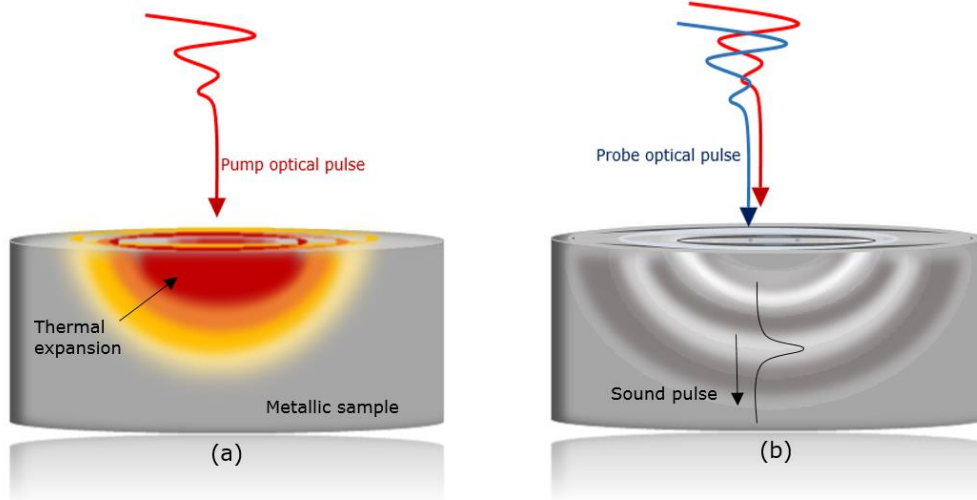


Figure 11: Thermo-elastic generation of acoustic pulse. Absorption of pump laser pulse in a sample increases temperature locally (a) which induces stress and strain in the sample (b). This causes vibration and launches acoustic wave due to thermal expansion. The second laser pulse (probe) is sent to probe reflected acoustic waves to the surface.

To generate ultrasound with femtosecond-laser pulses, the average power is kept low where the powers of peaks are high for generation of acoustic pulses. The shorter pulse induces generation of the more energy to be concentrated in high frequencies. The total energy per pulse can be determined by dividing the average power by the repetition rate (the number of pulses per second):

$$\epsilon_p = P_{avg}/R_{rate} \quad (1)$$

Where the resultant quantity of total energy (ϵ_p) is in Joules per pulse. The P_{avg} is the average power of the laser pulse in Watts and the R_{rate} is repetition rate in pulses per second. Then the peak power per pulse is calculated by the following equation:

$$P_{peak} = \epsilon_p/D_{pulse} \quad (2)$$

Where D_{pulse} is the pulse duration at the full-width-half-maximum (FWHM) point [51]. The maximum power of a pulse that can sustain for 12ns (pulse duration) was more important is considered in this study rather than power density which is the concentration of the power at that part. For example, a pulsed laser beam with

repetition rate of 80MHz, pulse duration of 150 femtosecond (fs) and average power of 1mW will produce pulses with energy of 12pJ, and a peak power of ~83.3W.

The laser pulses are absorbed in the first metallic thin film layer of FPI with duration of 150 fs. The absorption length or penetration (skin) depth (ξ) is calculated by the wavelength (λ) of the laser and extinction coefficient (κ) of the layer material. The following equation is expressed for calculation of the penetration depth of laser beam:

$$\xi = \frac{\lambda}{4\pi\kappa} \quad (3)$$

The absorbed light deposits some energy in the irradiated region at the z (thin film thickness) direction [51]. Then the total energy ($\epsilon_T(z)$) absorbed by the thin film can be calculated by the following equation:

$$\epsilon_T(z) = \frac{(1 - R)\epsilon_p e^{-\frac{z}{\xi}}}{a\xi} \quad (4)$$

Where R is total optical reflectivity which can be calculated by Fresnel Equations and the "a" is laser spot with a radius of r ($a=\pi r^2$). In general, the temperature increases ($\Delta T(z)$) and it can be calculated with using induced energy $\epsilon_T(z)$, and heat capacity "c" of the layer material as:

$$\Delta T(z) = \frac{\epsilon_T}{c} \quad (5)$$

The stress and strain can be calculated by the following equations where the temperature change is a function of time (t) and depth (z):

$$\frac{\delta\sigma_z}{\delta z} = \rho_0 \frac{\delta^2 \mu_z}{\delta t^2} \quad (6)$$

$$\delta\sigma_z = \rho_0 v^2 \eta_z - 3B\beta\Delta T(z) \quad (7)$$

$$\eta_z = \frac{\delta \mu_z}{\delta z} \quad (8)$$

Where the σ_z is the longitudinal stress, ρ_0 is the material density, μ_z is the displacement in z direction, η_z is the strain (deformation upon stress), v is the sound velocity, β the thermal expansion coefficient, and B is the bulk modulus. For an initial condition of $z = 0$ corresponding to the flat sample surface at $t=0$ and σz , the initial temperature rise and initial strain become:

$$\Delta T_0 = \frac{(1 - R)\epsilon_p}{a\xi C} \quad (9)$$

$$\eta_0 = \frac{3B\beta\Delta T_0}{\rho_0 v^2} \quad (10)$$

Once these equations were solved, the strain can be calculated by the following equation:

$$\eta(z, t) = \eta_0 e^{-z/\delta_p} - \frac{\eta_0}{2} \left[e^{-\frac{z+vt}{\delta_p}} + e^{\frac{|z-vt|}{\delta_p}} \text{sgn}(z - vt) \right] \quad (11)$$

2.3.2 Optical detection of ultrasound

Interferometry based technique is one of detection methods to detect laser-induced ultrasonic wave by an incident ultrashort optical laser pulse [54]. The interaction between the incident laser light and sound waves is an effective way to detect ultrasound while overcoming some limitations of piezo-electric transducers including limited bandwidth, the requirement for couplant and micro-scale fabrication. Small changes in overall reflectivity and transmittance can be measured by optimizing optical detection and signal processing by using the interferometric detection technique [82]. In this section we will discuss pulsed laser detection method used in LU.

2.3.2.1 Interferometric detection

Interferometry is the most common method for the detection of laser-induced acoustic waves [55]. In this study, the Fabry-Perot Interferometric (FPI) transducers were used for both generation and the detection of acoustic waves by using ultrafast laser systems. In interferometric detection using FPI, an optical cavity is formed in the space between the two reflectors. The thickness of optical cavity is modulated due to imposed stress (i.e. excitation by pump laser beam) to the transducer. The optical intensity differences between the two reflections occur after modulation of cavity thickness. The detection is achieved by interaction of laser light with the transducer to measure the corresponding intensity modulation. Another contribution to induce this modulation is to expose the surface of FPI to an incident acoustic wave which causes the displacement of an optical cavity [82][52]. The displacement induced by a strain can be calculated by the following equation:

$$\mu_z(z, t) = \int_0^{inf} \eta(z, t) \delta z \quad (12)$$

The phase of the optical beam is changed by this displacement at normal incidence. This can be calculated by the following equation by which the expected response can be quantified by a given strain:

$$\delta\varphi = \frac{4\pi}{\lambda} \mu_z \quad (13)$$

If the modulated beam is out of phase by 180 degree, the waves can be measured by converting the variations of phase to variations in amplitude [49]. Higher modulation depth ($\delta R/R$) is typically obtained by this method due to the alternation in the intensity produced by refractive index modulation, which is smaller in comparison to that produced by interference. These small changes in the intensity are commonly measured by using the FPI-based transducers [56].

2.3.2.2 Fabry-Perot Interferometer for detection of laser ultrasound

Laser-induced ultrasound generation is typically limited to metals which generate waves with particular characteristics [15]. The acoustic waves can be generated by the transducer when an adequate absorption occurs in the first metallic layer. The generated acoustic waves also can be detected using the transducers. Fabry-Perot Interferometer transducers are used in this study for the detection of ultrasound via optical cavity of the transducers. The FPI is made of two parallel mirrors around an optical cavity where interference occurs by transmitted light. Fabry-Perot produce multi waves interferences. Supposing that a monochromatic plane wave at the angle of incidence (θ_i) strikes upon a FPI. The waves were partially reflected and partially transmitted [57]. Those transmitted parts go under several round-trips inside the cavity before leaving it. An interference pattern with periodic dips (reflection) and peaks (transmission) is produced by the interference of multiple waves inside the cavity.

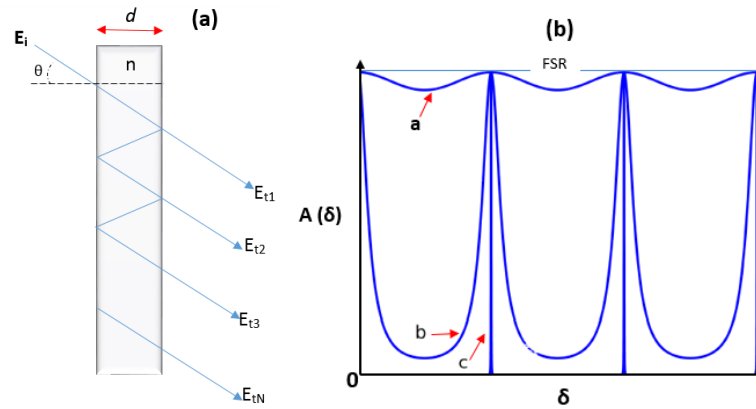


Figure 12: Multiple beam interference on the optical cavity of a FPI. a) Schematic of a simple cavity in which the beams follow the paths, and b) Transmission of a cavity. Sharpness of the peaks or finesses of the cavity depends on different parameters such as the reflectivity of the layers, losses within the cavity and coherence length of the light source. The images have been taken and modified from Perez Cota 2015 study [49].

The space between the peaks is called the free spectral range (FSR) for the given angle of incidence (Figure 12 (b)) [58]. The transmission of light inside the FPI depends on several parameters such as the wavelength (λ) of the light, the refractive index (n) of the material between the interfaces, the Fabry-Perot thickness (d), the

reflectivity coefficient of each interface (R) and the angle of incidence (θ_i). In the current study, all parameters are fixed, except the wavelength of the light as it travels from one medium to another medium with different refractive index where its speed also changes. When a phase-shift of $2m$ (m is an integer) occurs on a round trip, the maximum transmission will be achieved. Also the distance between two reflecting mirrors must be greater than the wavelength of the transmitted light. Since the reflected and transmitted light are bounced back and recombined in a sort of an optical feedback system, a dielectric film can be used as an optical cavity [58]. As previously mentioned the transmitted light inside the cavity goes under several round-trips before it leaves through partially reflecting mirrors. In this case, the resultant interference is equal to the multiple interfering waves (Figure 12(a)), with a phase shift given by the cavity characteristics, the beam coherence length and the wavelength. The intensity of the transmittance and reflectivity of the cavity can be calculated by solving the multiple interference between parallel layers (Figure 12(b)) [59][60]:

$$I_t = A(\delta) = 1 / 1 + F \sin^2 \left(\frac{\delta}{2} \right) \quad (14)$$

And

$$I_r = F \sin^2 \left(\frac{\delta}{2} \right) A(\delta) \quad (15)$$

Where $A(\delta)$ is known as the Airy function, δ is optical path differences that can be calculated by $2n d \cos \theta$. The optical path differences depends on thickness " d " and refractive index (n), and the propagation constant k_0 . The ratio between FSR and the linewidth (FWHM) of a mode determines the finesse of the Fabry-Perot. The width of the resonance peaks can be measured by the factor " F " which is defined as the finesse of an optical cavity. It is calculated by the following formula:

$$F = 4R / (1 - R)^2 \quad (16)$$

Where “ R ” is the reflectivity of the interfaces. The intensity of the transmittance of the cavity given by I_t is shown in Figure 12(b). As the reflectivity “ R ” of the interfaces increases, the transmission peaks become narrower which is also a useful effect. This is defined as high finesse. For example, in detection of ultrasound, if the wavelength of the probe laser beam is selected to be on the slope of one of the transmitted or reflected peaks, and then the intensity of the reflected and transmitted beams will be sensitive to the cavity size. The size of the cavity can be modulated due to the change in the distance between the layers once the acoustic waves are travelling through the cavity. In this case, the cavity size is comparable with an acoustic wave. The light will be modulated through this process. The sensitivity to the acoustic waves will be increased by the interference produced inside the cavity using this method for detection of ultrasound (see section 2.3.3) [15][82].

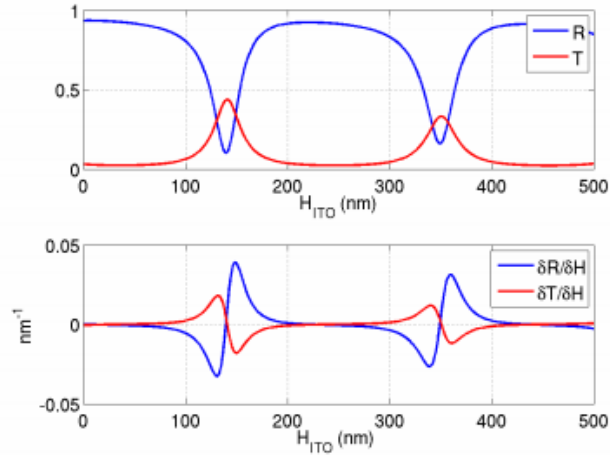


Figure 13: Optical behaviour of a FPI transducer against ITO dielectric thickness (H_{ITO}) where the gold layer is fixed at 30nm. Top graph represents reflectivity (R) and transmittance (T) and the bottom shows sensitivity to the change of the distance between metal layers. If the transducer is compress by a sound wave, the instantaneous reflectivity and transmittance will change depending on the sensitivity. The figure has been adapted from Perez Cota study [49].

The change in distance between two reflectors results in reflection (dip) and transmission (peak) change as shown in Figure 13. Those areas with the highest rate of change in transmittance/reflectance for a small change in distance between the reflectors are of the greatest interest. Therefore a transducer with an accurate dimension offers high sensitivity to detect incoming acoustic waves as a change in

size of the optical cavity. This sensitivity defines the ratio of the change in the reflectivity or transmittance within the transducer with respect to the change in the length of optical cavity (H) with the expressions $\delta R/\delta H$ and $\delta T/\delta H$, respectively. In the current study ideally the detection mechanism is performed in reflection. For that reason, the reflection should be as large as possible to achieve large detection levels. The high sensitivity can be achieved at slopes of reflection or transmission peaks as it can be seen the bottom graph of Figure 13. By using the mechanical properties of the transducer, the frequencies and amplitudes of acoustic waves generated by the transducer can be defined by the response of the transducers to a pulse of light excitation.

2.3.2.3 Fresnel equations and optical model

In this section, the transmissivity, reflectivity, and absorptivity as optical characteristics of the transducers are determined for both the pump and probe laser beams. The Fresnel coefficients are used in an optical model of the transducer in order to determine those optical characteristics. The angles of the reflected and transmitted beams propagating through a refractive index discontinuity are calculated using Snell's law (equation 17).

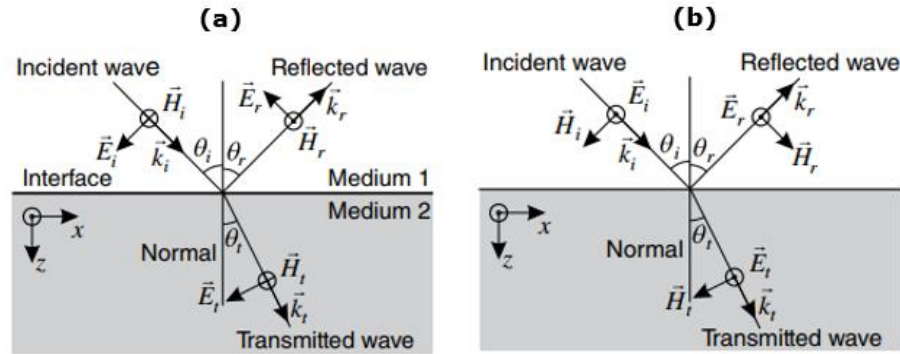


Figure 14: Reflection and transmission of an electromagnetic wave at an interface of materials with different optical characteristics. When (a) the electric field (E_i) is perpendicular (s-polarized) or (b) parallel (p-polarized). The figure has been taken from Thomsen et al., 1986 [54].

The Fresnel equations allows us to calculate the amplitude of the transmitted and reflected beams, and its relationship with the polarisation of the light. This approach

allows us to model numerically for those complicated multilayer systems such as the thin-film transducers [54].

$$n_i \sin \theta_i = n_t \sin \theta_t \quad (17)$$

The n_i , n_t , θ_i and θ_t are the refractive indexes of the incident and transmitted media, and their angles, respectively. Let us assume an electromagnetic beam incident to a highly transparent medium, and that the reflected and transmitted beams are all plane waves. There are two cases where the equation can be solved, when the electric field (E_i) is perpendicular (s-polarization) or parallel (p-polarization) to the plane of incidence (Figure 14). The ratio of amplitude of the reflected wave with respect to the incident for s-polarized light can be calculated by incorporating Snell's law:

$$r_s = \frac{n_i \cos \theta_i - n_t \cos \theta_t}{n_i \cos \theta_i + n_t \cos \theta_t} \quad (18)$$

$$t_s = \frac{2n_i \cos \theta_i}{n_i \cos \theta_i + n_t \cos \theta_t} \quad (19)$$

In the case of the electric field where it is parallel to the incidence plane (p-polarization), the reflection coefficient and transmission coefficient can be calculated by the following equations:

$$r_p = \frac{n_t \cos \theta_i - n_i \cos \theta_t}{n_i \cos \theta_t + n_t \cos \theta_i} \quad (20)$$

$$t_p = \frac{2n_i \cos \theta_i}{n_i \cos \theta_t + n_t \cos \theta_i} \quad (21)$$

The reflectivity (R) can be simplified as $R=|r|^2$ when $\theta=90$. If no light is absorbed in the material, transmitted power can then be calculated by the following equation:

$$T = 1 - R \quad (22)$$

The above equations (18, 19, 20, and 21) are known as the Fresnel equations and they give the relationship of amplitude of a wave depending on its incident angle and refractive index for parallel(p) or perpendicular(s) polarizations.

The optical model in this study has been developed based on the Fresnel equations due to the simplicity of the geometry of our three-layer thin-film transducer, which makes it possible produce the model in one dimension. This is because, the width of the transducer is greater than the optical wavelength [61]. The optical model consist of two main parts (Figure 15); the parameters such as thickness, refractive index, incidence angle can be changed in the main part (Figure 15(b)) and a second part that solves the transmittance and reflection for such parameters using the Fresnel equations (Figure 15(a)). The sensitivity of the transducers at λ_{probe} is calculate using an optical model for the transmission, reflection, and absorption cases. In the main algorithm, transmittance T, and reflection R are calculated for a range of thickness by using refractive indices of materials across various wavelength ranges. At this point from the second part of the model, the incidence angle, optical path, and Fresnel coefficient are calculated and then are fed into the main part for calculation of sensitivity for transmission ($\delta T / \delta H$) and reflection ($\delta R / \delta H$). At the end the results are plotted for the display matter. For this model the transducer is composed of multiple layers including air, material1, material2, material3, material4 and air again [49].

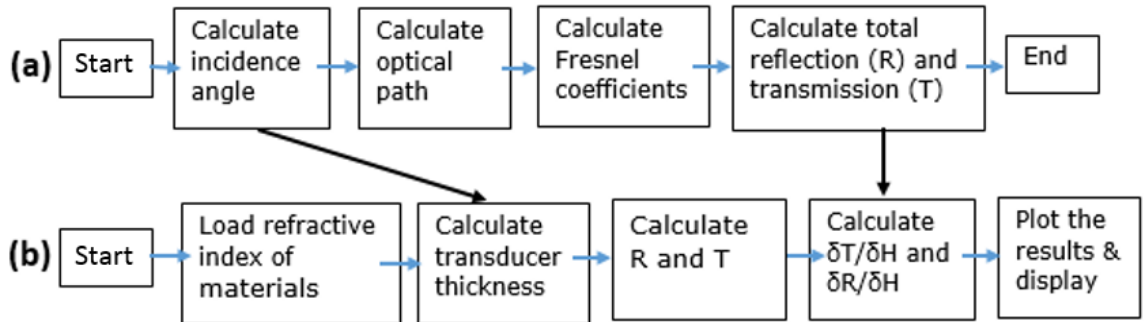


Figure 15: Flow diagram of two main parts of the optical model: (a) the T, R, and angle of incidence are calculated for a particular thickness. The results also are fed to (b) the main algorithm to calculate R and T for a range of thickness.

2.3.3 Pump and probe technique using picosecond ultrasound

The acoustic wavelength and optical wavelength are comparable when the acoustic waves are in the GHz range since the speed of sound is significantly slower than the speed of light. The high attenuation of sound in GHz range frequencies limits the generation of acoustic waves using conventional piezoelectric transducers. The pump and probe technique is used to overcome those limitations without use of special detectors or electronics [80]. This is because this technique offers a means after data acquisition to detect very high frequency acoustic waves [81]. The detection method presented in this study is based on Brillouin scattering (see section 2.2) [82]. This technique is used for both generation and detection of acoustic waves. This is feasible by separating the pulses in pump and probe parts with a time delay. In the context of generation and detection of ultrasound, the pump is used to generate the ultrasound where the probe laser is used to measure it. For separating the pump and the probe pulses, a controlled time delay is applied between two pulses [62]. This technique allows to sample the event in time by sequentially repeating the event and adjusting the delay. In every delay position, the trace of each event can be built from measurements. This approach does not require high-speed acquisition of photodiodes or detectors (ns Rise Time) (see section 2.4).

2.4 Brillouin scattering and oscillation

The acoustic waves are generated after the pump laser pulse incident which propagates in the film thickness direction. Scattered sound from acoustic waves in translucent solid is called Brillouin scattering [49]. Then the transducer is irradiated with a time-delayed probe laser beam in the same location as the pump. The probe laser beam then penetrates into the substrate, metallic layer and the cavity of the FPI. The penetrated light pulse is scattered by the sound wave and becomes matched when the Bragg condition is satisfied [48]:

$$\lambda_B = 2n\Lambda\cos\theta \quad (23)$$

Where "n" is the refractive index of the media, Λ the period of the grating and θ the incident angle (see Figure 16). A grating is generated by an acoustic wave due to the thermo-elastic effect in the strain regions. The reflectivity of the probe light inside the cavity shows oscillations corresponding to the frequency of acoustic waves propagating in the thickness direction.

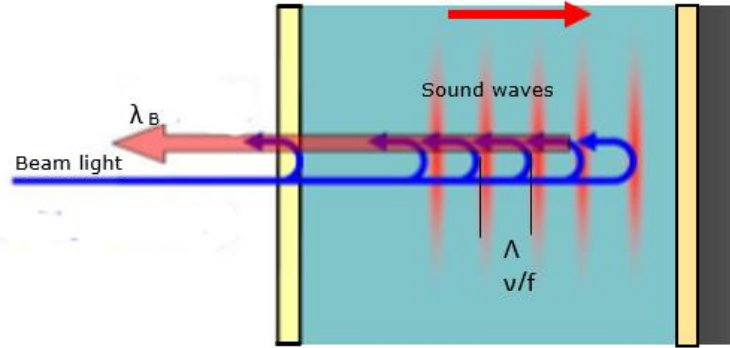


Figure 16: Bragg grating made out of a soundwave using probe light. The period grating depends on the acoustic source (frequency) and material (speed of sound). The incident light ($\theta = 0$) that satisfies the Bragg condition it is reflected by the grating. The figure has been adopted and modified from [48].

The scattered light beam experiences a frequency shift due to the movement of scattering element. The scattered light interferes with a reference beam or probe beam which results in a modulation of intensity (Figure 16). This is commonly known as Brillouin oscillations. The period Λ is given by a sound wave it can be written in terms of the speed of sound v and the acoustic frequency f as $\Lambda = v/f$ as:

$$\lambda_B = \frac{2nv}{f} \cos\theta \quad (24)$$

In order to scatter a light with specific wavelength λ in a translucent material, the frequency of an acoustic wave is needed which can be calculated by the following equation:

$$f_B = \frac{2nv}{\lambda} \cos\theta \quad (25)$$

Where “ n ” is the refractive index of the material, “ v ” is the speed of sound in a sample material, θ is the incident angel and λ the optical wavelength [62]. Brillouin oscillations can be observed when the scattered wave interferes with a reference beam. The frequency of this modulation is referred to as the Brillouin frequency (f_B) [49]. For example, the frequency of acoustic waves in water and glass are approximately ~5 and 22GHz respectively (at $\lambda=780\text{nm}$). In solids sound can propagate longitudinally (compression wave) but transversally as well. If the lateral area of the medium where the sound propagation is large in comparison with the acoustic wavelength, then only transverse waves are considered in a one dimensional case and the sound speed is expressed as:

$$v = \frac{\sqrt{E}}{\sqrt{\rho}} \quad (26)$$

$$f_B = \frac{2n\sqrt{E}}{\lambda\sqrt{\rho}} \cos\theta \quad (27)$$

Where E is the Young’s modulus (a measure of the elasticity of a material) and ρ is the density. The waves, which fit into a resonator of a particular length are also called oscillating modes. The Brillouin frequency is changed at a boundary and the time of flight (Δt) defines the time in which the Brillouin frequency stays constant before changing at the boundary. The speed of sound also can be calculated if the time of flight is known:

$$v = \frac{d}{\Delta t} \quad (28)$$

Where the “ d ” is the thickness of the medium and Δt also known as the time of flight. Then the f_B becomes;

$$f_B = \frac{2n\Delta t}{\lambda d} \cos\theta \quad (29)$$

Therefore, the frequency will be decreased either by lowering Young’s modulus of the material or by increasing the thickness of the cavity. The strain pulses are created by

the pump pulse incident into a metal film which propagates perpendicularly to the plane of the metal layer and parallel to the optical axis (Figure 17).

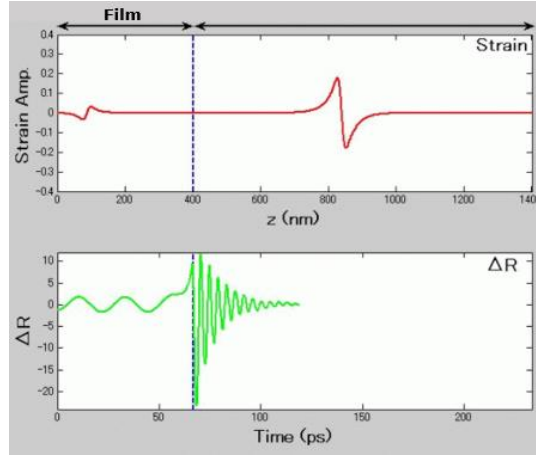


Figure 17: The simulation result shows the low-frequency and high-frequency Brillouin oscillations from the silica film and Si substrate, respectively. The top figure presents the propagated strain in the thin film and the substrate, and the bottom figure presents the change in reflection in the thin film and the substrate. The figure is taken and modified from the study conducted by Ogi et al. [48].

A part of probe pulse is scattered from the generated acoustic field in the time of its arrival and the other part is reflected in the transducer. The reflected part interferes with the scattered light to observe Brillouin oscillations by which the Brillouin frequency can be resolved in time [48][49]. The Brillouin frequency depends directly on the speed of sound, refractive index, and density of the sample materials. These parameters are used in theoretical models. The Figure 17 shows a theoretical simulation of the reflectivity change caused by the sound strain pulse.

2.5 Optical Fibre

As mentioned earlier in Chapter 1, one of the aims of this study is to fabricate a small-sized transducer on the tip of a single-mode fibre in order to generate and to detect the acoustic waves. Ideally the generation of acoustic waves and detection of Brillouin oscillations is performed in reflection configuration which means they can be performed from one side. Therefore, the fibre-based transducers can be deployed into

a needle or a catheter for detection of Brillouin oscillation from deep-seated biological tissue (see section 1.1). In this case the Brillouin oscillations are measured from a sample where it is in the vicinity with the transducer fabricated at the fibre tip (Figure 18).

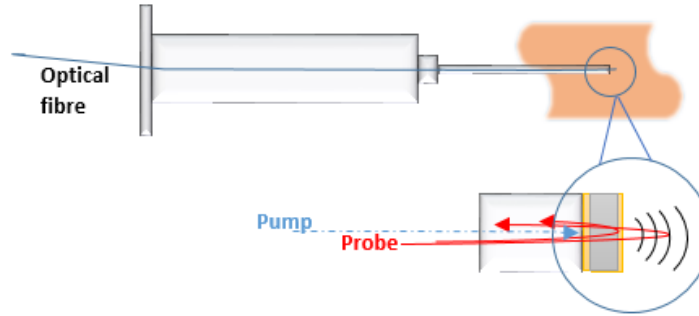


Figure 18: Fibre-based transducer, deployed into a needle to detect Brillouin oscillation from deep inside a biological tissue.

An optical fibre as a cylinder-shaped dielectric waveguide is comprised of a central core embedded in an outer cladding. The waveguide is made of silica glass as the low-loss materials where the outer cladding has slightly lower refractive index than the core [67]. The light is guided in the core of optical fibre and propagates based on total internal reflection at the core-cladding boundary. The principle of light transmission in the optical fibres is essentially same as transmission of light in planar dielectric waveguides [68].

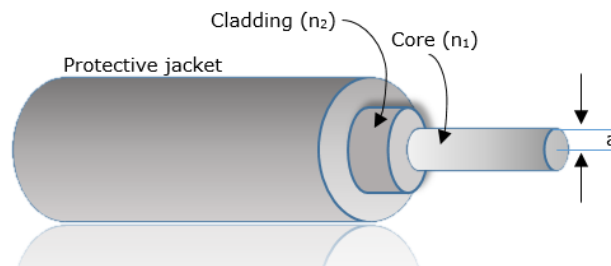


Figure 19: Schematic of single optical fiber structure.

There are two classes of optical fibres, single mode fibre (SMF) and multi-mode fibre (MMF). The SMF with smaller core diameter permits transmission of only a single mode and the MMF with larger diameter provides to transmit a wide range of modes.

In optics and photonics, modes describe the propagation of light and the number of photons in different modes what in turn describe the transport of energy or information [69]. The spatial distribution of optical energy in one or more dimension is called modes of light and its spatial distribution remains constant during propagation [70]. There are three types of modes: guided mode (a) (b), and substrate radiation mode (c) [71]. It is called guided mode because the light power is confined within the guided walls where the substrate radiation mode the light is confined at the air interface but varying at the substrate sinusoidally [71]. The light propagates in a waveguide as an electromagnetic field under certain boundary conditions where interfaces between planes have different refractive index [72].

2.5.1 Step index optical fibres

Different modes may have different group and phase velocities. This leads to some difficulties associated with propagation of light in the MMF (Figure 20). Since the modes have different velocity, their travel time also is different. So, as the light pulse travels through the fibre it is broaden which is known as modal dispersion. This phenomena limits the speed at which adjacent pulse can be sent without overlapping. The graded index fibre is a possible solution to reduce this effect. In this type of fibres the refractive index of the core cladding boundary is increased from a minimum value to a maximum value at its centre. In step-index fibres as a conventional fibres the refractive index of core and cladding are constant. Due to distinct structural feature of step-index fibres the light is exclusively guided in the core. This type of fibre is a frequently type of fibre that is used in different industries [68].

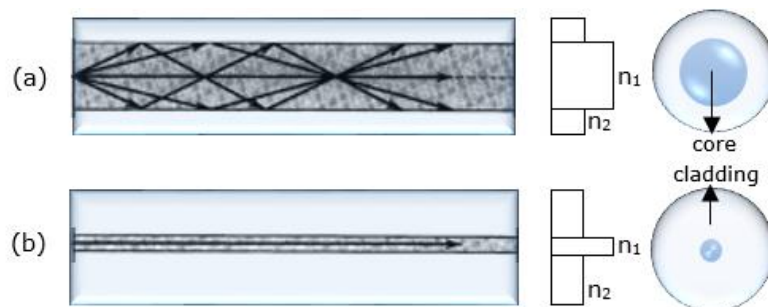


Figure 20: Geometry, refractive index and traveling light in: (a) a multimode step-index fiber and (b) a single-mode step-index fibre.

2.6 Manipulation of light absorption in dye-doped thin films

One of the important applications in Nano-photonics is manipulating light absorption in a nanoscale structure in order to create a specific absorption characteristic which is not provided by conventional materials. Various nanostructures can be fabricated using different techniques to enhance the light harvesting efficiency, such as strengthening localized resonances [76][77].

A new type of ultrathin-film system with the broadband light absorption properties based on interference effects has been reported recently [78][79]. This system consists of one or more nanoscale absorbing dielectric coatings that can be accommodated on a metallic back reflector. The thickness of such optical coatings are much thinner (i.e. a few nanometres) than conventional anti-reflecting coatings (i.e. $\lambda/4n$). The " λ " is the wavelength and " n " is the refractive index of the material. Note, in this study the absorption at the wavelengths between 720nm and 740nm were the greatest of interest. They can also retain the constructive interference of light waves due to their ultra-thin thickness. This ultimately results in localisation and absorption of incident light inside the absorbing dielectric. Highly dense absorbing media can be used for fabricating and designing such ultrathin dielectric coatings [78]. In this context, organic polymer films dyed with absorbing dye molecules coated on reflecting substrates can be a good option [79]. This is because the solution based fabrication of small-sized coatings offers high degrees of freedom to choose the dopants, hosts and the doping concentrations.

Chapter 3

Characterization of the thin film transducers

3.1 The selection of wavelength for probe (780nm) and pump (415nm)

The novelty of my study is to generate and detect acoustic waves using the same transducer. The probe beam (780nm) is used to detect the mechanical wave generated by the transducer by measuring variations in the beam intensity at the detector. The amplitude of the generated waves determines the power of single versus a level of noise power seen by the probe, the sensitivity of the device as well as the amount of light detected. In this section, we will concentrate on the sensitivity of the transducer. Figure 21 shows the example of the optical model for a wide range of gold (5-70nm) and ITO (100-200nm) thicknesses to optimise the sensitivity of the transducers before fabrication. There, the sensitivity for the reflected ($\delta R/\delta H$) and transmitted ($\delta T/\delta H$) beams are presented. The transmission and reflection are tuned based on a large range of possible thicknesses and different materials. In this study, the reflector layers and the optical cavity in a FPI transducer are made in different thicknesses and materials. Therefore, the sensitivity in the transmission and reflection are different for the FPI-based transducers in this study.

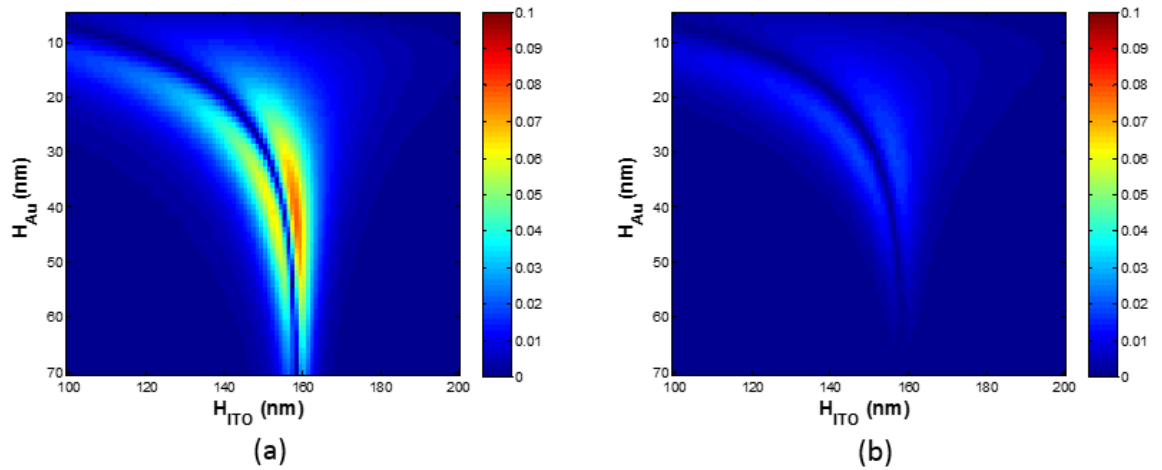


Figure 21: Transducer sensitivity in reflection $\delta R/\delta H$ (a) and in transmission $\delta T/\delta H$ (b) at λ_{probe} for a range of layer thicknesses. The colour bar shows the sensitivity (nm^{-1}).

As it can be seen in Figure 21(a), the sensitivity for the reflection case reaches a maximum of $0.5 \text{ (nm}^{-1}\text{)}$ while for the transmission case (Figure 21(b)) is about half of this value. This means that the reflection case is twice as good for detecting incoming waves.

The sound-waves are created by the thermoelastic effect via pump beam episode on to the sample. A rapid thermal expansion occurs when the optical energy of a pulse is rapidly absorbed by the transducer. Part of the energy is transmitted to the substrate and the rest resonates inside the generation transducer as the acoustic energy propagates [49]. The optical absorption coefficient of gold at 415nm is high ($\alpha=59.5 \times 10^6 \text{ cm}^{-1}$) [65] and approximately six times bigger than at 780nm, leading to a short skin depth ($\delta=36\text{nm}$). In the case of 415nm of pump laser beam, the skin depth is comparable to thickness of the layers in the generation transducer and this is a good feature from a design point of view. Because the skin depth is comparable to the dimension of the generation transducer, it will generate waves efficiently. That flexibility allows the tuning of the layers for high transmission and low sensitivity of λ_{probe} without compromising acoustic generation.

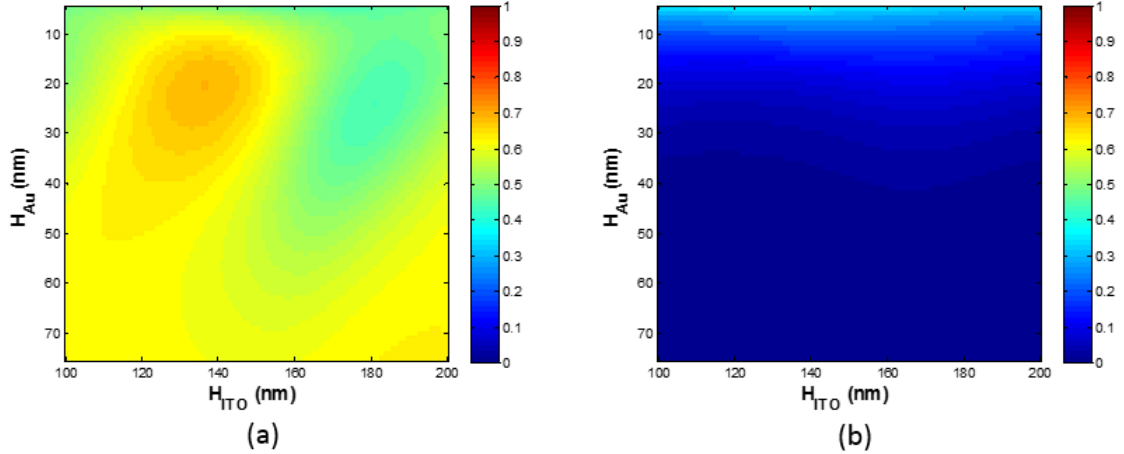


Figure 22: Result of optical model for pump wavelength. Colour bar shows percentage of light absorbed per nm by the transducer (a) and its related transmittance (b) at λ_{pump} in a range of thicknesses.

Figure 22 is an example of the stimulated optical model for λ_{pump} . As shown in Figure 22(a), the absorption remains almost similar in all layers with variations of less than 20%. From Figure 22(b) the transmission drops rapidly with gold thickness at 415nm of pump laser beam suggesting that little improvement to the exposure reduction can be achieved above 10nm of gold. However, thicker gold layers can make the device resonate at lower frequencies because the round trip time of the acoustic wave inside the generation transducer is increased. In practice, regardless of the layers' dimensions in the generation transducer, as long as they are comparable to the skin depth, the transducer can generate acoustic waves efficiently. That versatility allows the tuning of the layers for high transmission at λ_{probe} without compromising the generation of acoustic waves.

3.2 Layer material

First, the transmittance at λ_{probe} (780nm) for detection transducers was modelled for 150nm of ITO layer with 30nm of different reflectors including gold (Au), Silver (Ag), Copper (Cu), and Aluminium (Al). Figure 23 shows examples of these models. From there it is possible to see that Al shows low transmittance which makes it unsuitable for detection in reflection where the Au, Cu and Ag show similar capabilities with maximum transmittance in about same range.

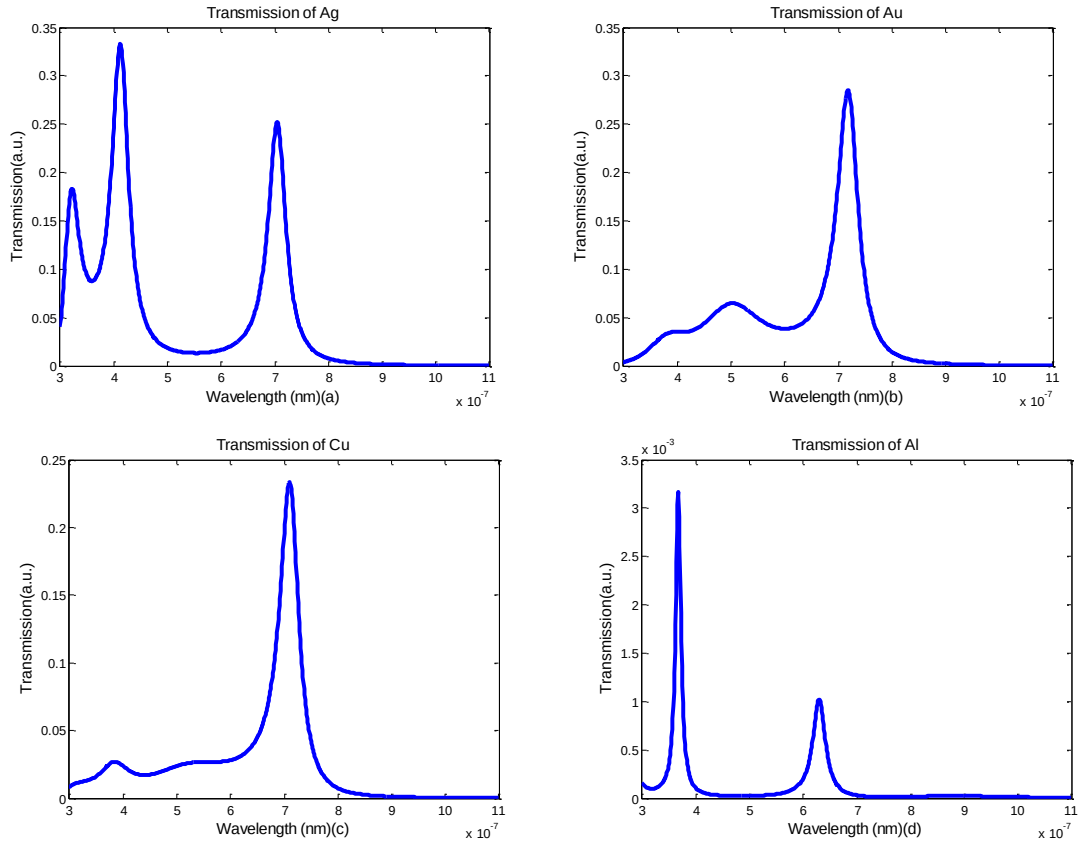


Figure 23: Comparison of the performance of detection transducers working in transmission build from silver (a), gold (b), Copper (c), and Aluminium (d) metals at λ_{probe} (780nm).

Absorption was also calculated for generation transducers ($\lambda_{\text{pump}}=415\text{nm}$) for the similar dimension and the result is shown in Figure 24. As it can be seen the absorptions for gold and copper are similar, however for aluminium and silver

absorption is ~ 4 times smaller meaning that it will generate less significant acoustic waves. That leaves only gold and copper (Figure 24).

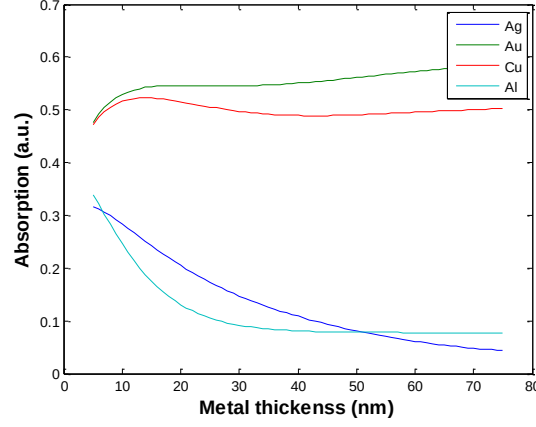


Figure 24: Comparison of the performance of generation transducers working in transmission build from different metals at λ_{pump} (415nm).

3.3 The principle of mechanical characterization of generation transducers

From the mechanical response of the transducer point of view, the acoustic waves are generated by the simple three layer FPI transducers. It resonates once it is excited by a pump laser pulse and the generated acoustic waves will be radiated if the transducers are in contact with another medium. It is important to know their resonance frequency when the transducer works as an acoustic source generator. The signals from generation transducer can be detected by independently fabricated detection transducers. The mechanical model in this study was developed using Matlab and signal processing tool box. In order to simulate the generation transducers, it was simplified to a single dimension (z , thermo-mechanical). The model takes the laser input as an injection of energy (Equation 4) which is associated with the laser power, pulse width and absorption of the layers in the generation transducer. Then thermal expansion is computed by the model. When the temperature is increased, the motion takes place and the laser light is absorbed. This model is thermally-mechanically coupled model.

The solution for the mechanical motion within the transducer is simplified to a one-dimensional problem. This is because the generated acoustic wave within the transducer is considered as a plane wave. This assumption is valid since the square of the laser spot size is greater than the distance from the generation point to the detection according to Rayleigh criteria.

3.4 Transducer layer characterisation

In order to compare the theoretical and the experimental outcomes, it is crucial to know the accurate dimension of the device. In our experiment, after fabrication of the devices, the thickness of each layer in the device is measured (to tolerances of $\pm 5\text{nm}$) through a convenient post fabrication measurement method. This measurement is also implemented for those devices that are deposited using sputter coater due to high variations in deposition rate given by the sputter coater. Thickness measurement of the layers is carried out based on the collected transmission spectra from the device. The device is a multilayer structure with two parallel reflecting surfaces, separated by an optical cavity. The two reflectors are gold layers and the spacing between them can be from ITO or a polymeric material such as polystyrene or cellulose acetate.

In the first step the theoretical transmission spectra $T(\lambda)_{\text{model}}$ from a gold layer with thickness of 1 to 70 nm is saved to a file. The experimental transmission spectra $T(\lambda)_{\text{exp}}$ also is saved in the same file and is loaded by a script in order to fit and compare the experimental spectra with all theoretical curves, as shown in Figure 25 (the top figure). The similarity between each theoretical curve and experimental one is scored from 0 to 1 (the bottom graph of Figure 25):

$$\text{Score} = 1 - |T(\lambda)_{\text{model}} - T(\lambda)_{\text{exp}}| \quad (30)$$

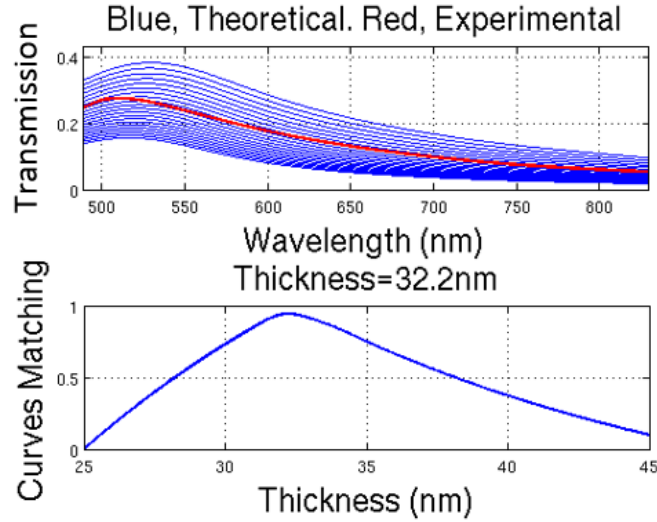


Figure 25: Example of the output that used to evaluate the thickness of a gold layer. In the top graph, the blue lines present the model spectra and red line represents the experimental traces. In the bottom graph, the maximum score corresponds to the best match between theoretical and experimental curves.

The scores are plotted as a function of thickness. Figure 25 shows an example of the output of the script used to measure the thicknesses of gold layers in FPI structures. The thicknesses of both gold layers at top and the bottom of an optical cavity are measured based on this method while fabricating a transducer through following steps: a number of clean substrates are placed inside the chamber. In the case of fabricating a gold layer on the tip of single mode fibre, a clean substrate also can be placed next to the tip where the gold is deposited. After depositing the first gold layer (bottom layer), one of substrates is taken out from the chamber to measure the thickness of the gold layer using the scoring method outlined above. Once the transmission spectra were collected, the sample either is returned to the chamber to add the ITO layer or is taken to deposit a polymeric layer using a spin coater. After the deposition of the cavity spacing of a FPI transducer, all the devices that have the first gold layer and the spacer are returned back into the sputter to deposit the second gold layer (the top layer). A clean substrate is also included in order to measure the top gold layer thickness. Once the whole multilayer structure is deposited on the top of substrates and the thickness of gold layers measured, the thickness of the spacer is evaluated by performing a fit to theoretical curves.

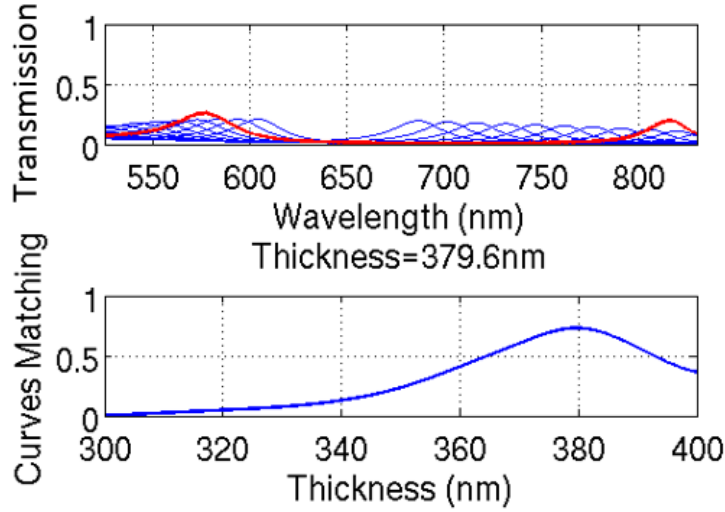


Figure 26: Example of the output of script used to evaluate thickness of an ITO layer in a three layer transducer structure. In the top graph, the blue lines present the model spectra and red line represents the experimental traces. In the bottom graph, the highest score is related to the thickness of ITO layer.

The theoretical transmission spectra are calculated for a range of spacer thickness and based on the measured thickness for gold layers at top and bottom, the thickness of spacer is evaluated experimentally. Since the thicknesses of the top and bottom gold layers are slightly different for each deposition, a theoretical curve to measure the spacer thickness is need to be calculated each time the Matlab script runs. As shown in Figure 26, the same method as with the gold layers was used to measure ITO thickness. The method of scoring between the experimental and theoretical curves is calculated as it was described for scoring gold layers. All the deposited layers within a device can be measured at the end of this process. The optical (i.e. optical sensitivity ($\delta R / \delta H$ and $\delta T / \delta H$)) and mechanical (i.e. frequency response)) behaviour of the devices are also validated using the measured thickness of the layers (see section 2.4).

In summary, the transmission spectra as a function of wavelength are modelled using Fresnel equations. The theoretical and experimental spectra are used to calculate a score between experimental and theoretical spectra using Equation 30. The score point is fitted to a curve and finds the position of its maximum. The result then is displayed.

3.5 Fabrication methods to deposit thin films

Different fabrication methods are used in this study based on the material used in each experiment. In this study, a wide range of frequencies from several GHz to hundreds MHz were produced using several different approaches. These approaches included using different materials, and increasing the spacer thickness which require different fabrication methods. Sputter coating is one of the fabrication methods which involves deposition of a metal-multilayer-dielectric structure and spin coating is used in micro-fabrication of polymeric thin films on the top of glass substrates. As an alternative to spin coating, dip-coating method is employed to fabricate a polymeric thin film on the tip of an optical fibre.

Another approach for lowering the frequency to MHz range, rather than fabricating a thicker spacer, is changing material from ITO to polymer. Typically, a few microns of polymer thin film offers the prospect of several tens megahertz [49]. Polymer materials offer many attractive properties including high flexibility, small Young's modulus relative to silicon/glass, low cost, biocompatibility, relatively good processability and tuneable properties. In addition, transparent polymeric thin films have a high transmittance in the Infrared region, making them suitable materials substitute to ITO. Most importantly, the properties of polymers can be tailored with greater flexibility as they can host a variety of functional dopants such as absorptive dyes [83]. Deposition of thin film layers using polymeric materials requires a specific fabrication method such as spin-coating or dip-coating.

3.5.1 Deposition of thin film layers using sputter coater

Deposition of conducting materials such as gold and ITO were performed with a sputter coater. Tooling factor is one of the FTM (Film Thickness Monitor) parameters that can be changed from the front panel keys. A correction for the difference in material deposited on the quartz sensor versus the substrate is called tooling factor [84]. A proper tooling factor (actual tooling factor) can be calculated using the following equation:

$$\text{Tooling}_{\text{Actual}} = \text{Tooling}_{\text{Approximate}} \times \text{Thickness}_{\text{Actual}} / \text{Thickness}_{\text{QCM}} \quad (31)$$

Where $Thickness_{Approximate}$ is an approximate value, $Thickness_{Actual}$ is the substrate's actual film thickness and $Thickness_{QCM}$ is a given value to deposited material. This procedure is needed to be repeated for a second or third time to improve the accuracy of measurements in this procedure [84].

3.5.2 Deposition of thin films using spin coater

Spin coating as a fast and easy method offers fabrication of thin and homogeneous films from solutions. There are several important factors that affect the generation of thin films with low roughness, including evaporation rate of the solvent, concentration of the solution, angular velocity (rotating speed), and spinning time. It is necessary to have a solvent with relatively high evaporation rate at room temperature such as acetone or toluene. The flow of the solution is influenced by the evaporation process. A solution with high evaporation rate solvents will flow slower as the viscosity of the solution increases. The film thickness depends on the evaporation rate of the solvent. There are 3 possible cases [85]:

- The solvent with relatively low evaporation rate in which the film thickness (d) depends only on the rotational speed (ω) and spinning time (t). $d = \omega^{-1} t^{-1/2}$
- The solvent evaporates with a constant rate. $d = \omega^{-2/3}$
- Solvent evaporation varies with the square root of the angular velocity. $d = \omega^{-1/2}$

It is expected that by increasing ω , the thickness (d) is decreased and *vice versa*.

3.5.3 Dip coating method

Dip-coating as a simple and economical method is widely used to deposit thin films onto any substrate. In the dip coating process, an aqueous based liquid solutions is used to coat the surface of any substrate directly with a single or multi thin film layer. Generally, the approach involves three important (Figure 27); immersion (dipping)

and dwell time, deposition and drainage (withdrawing), and evaporation (drying) [86].

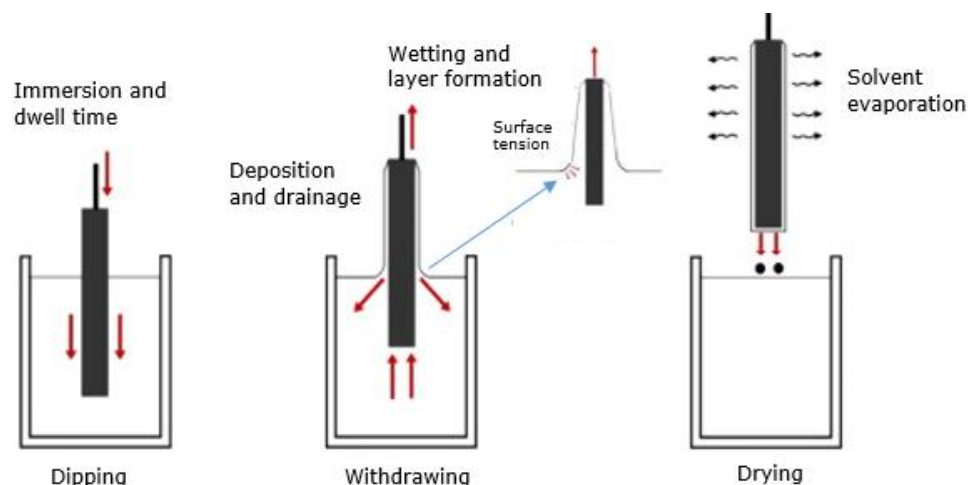


Figure 27: Schematic steps of dip coating; immersion (dipping) and dwell time, deposition and drainage (withdrawing), and evaporation (drying). This figure has been adapted and modified from [73].

The substrate is immersed into a solution at a constant speed (u_0) in order to provide enough time for sufficient interaction between the precursors and substrate in coating solution. The wetted substrates are then withdrawn from the solution at a constant speed. This allows a thin layer of precursor solution to be deposited on the surface of the substrate and excess liquid is drained from the substrate. A thin film is deposited once the solvent evaporates from the solution. The evaporation can be promoted by heating the substrate in order to obtain a dried thin film [87].

Thin film formation via dip-coating process is controlled by several parameters which are complex chemical and physical multi variable. Immersion time, withdrawal speed, dip-coating cycles, density, solution concentration, substrate surface and evaporation rate of solvent are the most important factors that determine the thickness and morphology of deposited thin films [88].

The withdrawal speed of substrate is commonly used in order to control the thin film thickness. The following equation estimates the relationship between the thin film thickness and the speed of the substrate withdrawal.

$$h \propto u_0^x \quad (32)$$

Based on the results obtained by Baupool et al., the “h” is the film thickness and u_0 is the withdrawal speed, where x is in the range of 0.4–0.6 [89]. These equations show that there is a strong effect of the withdrawal speed on the film thickness. The process conditions as well as the solution properties determine the differences in the value of x. A weaker film thickness is obtained by a smaller value of x. It was found that at faster withdrawal speed of substrate from the solution, the film is exposed to solvent evaporation at the surface of the liquid for a shorter time. Whereas at slow withdrawal speeds, longer time exposure of the film to solvent evaporation at the liquid surface leads to thinner thickness [89].

In summary, in this study a three layer structure consisting of an optical resonating cavity surrounded by two partially reflector layers is fabricated, similar to a zero order Fabry Perot. Ultrasound is generated by the thermoelastic effect after exposure of the transducer to a short laser pulse. The thickness of layers determines the resonance peak of the generation bandwidth of the transducer. By selecting the correct thickness of the layers, the sensitivity of the transducer increases as its reflectivity and transmittance changes in respect to its own height. This makes the transducer an efficient detector. The Brillouin scattering can be also detected by the transducer when the generation and detection occur in the same transducer. A high transmittance of probe laser beam is provided by an optical cavity where the correct materials and dimension of the layers are chosen. This allows the detection of Brillouin scattering both in reflection and transmission. Different fabrication methods and materials are used to deposit the layers in FPI structure.

Chapter 4

Material and Methodology

4.1 Introduction

In this Chapter, the sample preparation, the experimental set-up, signal processing, fabrication methods and characterization methods will be presented and described. Different materials were used in a series of experiments in this study. From the mechanical and optical points of view, different materials were used to fabricate a three layer FPI structure. A thin layer of metal is deposited onto the glass substrate, forming a partially reflector surface with high absorptivity properties at the wavelength of the pump laser beam (415 nm). The spacer is fabricated from a material with high transparency property at the wavelength of the probe laser beam (780 nm) to increase the power level of signals available for detection.

Table 1: Thermal and mechanical properties of materials used in this study [90][91].

Material	Density Kg/m ³	Acoustic Velocity (m/s) V	Thermal conductivity W/(mK)	Thermal expansion coefficient (K ⁻¹)	Specific heat capacity J/(KgK)	Young's Modulus (E)
Polystyrene (PS)	1050	2450	0.33	0.7×10^{-6}	1300- 1500	3-3.5 GPa
Cellulose acetate (CA)	1250	3100	0.167-0.335	180- 270 $\times 10^{-6}$	1451- 1509	2.4-4.1 GPa
Gold (Au)	7140	3240	318	6.5×10^{-6}	129	70 GPa
Indium Tin Oxide (ITO)	998.3	3600	429	14.2×10^{-6}	700	150 GPa
Air	1.2	340	0.025	-	1.0035	0.1 MPa
Silica Glass	3965	5700	1.38	2203	703	73.1 GPa
Water	-	1496.5	0.6	0.5×10^{-6}	4180	-

Table 1 shows the thermal and mechanical properties of the different materials which are used in this study. These properties are then used to constrain numerical models in order to predict the sensitivity and to optimize the mechanical response of the transducers, respectively. Single-mode optical fibre (780-HP, Thorlabs) is used in this study in order to increase the deposited energy per pulse relative to core diameter of the fibre. As it can be seen in equation 4 (section 2.3.1) the deposited energy per pulse increases as the core diameter of the fibre is decreased. The fibre is designed to operate at near IR wavelength with cladding, coating, and core diameters of 125 ± 1 , 245 ± 15 , and $4.4\mu\text{m}$. The operation wavelength is between 780nm-970nm with second mode cut-off wavelength of $730\pm 30\text{nm}$. The pump laser beam at the wavelength of 415nm was also coupled into the same fibre. However, the delivery of two laser beams from different wavelength regions into the same fibre was challenging. This is because the fibre is a single mode at all wavelengths above the cut off wavelength and high attenuation occurred due to grating damage (see section 1.2, page 15).

4.2 Substrate preparation

For the purposes of this study, two types of transducers were fabricated (i) using large glass substrate (i.e. plate transducers), and (ii) small (sub-mm) transducers on the tip of the optical fibre (i.e. fibre-based transducers). The substrate must be cleaned before fabrication of the transducers. The substrate cleaning is counted as an important part of the fabrication process due to the very small dimensions of the thin layers. This allows the thin films in the transducers to be fabricated as a smooth and contamination-free surface. Cleaning the substrates also increases the adhesion of the transducer to the glass substrate.

4.2.1 Preparation of the substrate for plate transducers

For large plate transducers, microscope glass slides were used as substrates. In four steps the substrates were prepared for fabrication by submersion of the substrates in four different solvents (i.e. ethyl-lactate, acetone, methanol and isopropanol) using an ultrasonic bath (Fisher SW3H). The substrates were immersed in each solvent in the given order for 8 minutes in each cycle. Bear in mind that before submerging the

substrate in the next solvent, they were rinsed with the next solvent. The substrates were dried using pressurised nitrogen after the last cycle. The cleaning process is the use of different vapour solvents to remove contaminations such as grease, oil, carbon, and organic impurities in four steps. Ethyl lactate is biodegradable solvents, acetones removes organic impurities such as greasy/oily contaminations, methanol removes acetone residues, and isopropyl is used to avoid striations on the substrates. The process was performed based on published patent by Kennison et al [74].

4.2.2 Preparation of the substrate for fibre-based transducers

Before fabrication and cleaning the optical fibre tips, the outer jacket of the fibres were carefully removed to avoid damaging the fibre using a stripping tool (Three-Hole Fibre Stripper, FTSA (Thorlabs)). The tips of the optical fibres were then cleaned with an acetone wipe. To create a flat surface at the tip, the optical fibre was cleaved using a cleaver (High Precision Inno Optical Cleaver Vf-15hE). The fibre tip was then monitored using an ordinary light microscope to check whether it is flat and clean.

As shown in Figure 28 , a container was designed in order to hold and expose the fibre tips and protect the rest of the fibre inside the chamber of a sputter coater. It consisted of a cylinder shaped dish with a loose fitting cover over the top and sides. Multiple holes with diameter equal to outer diameter of ceramic ferrules (Thorlabs, UK) were created on cover the top in order to hold the ferules. Several ceramic ferrules for LC/PC connectors (Thorlabs, UK) were fixed in the holes: outer diameter of 1.25mm x 6.4mm long, with a 126 μm hole size. Once the fibre was prepared, it was carefully inserted into the ferrule so that the tip was flat with its end face PC-polished by a fibre stopper.

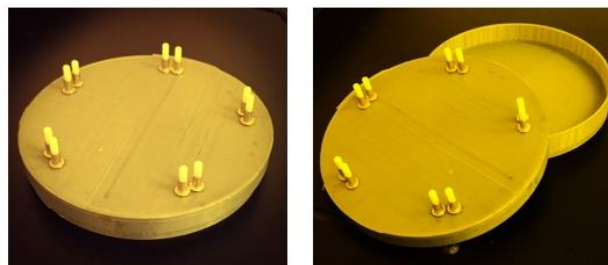


Figure 28: A container designed in order to expose the tip of fibre and protecting the rest of the fibre inside the chamber of sputter coater.

4.3 Experimental setup

For the experimental setup, two 150 femtosecond pulsed lasers (150 fs pulse width) with the repetition rates of 80 MHz and 12 ns time delay between pump and probe were used. The experiment is based on a dual laser asynchronous optical sampling (ASOPS) laser from Menlo. The wavelength of the probe laser can be tuned from 700nm to 900nm. A maximum of 4 mW average power was used in the probe and 1 mW in the pump (415 nm) corresponding to pulse energies of 40 and 10 pJ and peak powers of 240 W and 60 W, respectively (see section 2.3.1, equation (1) and (2)). A laser power meter (PM206, Thorlabs) was used to measure the power of the pump and probe laser beams. Different arrangements were used to test the transducers either in a free-space or a fibre-based setups. A lower average power was used for excitation of the samples in the fibre-based setup (about 0.3 mW to produce a peak power of 18 W) to prevent damage to the optical fibres.

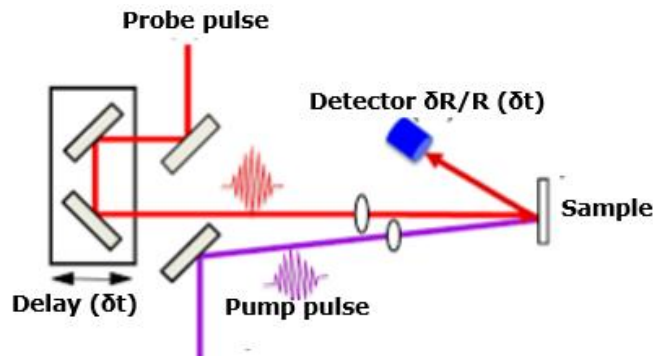


Figure 29: Schematic of ultrafast time resolved pump and probe technique. The figure has been modified from the www.phys.com website [63].

Figure 29 shows a simplified schematic of the pump and probe technique. In the current experiment two pulsed laser systems were used to provide a time delay between the pump and probe laser pulses. The electronics allow one laser to be locked at 100 MHz repetition rate and the other laser to be locked at 100 MHz + 12 KHz. This means that the lasers provide the equivalent of a 12 ns delay line sweep every 120 microseconds. The pump and probe beams are focused to the same location on the top of the transducer and the probe response is recorded as the delay is swept [65].

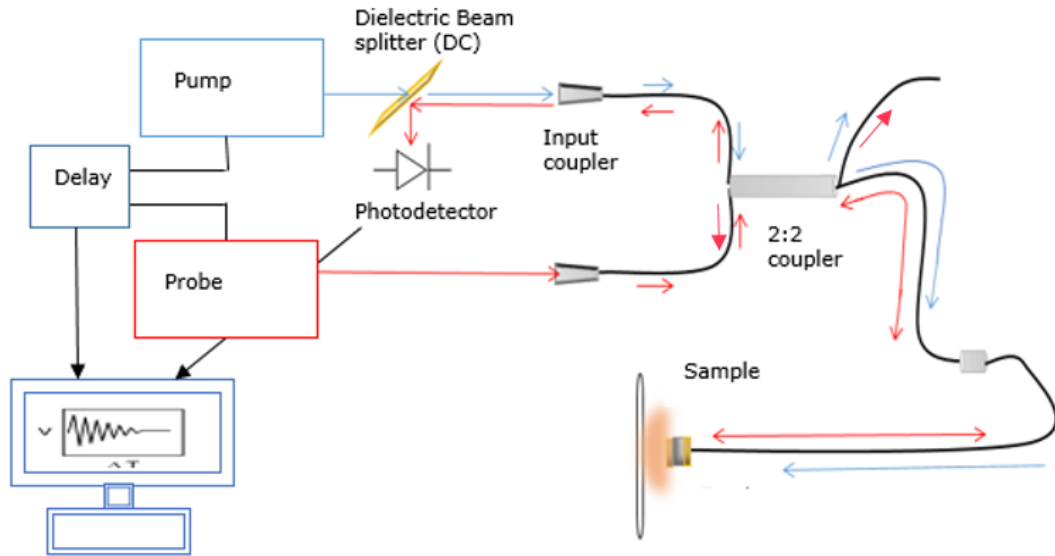


Figure 30: Schematic of a fibre-based experimental system. The system comprise of two optical paths for the pump (blue line) and probe (red line), and the detection is taken place in reflection mode in this case. The components of the system are explained in the related section. Only the probe beam pathway in reflection is considered in here.

Figure 30 shows the schematic of an ideal fibre-based experimental setup since the signals are detected in reflection by a photodetector coupled in one of the input arms of a 2:2 single mode fibre coupler (50/50, 780nm, Newport). In this case the delivery of two laser beams at the wavelength of the pump and probe take place in two illumination optical paths. Both beams travel down to the sample via the same optical fibre. The essential parts of these setups are two illumination optical paths from pump and probe beams and a detection arm. The optical paths were designed to allow control of the time delay between synchronously emitted short laser pulses. It should be noted that coupling the two laser beams with different wavelengths into the same fibre using the directional coupler cannot be 50:50 at all wavelengths.

The pump laser pulse was used for excitation of the sample and the probe laser pulse was used for measuring the response of the sample (see section 2.3.1 and section 2.3.2). The wavelength of 415nm was chosen because gold absorbs the light at this particular wavelength leading to a strong thermo-elastic generation of acoustic waves.

The probe laser pulse was chosen at the wavelength of 780nm where the spacing between two gold reflectors shows strong light transmission.

As shown in Figure 30, following the laser pulses from two different pathways, the pump laser beam is delivered into a 2:2 optical fibre coupler via a Dichroic Coating (DC) beam splitter. The coupler is used in order to reduce the peak power of the laser pulses on the sample (see section 2.2.1, equation 1 and 2). The DC beam splitters are designed to split laser beams operating in the visible through infrared wavelengths. 50% of the pump laser beam is then delivered onto the fabricated transducer on the tip of a single mode fibre (SMF). The probe laser pulses are then delivered to the same pathway as the pump laser pulses have been delivered to the sample. The reflected probe laser beam (i.e. the response of the transducer) from the sample travels down to the coupler and 50% of reflected beam is delivered to a photodetector by a DC at the wavelength of the probe laser beam. The intensity variation of the probe beam is monitored by the photodetector, which is captured and averaged on a digital oscilloscope (DSO).

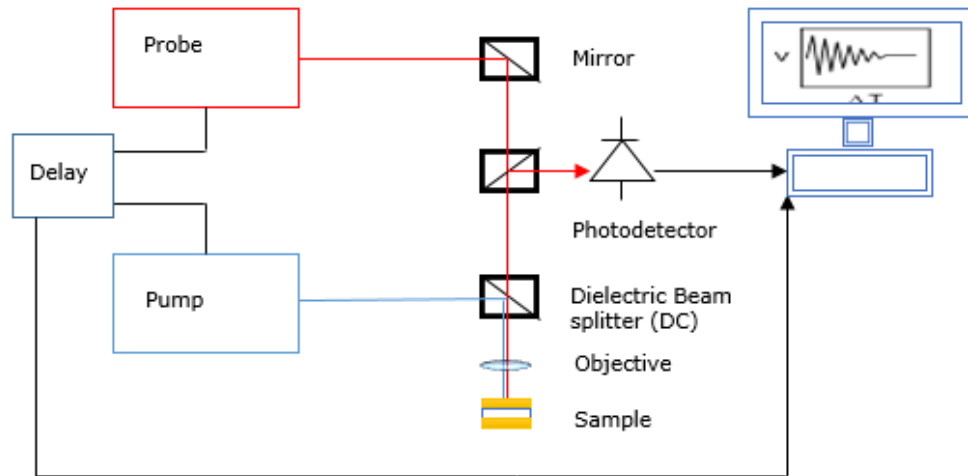


Figure 31: Schematic of a free-space experimental system. The system comprises of two optical paths for (pump and probe) and the detection takes place in reflection mode in this case where the probe light reflected from sample to an objective lens, two beam splitters and a photodetector.

The plate transducers were tested in a free-space experimental setup as shown in Figure 31. The optical and mechanical characterization of a particular size of the fibre-based transducers can be evaluated using plate transducers before fabricating the same transducers on the tip of a single-mode fibre. This is because the fabrication and characterization of the fibre-based transducers is more time consuming and challenging where the plate transducers are easy to fabricate and to test. The fibre coupling is also slightly restricting to test the fibre-based transducers where they can be easily damaged during handling. The irradiated area on both sample types are $\sim 4.5 \mu\text{m}$ diameter, for a laser spot and the core diameter of a SMF. The beam from the pump laser pulse is reflected by a DC and focused by an objective lens (i.e. planar-convex) to a sample. The laser was focused on the samples using a 250 mm focal length to a focal spot size of $\sim 4.5 \mu\text{m}$ diameter, providing a laser intensity of $\sim 0.5 \text{ W/cm}^2$ with 150fs pulses. The beam from the probe laser pulse is reflected into the same optical path from a mirror at a given wavelength of the probe laser beam. It is then transmitted through a partly transmitting mirror (beam splitter) through a DC beam splitter to the sample. Before the beam interacts with the sample, it is focused by an objective lens. The reflected beam from the sample is then collected by a photodetector.

An alternative fibre-based setup was used in this study as shown in Figure 32. In alternative configurations, it is possible to excite the sample by the pump laser beam on the other side of sample rather than delivering the pump laser beam via the SMF. This is performed due to the high attenuation where two different wavelengths of the pump and probe were coupled into the same fibre. It may also be possible to detect both reflected and transmitted beams simultaneously.

Detection in transmission mode was performed where there is high transmission for the probing wavelength λ_{probe} . An identical fibre-based experimental setup was built for detecting the probe laser beam in transmission mode with the photodetector placed on the other side of the sample, as shown in Figure 32. Detection in reflection mode also can be provided using different configurations where the sample is excited by delivering the pump laser beam outside of the coupler and detection takes place by delivering the probe laser beam by one of the coupler arms. In this configuration,

the photodetector is connected in one output arm the coupler where the pump laser beam was connected to the coupler and the sample excited externally (Figure 33).

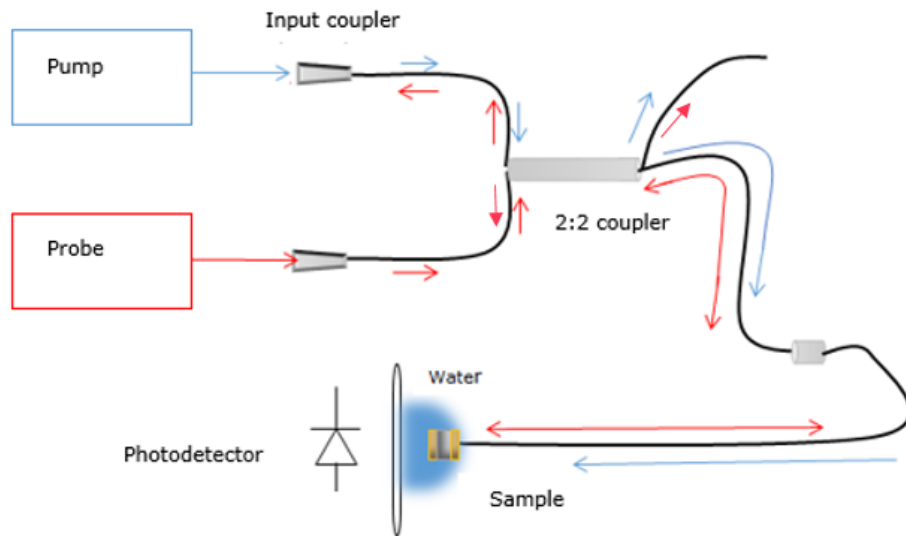


Figure 32: The fibre-based experimental setup for detection in transmission mode where the photo detector is placed on the other side of the samples. Only the probe beam pathway in reflection is considered in here.

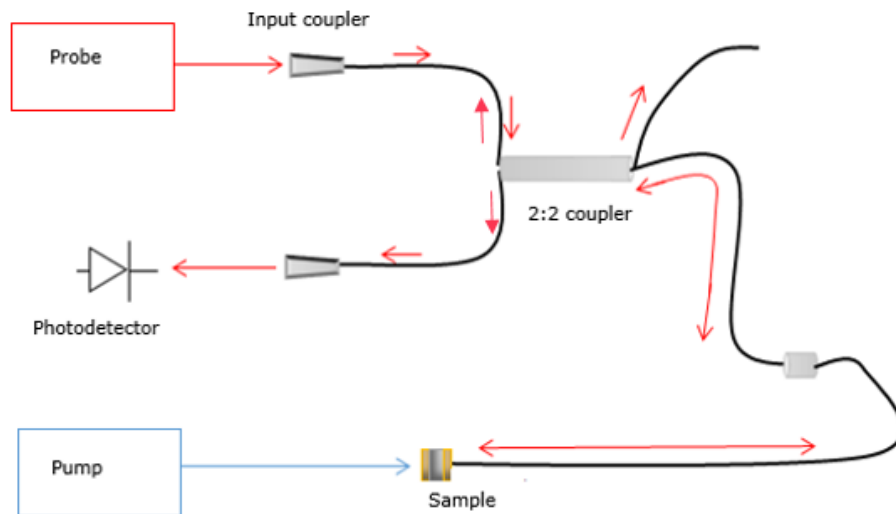


Figure 33: The fibre-based experimental setup for detection in reflection mode and the sample is excited by delivering the pump laser beam from the other side of the samples.

4.4 Signal processing

Either a single or an array of time traces is collected to evaluate the mechanical frequency response of the transducers. An array of time traces is collected because the larger sampling rate provides the accuracy measurement. Each time trace is decayed and stabilised after a rapid drop of signal level which is known as coincidence peak due to the transient change of the optical reflectivity induced by absorption of the pump laser pulses on thin film of gold (Figure 34(a)). The absorption of the short pulse of light produces heat which causes the material to expand rapidly generating stress (thermal expansion) which is followed by the relaxation of the materials in the transducer. There are several signals of interest, such as the resonance of the transducer, Brillouin signals (see section 2.4) from substrate and tested samples. The mentioned signals are decaying sinusoidal waves that are presented with different amplitudes, frequencies, and decay rates (see section 3.4). By the following steps each trace can be processed to obtain desired information. The area of interest is truncated from the main trace and the thermal background is removed by subtracting a fitted function. For this a polynomial functions (polyfit and polyval) are performed, plotted, and subtracted from the main trace. Then the peak frequencies are evaluated by performing a fast Fourier Transform (FFT). Filtering in time domain is performed for display purposes (see Appendix, A.2.1).

4.4.1 Thermal background removal

The temperature of the sample is increased after the absorption of a pump pulse by the sample. The refractive index is modulated by this change in temperature. This is because the temperature change in the materials induces a change in the density of the materials. The refractive index is not only modulated by a change in temperature, but also by the induced strain. The reflection and transmission in a provided material are locally affected by the modulation in refractive index. This leads to a slow modulation of the light intensity in laser ultrasound experiments, which is referred to as the “thermal background” effect. The period of the thermal background is longer in comparison with the duration of the acoustic signals due to thermal expansion but they can overlap in time (see section 2.3.1). The thermal background removal is

important in order to observe the signal of interests. In order to remove the thermal background from the main signals, the area of interest was cropped from the raw trace and fitted to a polynomial curve with fourth to sixth order. The higher order of the polynomial fit causes to lose the main signals. The fitted curve is then subtracted from the source trace where it was verified by overlapping it with the source trace on a graph.

Figure 34 shows an example of the described process to obtain the main signal. The signal presents the resonance of the PS plate transducer that was obtained experimentally using free-space setup (Figure 33). In Figure 34 (a) presents a graph of a cropped raw trace and fitted curve on top of it is shown in green line in this part. The fitted curve is subtracted from the source trace once it was verified that the fitting curve follows correctly the source trace¹. Then the fitted curve is subtracted and Figure 34 (b) shows a filtered version of the thermal background-free trace.

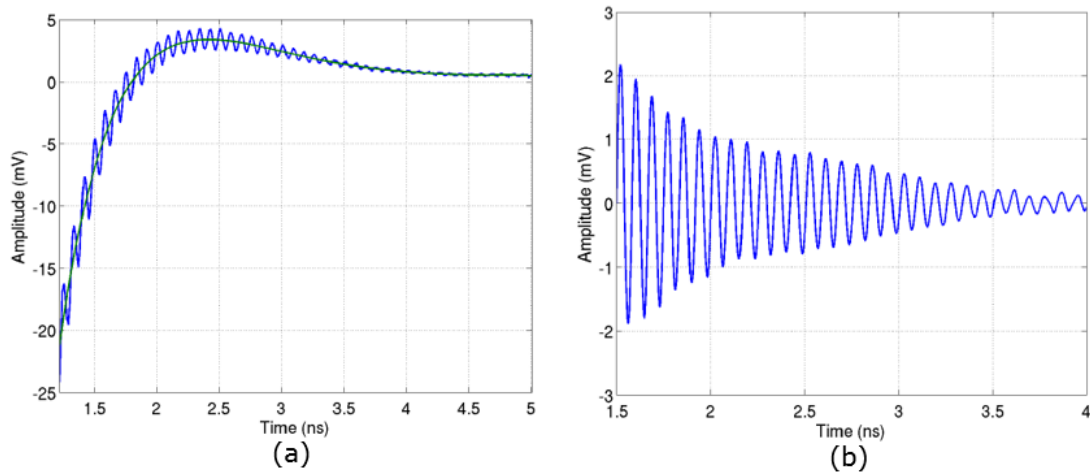


Figure 34: This is an example of thermal background removal for a PS plate transducer: (a) cropped data (blue line) and fitted thermal curve (green line). Then the fitted curve (green line) subtracted from data to remove thermal background and obtain the filtered trace (b). The figure is adopted and modified from Perez Cota study [49].

4.4.2 Determination of peak frequency

The frequency components of the signal were evaluated by calculating the FFT of a filtered trace. The frequency can be investigated by observing its peaks. In order to save time, a range of expected frequencies and a threshold on the amplitude were set to evaluate the frequency of a peak automatically (Figure 36). There were several signals produced from a measurement such as noises or undesirable peaks which were prevented to be evaluated through this process. For example, the high frequency peaks ($\sim 27\text{GHz}$) mostly are produced by gold layers as shown in Figure 35. These peaks have a low level of intensity in comparison to the intensity of resonance frequencies. By setting the threshold on amplitude (black line in the Figure 35), the frequencies of noise peak are prevented to be measured.

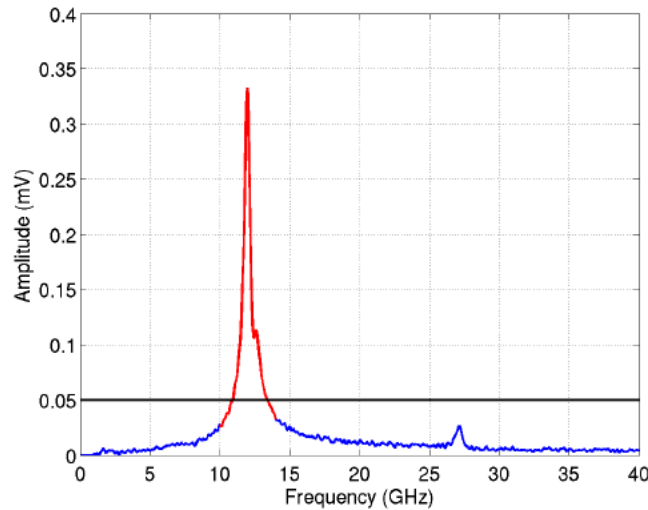


Figure 35: Example of determine of peak frequency. The red race is a range of frequencies to investigate and the blue trace indicates the original trace, where the black line indicates the threshold level. The red and black traces are set by the user manually. The figure is adopted from Perez Cota study [49].

4.5 Fabrication methods

Different fabrication methods were used in this study based on the materials used to construct the transducers designed for each experiment (see section 2.8).

4.5.1 Deposition of thin film layers using sputter coater

Depositions of conducting materials such as gold and ITO were performed with a sputter coater (Emitech K575X). An additional thickness measurement is carried out due to the poor accuracy of this method (see next section). The dimensions of the transducer were measured by comparing the transmission spectra from transducers and predicted spectra from numerical models. This method offers a high accuracy of measurements and prediction of the transducers' performance based on the optical and mechanical models.

4.5.2 Deposition of thin films using spin coater

Polystyrene (PS) (GPC: MW=100,000) films were spun on the top of glass substrates coated with gold from five solutions with different concentrations of PS (from 4 to 20% W/vol). Toluene anhydrous 99.8 % (Sigma Aldrich) was chosen as a solvent for PS. The initially stationary substrate was covered with polymer solution and then accelerated to the desired rotation rate for 30 seconds. Spin speeds were ranged from 500 to 3000 round per minute (rpm). To cure the coating, the substrate was heated at 100° C for 60 min. Another thin film layer of gold was deposited and transmission spectra are collected from the structure. The spectra were used to calculate the thickness of the PS thin film layer as it is explained in the next section.

4.5.3 Deposition of thin films using dip coating method

For fabrication of the transducers on the tip of an optical fibre, dip coating is a suitable method to deposit the polymeric thin films. In this study, a custom-made dip-coater (Figure 36) was used. The machine was equipped with a servo motor with 180 degree rotation limit both in clockwise and anti-clockwise direction. The speed of rotation was adjusted between 1° and 100°. In this study, the rotation degree (10°/sec) was used as a speed and therefore we could not measure the actual speed in terms of m/s. The cleaned fibre-based substrates were immersed into the solution bath at a constant dipping rotation speed (10°/sec) and kept inside the bath for different time windows (dwell time). The substrate was then withdrawn from the bath with a constant speed

as it was immersed into the solution bath. The film was dried using a few different protocols (explained in details in next paragraph) and the resulting thickness and uniformity evaluated using SEM.

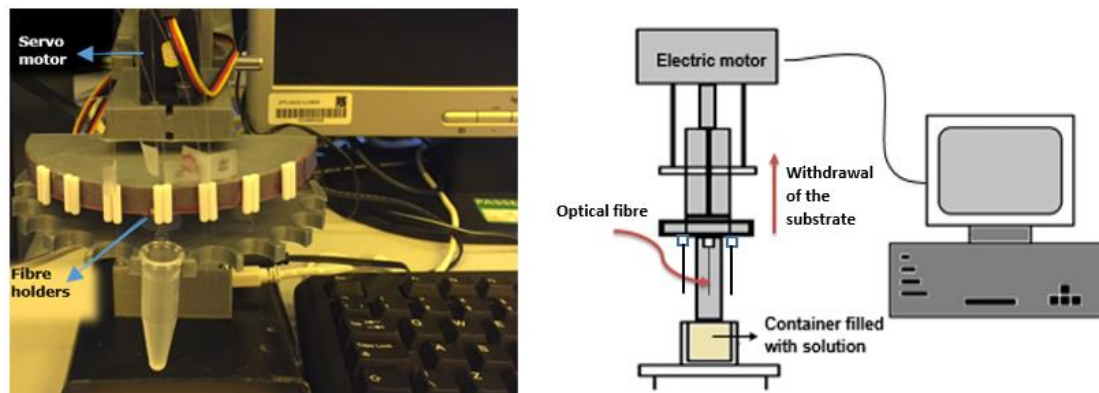


Figure 36: A custom-made dip-coater to hold the fibres and immerse them into the solution bath, and a schematic of dip coater machine which is controlled by Matlab code.

Two drying methods were applied to the polymer films on the tip of fibres, ambient drying and heat treatment. For ambient drying, the fibres were left at the vertical orientation at room temperature (25°C) for 30 min to dry where the evaporation rate of the solvent was high such as acetone or toluene. For heat-treatment drying, the fibres were placed into an oven, when the oven temperature was about 30–50°C. The ambient drying and heat-treatment drying are referred to as “slow” and “fast” drying, respectively.

4.6 Characterization of thin films

Different techniques are used to characterise the samples and to measure the thickness or to observe the morphology of the samples. Transmission spectroscopic methods were used to measure the spectra in order to calculate the thickness via its effect on the spectrum transmitted through multi-layered structure of the transducers. Other techniques such as optical microscopy and SEM were used to observe the morphology and patterned roughness in a subset of samples.

4.6.1 Transmission spectroscopy

The thickness of gold layers can be measured using their transmission spectra. As shown in Figure 37, a transmission spectroscopy setup is used to collect optical spectra from the samples. An Ocean Optics USB2000+ fibre coupled spectrometer and a conventional light source are used to obtain the transmission spectra from the device.

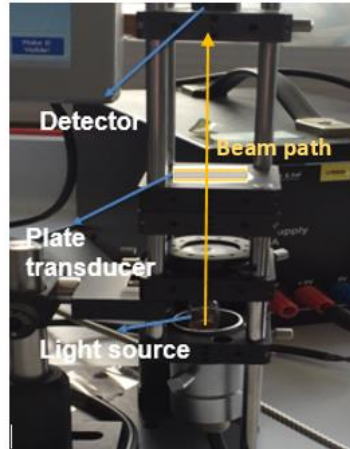


Figure 37: Transmission spectroscopy setup consists of a halogen lamp as a white light source, and a detector to collect optical spectra transmitted through the plate transducer which has not been shown in this picture.

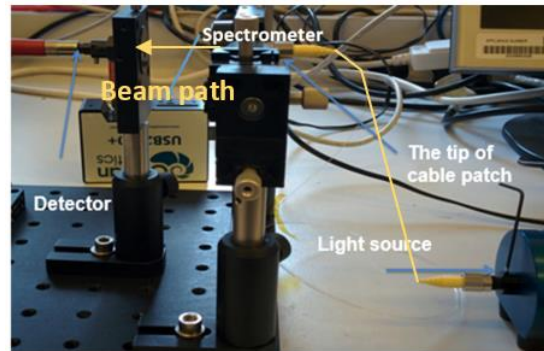


Figure 38: Fibre-based transmission spectroscopy setup consists of a white light source, and a detector to collect optical spectra transmitted through the transducer fabricated on the tip of a single mode fibre (cable patch).

The dimension of the devices fabricated on the tip of single mode fibre was measured directly from the fibre. The transmission spectroscopy setup is shown in Figure 38 , in

which the optical fibre with the transducer under test fabricated at its tip is directly connected to the light source. The transmission spectra is collected by an Ocean Optics USB2000+ fibre coupled spectrometer.

4.6.2 Spectroscopic Ellipsometry method

Ellipsometry is the best known technique to characterize materials using the polarization of the light. In this study, a spectroscopic ellipsometer (J.A. Woollam Co., USA) was used to calculate the thickness of transparent thin film deposited on the glass substrate. The thickness ranges from sub-nanometers to a few microns can be measured using ellipsometry. However, a limitation in measuring thin film thicker than 200 nm was observed. This is because in thicker thin films the oscillations become increasingly difficult to resolve, except with longer infrared wavelengths. In this case other post characterisation techniques are preferred such as the transmission spectroscopy as previously mentioned [59].

4.6.3 Scanning Electron Microscopy

In this study, the morphology and topography of the fabricated transducers either on the tip of optical fibres or on top of plate substrates were examined using Scanning Electron Microscopy (SEM). The samples must be cleaned and dried before examining them. They must be dried because the vacuum will remove the water which damages the vacuum system [93]. Then the samples were fixed using a double sided sticky Kapton tape and were mounted on a holder which is typically a metallic stub. The proper orientation of the specimen on the mounting stub is important before attaching it. Re-orientation of samples after inserting the sample into the SEM can be difficult and can cause damage to the sample. The sample can be also coated with about 20 nm to 30 nm of conductive metals such as platinum or gold to increase the conductivity of the sample in SEM and to prevent damage due to building up of high electrical charges on the samples. Accumulation of charge on the sample can distort the image.

4.6.4 Optical microscopy

The detection of large in-plane length scales as compared to SEM was performed with optical microscopy using a Zeiss AxioTech 25H optical microscope with magnifications between 10x and 50x. The thin films were displayed by a camera (Hitachi KP-D50 CCD).

4.7 Manipulation of light absorption for the pump laser beam

In this experiment, an obstacle was experienced in delivering of the two pump and probe laser beams in the same single mode fibre due to a high attenuation (see section 3.1) at the wavelength of the pump and probe laser beams. It was suggested to tailor transducer for an available wavelength for the pump laser beam by adding an absorptive dye to a polymer-based medium to bring two pump and probe beams in almost same range of Near Infrared (NIR) (see section 2.7). At this point different polymers rather than PS were also tested to find the best mixture of polymer along with the dye dissolved either in the same solvent or compatible solvent for the chosen polymer.

4.7.1 Materials

Aluminum 1,8,15,22-tetrakis(phenylthio)-29H,31H-phthalocyanine chloride (APC) with 90% dye content and absorption ~ 748 nm and Cellulose acetate with the average molecular mass (M) $\sim 50,000$ by GPC were purchased as a white powder from Sigma-Aldrich and used without any additional purification [75]. The molecular formula of the dye is $C_{56}H_{32}AlClN_8S_4$ (APC) its chemical structure is shown in Figure 39. The ethyl acetate, acetone, and dimethylsulfoxide (DMSO), ethanol, toluene, water and chloroform were used as organic solvents and utilize as purchased. All the experiments are made at room temperature (25°C).

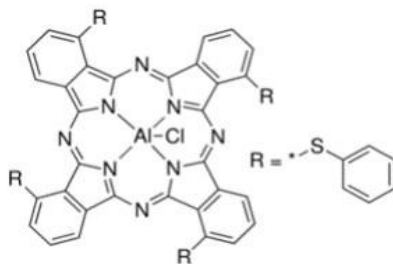


Figure 39: Chemical structure of APC dye compound. The picture is taken from the www.sigmaaldrich.com website [75].

Cellulose acetate (CA) is the most important cellulose ester that are commercially available as a white powder product. It is nontoxic, and odourless amorphous product. Cellulose acetate is the acetate ester of cellulose that is an organic compound with the formula of $(C_6H_{10}O_5)_n$, a polysaccharide. The general structure of the cellulose acetate is shown Figure 40.

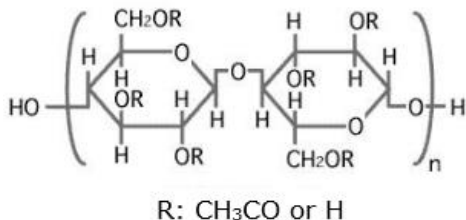


Figure 40: General structure of cellulose. The image is taken from www.daicel.com [94].

They are usually dissolved in a suitable organic solvent such as acetone or ethyl acetate. Some physical and mechanical properties of the materials are given in Table 1. Cellulose acetate is a semi-synthetic polymer obtained through the esterification of acetic acid with cellulose as a natural polymer. Cellulose is a polymer consisting of anhydroglucose (AUG) units as repeating units, and has three hydroxyl groups per unit [62]. Cellulose acetate is used in coating thin films because of its high resistance to sunlight, particularly to ultraviolet rays. It is the most stable among cellulose derivatives, and is a good pigment carrier. It is also a biocompatible material that can be utilized in medical equipment. It has a glass transition temperature (T_g), melting point of (T_m), and refractive index (n) of 180°C, 230-

250⁰ C , and 1.476 respectively [90]. The T_s and T_m are important where annealing or a high temperature is need to dry samples.

4.7.2 Solvent selection for both polymer and dye

The cellulose acetate was tested as a polymer to deposit the spacer. The aim of this part of study was to find a compatible solvent for both host polymer (i.e. CA) and APC dye as a guest substance. Also the compatibility of a solvent for a mixture of CA polymer and APC dye were investigated.

4.7.2.1 Choice of solvent for dye

The chemical structure and molecular mass of the dye was provided by the Sigma but no information was provided in regards to compatible solvents to dissolve the dye. To investigate the solubility of the dye in different solvents, it was dissolved in different solvents including acetone, DMSO, chloroform, ethanol, toluene, and water. The samples were left overnight on a stirrer to be dissolved. Each solution was spin coated on a glass substrate (Figure 41) and transmission spectra were collected at both wet and dry state of samples.

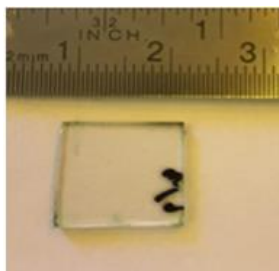


Figure 41: A small piece of glass microscope slide shows the approximate size of the plate transducers.

4.7.2.2 Solubility determination of polymer in different solvent

In this part of the study, the first solvent needs to be selected that is suitable for both dye and CA as a host polymer, and by selecting the second solvent that is soluble with the first selected solvent. The first solvent was included ethyl acetate and DMSO

solvents and the second solvent was a mixture of acetone with either ethyl acetate or DMSO at different ratios.

4.7.3 Compatibility of dye within Cellulose acetate host polymer

In this section, the incorporation of the APC dye and CA polymer was investigated. DMSO was selected as the suitable solvent. A mixture of CA and dye solution was prepared in a micro test tube at the ratio of 2:1 respectively by adding 200 μl of 40% CA solution (already prepared solution) dissolved in DMSO to 100 μl of prepared 4% wt./vol dye solution. The polymer-dye thin films were prepared at the spin speeds ranged from 200 to 3000 rpm (increasing order) with the spinning time of 30 seconds (see section 3.5.2). The same procedures were repeated to make the same samples but without adding dye, to act as a control for each samples. The thin films were then baked at 30°C for 30 minutes. The texture of polymer-dye thin films was observed under a light microscope and transmission spectra were collected both in wet and dried states. The spectra were normalised with respect to the control spectra, then they were plotted against wavelength.

4.7.4 Thickness analysis of thin films with different polymer concentration

In this part of study the effect of concentration of CA dissolved in acetone, and DMSO was observed separately. 10%, 20%, 30%, 40%, and 50% CA solutions dissolved in acetone were made and spin coated separately. The velocity used in this experiment was 3000 rpm. The spin coating duration for all samples was 30 seconds. To remove residual solvent from the layer, the samples were placed in the oven at a temperature of 30°C for 30 min after spin coating. Ellipsometry was used to calculate the thickness of each thin film.

4.7.5 Optical constant determination of dye-polymer thin films

In this part of experiment, the effect of dye concentration on the optical properties of APC dye mixed with CA thin films were investigated: extinction coefficient (κ), and absorption coefficient (α) of thin films were calculated (see Appendix, A.1).

Fifteen thin film samples with different concentrations of dye in CA solutions were prepared by the spin coating method on the glass substrates.

30% wt./vol dye-CA solution was made as previously explained (dissolved in mixture of acetone and DMSO solvents) using three different concentrations of dye solution (10, 15, and 20 %) at the ratio of 3:1. A colourless CA solution was also made using the mixture of solvents at the same ratio. Three sets of samples (Figure 42) were prepared at different spin speeds. Each set contained five samples. In each set, two samples were coated with the colourless CA solution: one on a gold coated glass substrate, and on a glass substrate as a reference.

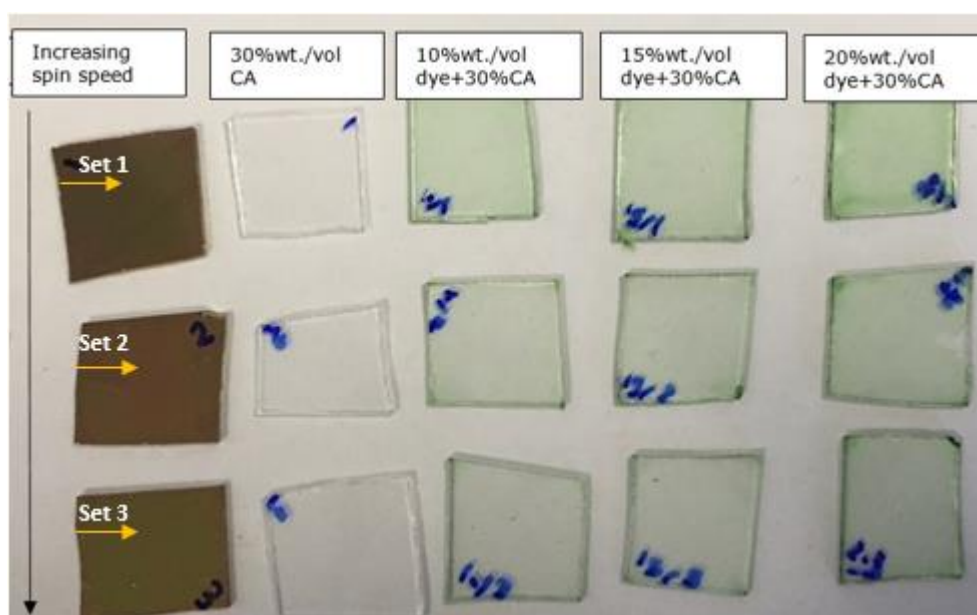


Figure 42: Three sets of samples, prepared with different concentrations of dye in CA solution and spin coated at three different spin speeds: 1500, 2500, and 3000 rpm.

The other three samples were coated with a dye-CA solution on the top of glass substrates. Three different concentrations were used to coat these three samples. All the samples was spin coated at the spin speed of 1500 rpm. The spin coating duration for all samples was set on 30 seconds. To remove the extra solvent from the layer, the samples were placed in the oven at a temperature of 30°C for 30 minutes after spin coating. The experiment was repeated at the spin speed of 2500 rpm and 3300 rpm using the same procedure. Another gold layer was deposited on

top of those samples with glass substrate coated with gold in the first place. Transmission spectra were collected from each sample separately and the thicknesses were calculated from the samples with the gold at the top and bottom layers. The thickness of thin film was determined by carrying out optical transmission measurements.

Transparent samples without dye were used as a reference for each sample. The thickness of each sample was calculated and used to calculate the absorption and extinction coefficients (κ) of each sample with specific dye concentration. For example for calculation of the κ of 20% dye solution, each thickness at different spin speeds were used to calculate the κ for 20% dye in CA polymer thin films for different thicknesses. For this, each spectrum was normalized by associated control sample and then absorption coefficient (α) was calculated. The α then was used to calculate κ (see Appendix, A.1). The samples were also observed using both optical microscopy and SEM.

4.7.6 Fabrication of thin and thicker devices to evaluate mechanical response

In this part of experiment the evaluated optical parameters, κ , and α for a wide range of wavelengths were used to optimise the fabrication and characterization of dye-CA thin films. The fabricated device from this part of experiment were used to evaluate the mechanical response for the dye-CA thin films to investigate the generation of acoustic waves via CA medium cavity.

Thinner devices (lower order devices) were fabricated using 30% dye-CA solution as previously explained (dissolved in mixture of acetone and DMSO solvents) adding 15% dye concentration solution at the ratio of 3:1. The dye-CA thin film was deposited on the top of a glass substrate coated with 22nm of gold layer (see section 2.3.1) at the spin speed of 5000 rpm. The spin coating duration for all samples was 30 seconds. To remove residual solvent from the layer, the samples were placed into the oven at a temperature 30°C for 30 minutes after spin coating. The second layer of the gold with the thickness of 22nm was deposited. The laser damage threshold was tested at different points for power levels of 1.5mW, 1mW, 0.5mW and

0.3mW for the pump laser. A thinner device was also investigated optically and mechanically.

4.7.7 Improving surface smoothness using single solvent

In this part of study, a single solvent (i.e. DMSO or acetone) was used to dissolve the CA. The procedure was the same as previously was described. The thickness and roughness of each sample were calculated using ellipsometry. The thickness of the samples at different spin speeds are presented in Table 5 (see section 5.4.3).

4.7.8 Increasing thickness and uniformity of thin films using sputter assisted layer by layer deposition

This part of experiment was performed to improve the uniformity and increase the thickness of CA thin films using sputter assisted layer by layer deposition of the thin layers of a multi-layered transducer. The simple multi-layered deposited whole stack is shown in a schematic diagram (Figure 43).

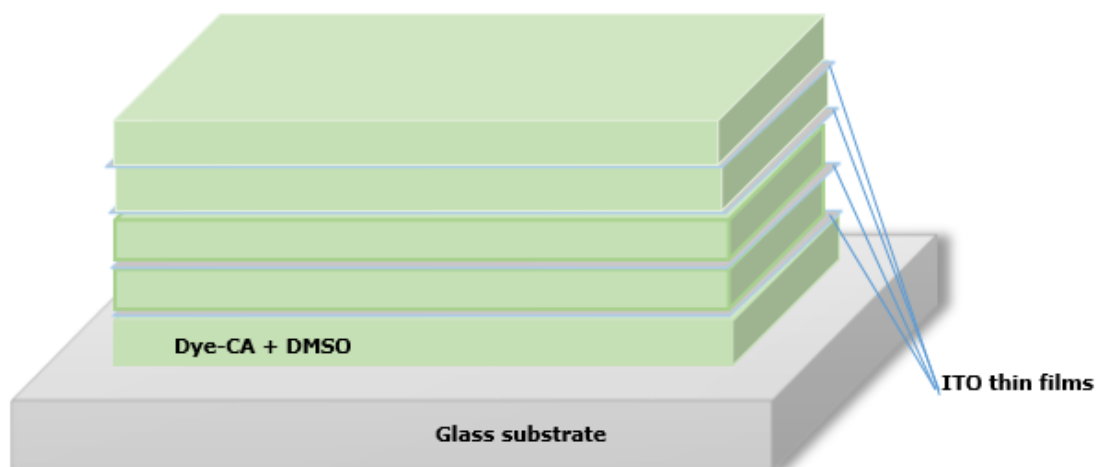


Figure 43: Schematic of a multi-layered transducer. The transducer comprises of polymeric thin films (dye-CA dissolved in DMSO) separated by thin film layers of ITO ($\sim 1\text{-}3\text{nm}$). The layer of thin films deposited on the top of a glass substrate.

Compared with single spin coating process to fabricate deposited film on top of glass substrates, this is an alternative method to produce a multi-layered transducer from the same material. Increasing concentration is not always an appropriate way to increase thickness; rather, to obtain a uniform thickness it was needed to find the optimal concentration and modify other factors to achieve the desirable thickness. Several samples were prepared with 10% dye mixed in 30% CA solution using acetone or DMSO as a single solvent. The thickness, roughness, and refractive index of each thin films were measured using ellipsometry and the results are presented in Table 9 (see section 5.4.8).

Chapter 5

Results and discussion

5.1 Introduction

In this Chapter, the theoretical and modelled results are compared with experimental data for fabricated fibre-based transducers and plate transducers. This Chapter presents outcomes from optical, mechanical models and incorporated experimental data with numerical models for ITO, PS and CA devices. The results from absorbing nanoscale thin films will be also presented in the current Chapter. An optical absorber system in a FPI was developed experimentally and numerically to tailor the transducer for an available wavelength for the pump laser beam, in order to control absorption behaviour of the dielectric layers (see section 2.6).

5.2 Results from fabrication of dielectric Indium Tin Oxide

A three-layered transducer based on FPI for the detection of acoustic waves in both reflection and transmission were described. The acoustic waves are generated by the same detection transducer. The sputter coating method was used to deposit the dielectric layers in the FPI-based transducers. The transducer design based on a gold-ITO-gold topology has been established. Then the optical and mechanical characterization of the transducers were performed and the outcome data were compared with numerical simulation output. The Fabry-Perot resonance frequency was lowered from several of GHz to hundreds of MHz by increasing the thickness of the thin-film cavity (section 2.4, equation 28), and the results are discussed.

5.2.1 Evaluation of optical and mechanical models

Many important characteristics of the transducers are defined by the optical model such as the optical transmittance, and the reflectivity which ultimately affect the mechanical response of the transducers directly (section 3.3 and section 3.4). In order to optimize the optical sensitivity and mechanical response of the transducers, those characteristics are essential for design purposes. The outcomes from numerical models also are fundamental to validate them against experimental measurements.

Reflectance for λ_{pump} (415nm) and λ_{probe} (780nm), and sensitivity of λ_{probe} can be evaluated using the optical model (see section 3.3).

Figure 44 shows the results of the optical model for a transducer with a wide range of gold (5-70nm) and ITO (100-600nm) thicknesses. The sensitivity of the reflected ($\delta R/\delta H$ (nm⁻¹)) beams are presented. As can be seen in the Figure 44, the sensitivity for reflection case decreases with increasing thickness. Also the resonance frequency is decreasing with increasing thickness of ITO from 10 GHz to 4.7 GHz and 3GHz (Table 2).

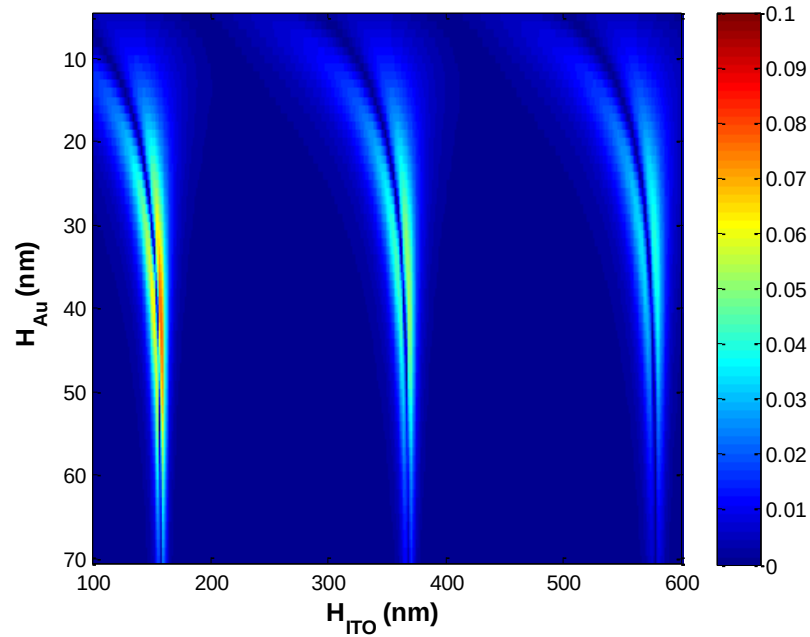


Figure 44: Transducer sensitivity $\delta R/\delta H$ at λ_{probe} (780 nm) for the ITO transducer with a wide range of thicknesses from 100 to 700nm. The colour bar presents the sensitivity per nm.

An example of transmittance comparison between model (blue) and experiment (red) is shown in Figure 45. As it can be seen there, is a good agreement between the model (blue lines) and experimental trace (red line), as the score is nearly one for an ITO plate transducer with thickness of 561nm (see section 3.4). The transmittance is 0.16 for λ_{probe} (780nm) and is 0.01 for λ_{pump} (415nm) (see section 3.1). To calculate the thickness of ITO, the transmission spectra of a single device, whose ITO layer thickness was unknown, was measured and compared with its modelled trace. The

evaluated transmitted spectrum of the transducer fits well to a 561nm thick layer of ITO. To calculate the resonance frequency of the transducer, the evaluated thicknesses are used in the mechanical models.

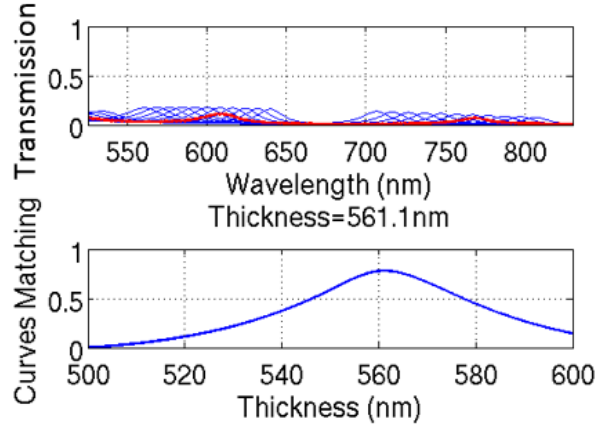


Figure 45: Example of transmittance comparison between the model trace (blue) and experiment (red). The blue traces represent different ITO layers. The experimental trace fits well to a 561nm layer of ITO fabricated on the top of a glass substrate.

Table 2 presents the evaluated data from the optical and mechanical models for the thicknesses of the fabricated ITO transducers that were measured experimentally. It is expected that the frequency will be decreased as the dimension of the transducer is increased (see section 2.4, equation 26).

Table 2: The sensitivity ($\delta R/\delta H$) at probe wavelength and frequency at pump wavelength are evaluated for ITO thin films with the increasing order of thicknesses that were measured experimentally. As the thickness of the thin films are presented by order.

Deposited ITO thin films on the tip of a single mode fibre			
Order of device	Thickness of samples (nm)	Sensitivity ($\delta R/\delta H$) (nm^{-1}) at $\lambda_{(\text{probe})}$ (nm)	frequency (GHz) at $\lambda_{(\text{pump})}$ 415 nm
First order	29-143-31	0.076 at 750	10.123
Second order	30-392-35	0.049 at 820	4.7760
Third order	32-602-35	0.037 at 810	3.4211
Deposited ITO thin films on plate transducers			
Order of device	Thickness of samples (nm)	Sensitivity ($\delta R/\delta H$) (nm^{-1}) at $\lambda_{(\text{probe})}$ (nm)	frequency (GHz) at $\lambda_{(\text{pump})}$ 415 nm
First order	26-150-30	0.083 at 813	9.8
Second order	29-360-30	0.082 at 825	6.03
Third order	29.9-561-30	0.030 at 780	4.988

The transducers with approximately 30-35nm of gold layers and higher order (i.e. thicker ITO layers) were fabricated either as the plate transducers or on the tip of single mode fibres. For the ITO transducers fabricated on the tip of a single mode fibre, the sensitivity in reflection case is decreasing from 0.076 to 0.049, and 0.037 (nm^{-1}) for the thickness of 144, 392, and 602nm, respectively. These ranges of sensitivity are for different probe wavelengths, as it can be seen in Table 2.

The mechanical model predicts the resonance frequency of the transducers depending on their layer thickness (see section 2.4). Figure 46 shows an example of the change of H_{ITO} with respect to time (a) for a third order ITO device (560nm). The transducer is in air and attached to a fused silica substrate. The Fourier transform shows a resonant frequency of approximately 3.4GHz. The high frequency contents (b) are from the motion of H_{Au} , that radiates waves into the transducer as well as glass substrate (about 18GHz) (see section 2.2). A more accurate way to measure the resonance frequency of the transducer is achieved by the long lasting wave since it provides a greater number of cycles (oscillations).

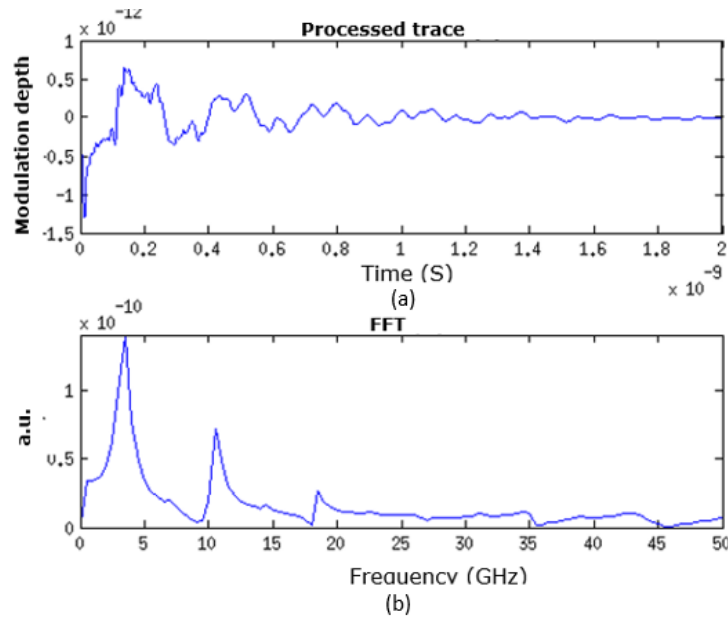


Figure 46: Simulation of the mechanical motion of a transducer with 29.9nm gold layers and 561nm ITO layer. Modulation depth (a) and its Fourier transform (b).

A thick (second order) device with 392nm thickness of ITO layer was also characterised using SEM and optical microscopy. Since the wavelength of the excited high frequency acoustic pulses in the thin film is in the nanometre range, the surface roughness should be also in the same order of magnitude. Therefore it is important to fabricate a thin film with constant thickness. The fibre-based transducers have a dimensions of sub-mm, deposited in a $4.4\mu\text{m}$ area (fibre core diameter), and it is essential to have a contamination-free and flat surface.

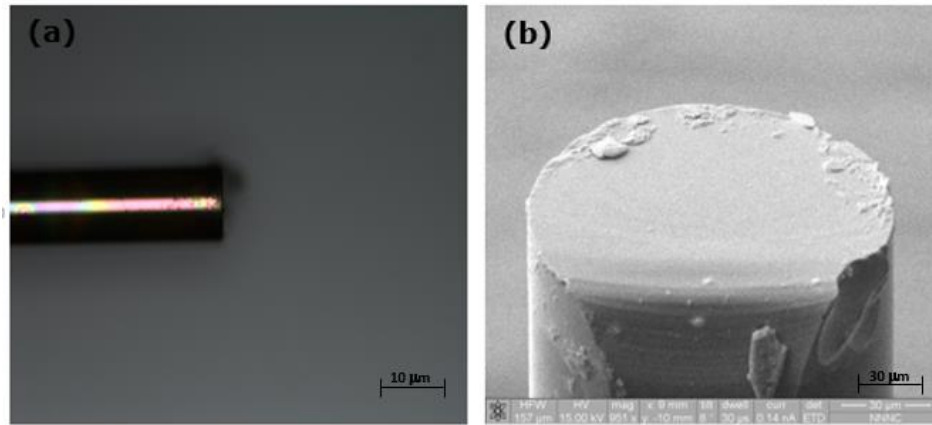


Figure 47: The images of a fabricated ITO transducer on the tip of a single mode fibre taken by (a) a light microscopy and (b) Scanning Electron Microscopy.

The images from SEM and optical microscopy are shown in Figure 47. It should be noted that all SEM measurements were done with gold metallic coating owing to the conductivity of the thin film at the magnification of $\times 951$ and 15kv accelerating voltage (the information are provided in each image). The optical microscopy image in Figure 47 (a) shows a flat surface while in the SEM image in Figure 47 (b) a broken edge can be seen on the tip of the optical fibre. Since the fibre has a cladding diameter of $125\mu\text{m} \pm 15$ with a core diameter of $4.4\mu\text{m}$, the broken edge will not cause any problem in terms of optical operation and mechanical response of the transducer. This is because the fabricated transducer on the top of fibre's core with $4.6\mu\text{m}$ diameter, is the area of interest where the pump and probe laser beams were delivered into the transducer.

5.2.2 Comparing the experimental and numerical traces

To validate those results, experiments were performed both in reflection and transmission. However, there is greater sensitivity to the optical resonance for the reflectivity case compared to the transmission case. The theoretical transmission of the devices is validated using optical measurements where the modelled resonance frequency is validated by variations in reflectivity (ΔR) of the transducer. This reflectivity is induced by the generated acoustic waves. The modelled response (ΔR) can be obtained by the sensitivity ($\delta R / \delta H$ (nm^{-1})) and the change in ITO thickness (δH_{ITO}) through the following equation:

$$\Delta R = \frac{\delta R}{\delta H} \delta H_{\text{ITO}} \quad (17)$$

The theoretical and experimental modulation depth can be compared directly if the modelled response is normalised by the static reflectivity value (DC) [49]. Three fabricated ITO plate transducers were characterised and tested in reflection. The transducers are from lower order (thinner) to higher order (thicker) with starting thickness of 150nm to 360nm and 561nm ITO layers. Before fabrication of the device, the optical model was applied to optimize the transducers' sensitivities by an accurate selection of the thicknesses of layers in a transducer. Figure 55(a) presents a schematic of a first order ITO device with gold layers of $\sim 30\text{nm}$ and ITO layer of 150nm thicknesses.

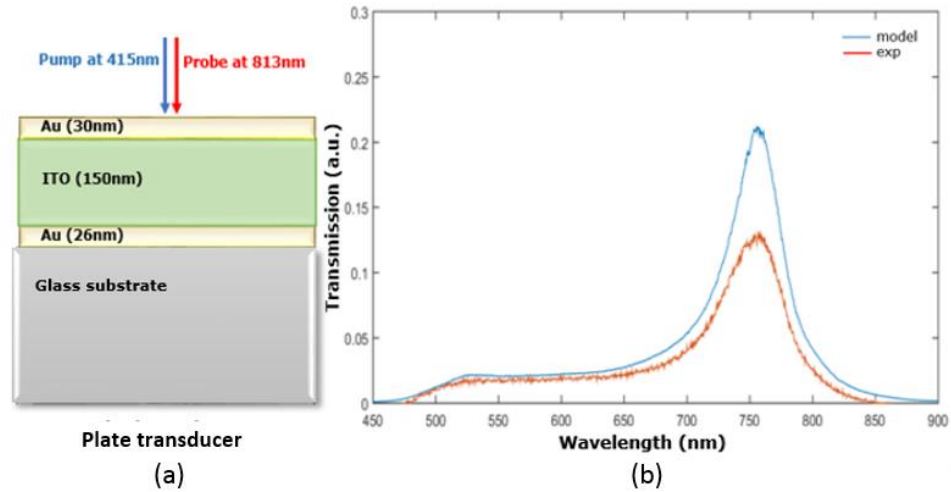


Figure 48: Schematic of (a) first order transducer with thickness of 150nm of ITO and ~30nm of gold layers. Transmittance comparison between model (blue) and experimental (red) traces for the ITO thickness of 150nm (b).

The Figure 48(b) also shows the corresponding results of thickness measurements using the transmitted spectrum of the theoretical trace (blue) and experimental measurements (red). As can be seen from Figure 48(b) experimental trace matches very well with its modelled trace. These values are used in the mechanical models to calculate the resonance frequency.

The frequency response of the thinner ITO device (first-order device) in reflection was measured using a free space experimental setup (Figure 31) and the results are shown in Figure 49. As can be seen there is a main frequency signal at ~9.5GHz associated with the mechanical resonance frequency of the transducer. From integrated results obtained from numerical model and experimental, it can be seen clearly there is a very good agreement between the modelled and measured variations in amplitude, despite of multiple source of errors such as variations in gold layer thickness at the top and bottom due to different sputtering cycles. This is another conformation of using numerical models to predict the performance of the transducers before testing them.

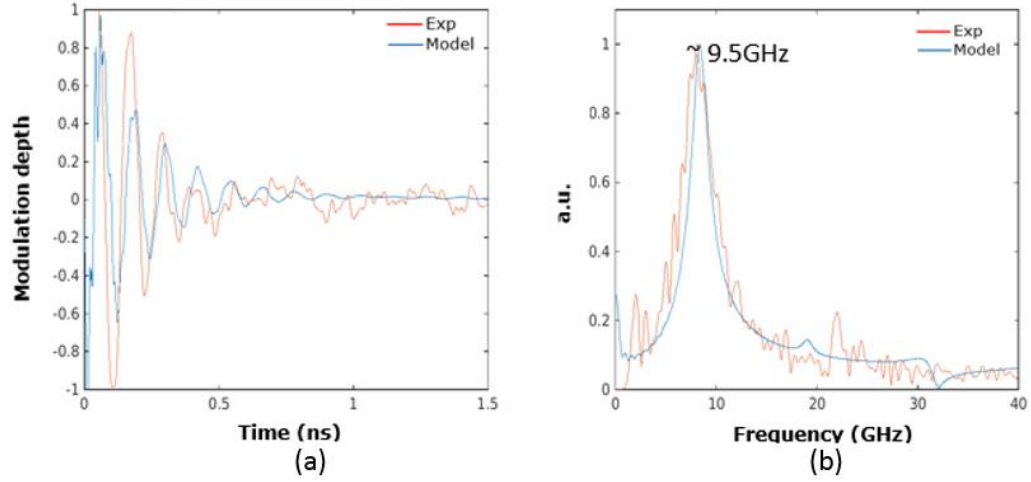


Figure 49: Comparison between experimental (red) and stimulated (blue, $\delta R/R$) modulation depth response of a first order transducer with thickness of 150nm of ITO and 30nm of gold layers to a single pump laser pulse. Temporal response (a) and corresponding Fourier transform (b).

Other high frequency signals are also presents which are due to radiation of waves from the glass substrate ($\sim 22\text{GHz}$) (see section 2.5) where this signal can be observed around 18GHz in modelled trace (blue). This possibly occurred due to variation in thickness of the gold layers in the transducer.

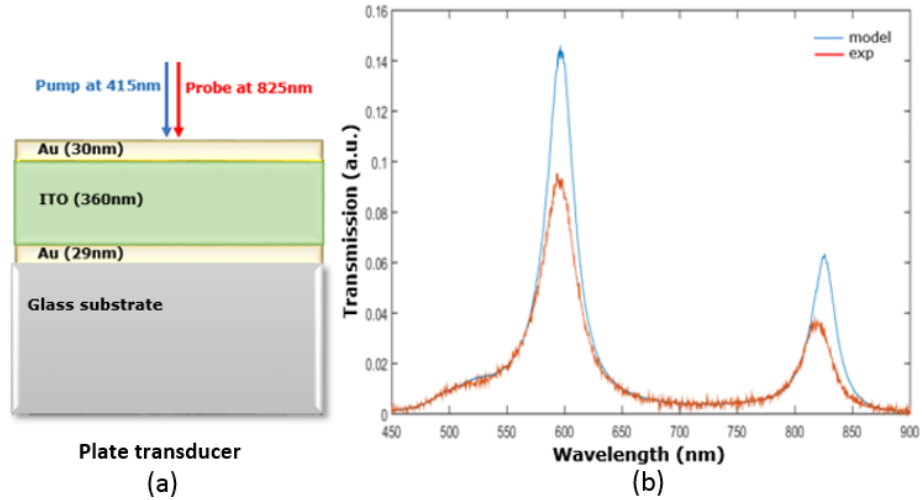


Figure 50: Schematic of (a) a first order transducer with thickness of 360nm of ITO and $\sim 30\text{nm}$ of gold layers. Transmittance comparison between model (blue) and experimental (red) traces (b).

Similar experiments were conducted for 360nm (second order) and 561nm (third order) thicknesses of ITO transducers (see Table 2). Each transducer was excited with the same energy of the pump laser beam and detection was performed in transmission. The results from the second order ITO plate transducer are also shown in Figure 50(a) with gold layers of ~ 30 nm thicknesses and ITO layer of 360nm thickness.

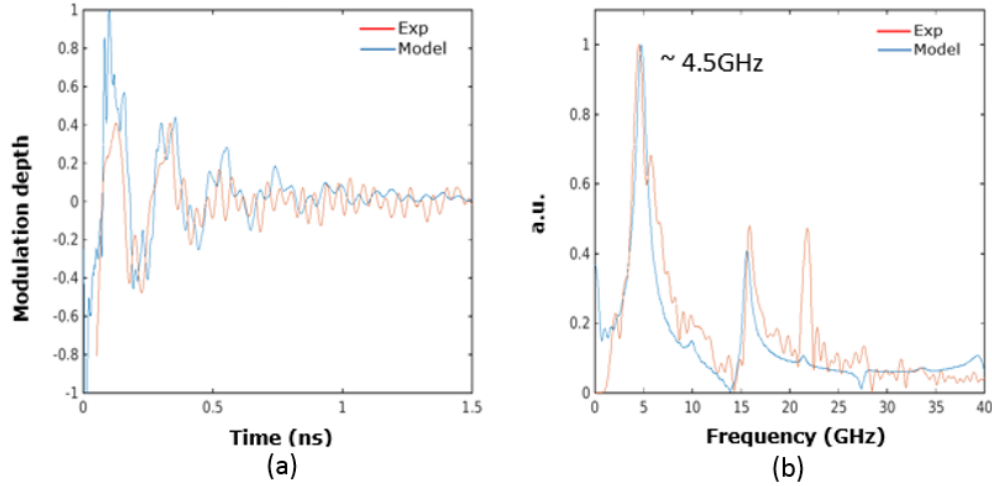


Figure 51: Comparison between experimental (red) and stimulated (blue, $\delta R/R$) modulation depth response of a second order transducer with layer thicknesses 39-360-30nm of gold-ITO-gold to a single pump laser pulse. Temporal response (a) and corresponding Fourier transform (b).

From the corresponding results of thickness measurements using the transmitted spectrum (Figure 51(a)), the experimental trace (red) matches very well with its modelled trace (blue). The frequency response of the second order ITO transducer is shown in Figure 51(b). There is a main frequency signal at ~ 4.5 GHz and a good agreement between the modelled and measured variations in amplitude.

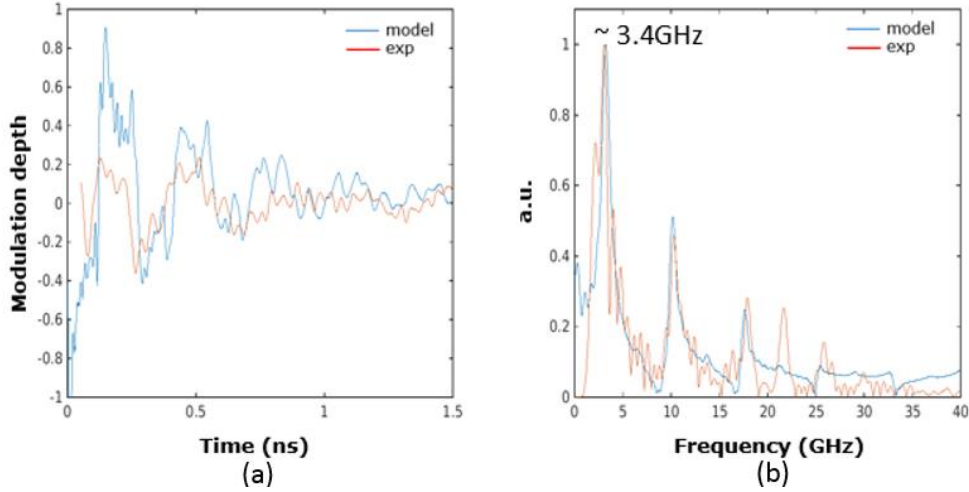


Figure 52: Comparison between experimental (red) and simulated (blue, $\delta R/R$) modulation depth response of a third order transducer with thickness of 561nm of ITO and 30nm of gold layers to a single pump pulse. Temporal response (a) and corresponding Fourier transform (b).

The same procedure was used for the third order device (thicker device) with the thickness of 561nm of ITO layer. The thickness measurement for this device was previously presented in Figure 45. The resultant frequency response of the third order ITO transducer is shown in Figure 52. There is a main frequency signal at ~ 4.5 GHz and the modelled data agreed with measured variations in amplitude. However, there are some difficulties in regards to detection of lower frequency using the initial laser system due to its pulse duration (see section 3.2, delay between two laser systems). It seems oscillation in the higher order transducers lasts longer than 12ns where the transducer is excited in 12ns time window. But still lower frequency in hundreds MHz range can be detected using the same system. Because of this reason in the modelled response in Figure 52(a), the experimental trace is slightly offset from the numerical output.

Figure 53 shows the evaluated transmitted spectrum of a first order ITO transducer fabricated on the tip of a single mode fibre with gold layers of ~ 30 nm and ITO layers of 143nm. The experimental trace fits well to 160nm thickness of the ITO layer with a high score in the scoring system (~ 1). The sensitivity of the devices ($\delta R/\delta H$ (nm^{-1}) for the reflectivity) is about 0.076 for the 750nm probe beam.

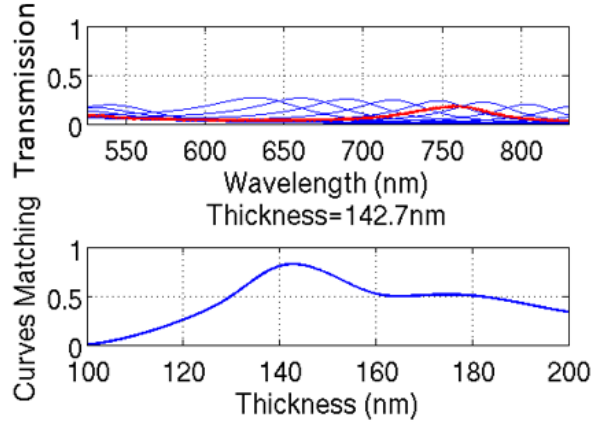


Figure 53: Example of transmittance comparison between model (blue) and experiment (red). The blue traces represent different ITO layers. The experimental trace fits well to $\sim 143\text{nm}$ layer of ITO fabricated on the tip of a single mode fibre.

The experimental traces for a first order ITO device ($\sim 144\text{nm}$ ITO layer and $\sim 30\text{nm}$ gold layers thicknesses) fabricated on the tip of a single mode fibre is shown in Figure 54 after truncations, thermal removal, filtering, and performing an FFT-based signal analysis measurement to evaluate the peak frequencies.

5.2.3 Brillouin detection from media in transmission

Detection of Brillouin oscillation from water was performed in transmission. The experiment offers the opportunity to investigate the propagation of acoustic waves into a medium generated by the transducer. However, it was expected to detect other Brillouin signals both from the glass substrate and the transducer. Water was selected as a medium since its properties is similar to the cell media.

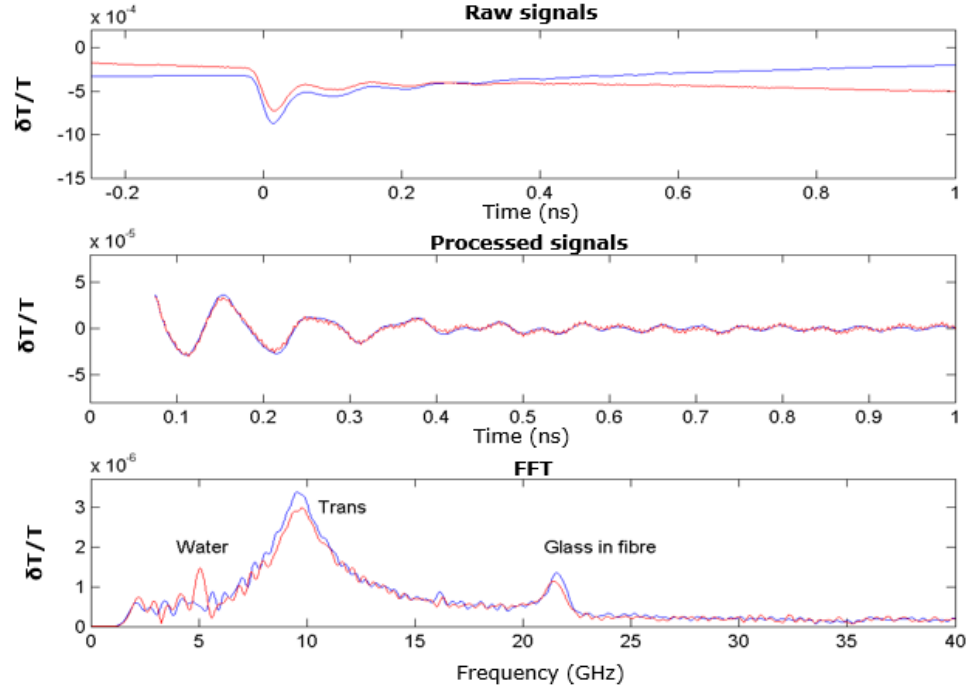


Figure 54: Comparison between experimental results tested in air (blue line) and in water (red line) for a first order fibre-based ITO transducer ($\sim 142\text{nm}$). The top graph shows unprocessed traces, the middle graph shows processed traces, and the lower graph shows Fourier transform of both traces. The detection was performed in transmission.

The blue line in Figure 54 presents the experimental trace performed in air and the red line is associated with the experimental trace performed in water. Both experiments were conducted with the same experimental setup shown in Figure 32 and were excited by the pump laser pulse with the same power (0.3 mW). There are signals at around ~ 10 and $\sim 22\text{GHz}$ in both experimental traces in air and water, which are associated with the resonance frequencies of the transducer and Brillouin scattering from the glass substrate (fibre core) (see section 2.4). Another signal around 5GHz can be only observed in the experimental trace performed in water. It is therefore concluded that a transition of the acoustic waves from at the transducer-medium (water) boundary is clearly observable. This transition is taken place due to different mechanical properties of the two materials which produce different speed of sound and refractive index in the materials. However, the result can be unlike by using different test mediums such as glycerol or biological tissue-mimicking liquid since different solutions present different mechanical properties. The great differences

between the two Brillouin frequencies of the two materials make the transition easy to observe.

5.2.4 Resonance frequency detection in reflection

Single-mode delivery of UV and IR wavelengths is desirable in many scientific applications in which a high quality beam is required. The single mode silica step index fibres are widely used at visible and IR regions where their availability is generally limited for applications in the UV region. A major cause of this insufficiency is the optical absorption of UV light in the pure silica core of the fibres due to high -OH content which leads to transmission degradation [80].

The dimensionless frequency or the V-number of an optical fibre is a parameter that can be considered to predict the behaviour of the single mode fibres (see section 2.5.1). The fibre is no longer single-mode below the cut-off wavelength, whereas it stays single-mode at any wavelengths above the cut-off wavelength. The cut-off wavelength can be lowered by reducing the numerical aperture (NA) at a fixed core dimension and vice versa. In principle, the short wavelengths below the cut-off of standard single mode step-index fibres can be also delivered which is dependent on laser power, measurement duration and operating conditions. Such effects were practically observed in this experiment in which the transmission reduced after about 1 hour due to optical damage of silica by UV absorption [61]. The damaged gratings was also created in the pure silica fibre core due to delivery of two laser beams at different wavelengths into the same single mode fibre. This contributes to a high loss of power of the laser beams (attenuation) [111].

A commercial single-mode fibre operating at 780 to 900 nm with cut-off wavelength of 730 nm with the core diameter of 4.6 μm was used in this experiment. The fibre is no longer a single mode fibre (un-real multi-mode fibre) at 415 nm of our pump laser beam as the core diameter is larger at this wavelength. It is referred to as few-mode fibres, supporting a small number of modes where the single-mode fibres support only one guided mode per polarization direction. This results in a non-linear effect such as cross-phase modulation by which a change occurs in the optical phase of a beam due to the interaction with another beam [112]. Since the

pump and probe laser beams at the particular wavelengths propagate through the same fibre, this phenomena can influence the phase change of a beam to the optical intensity of another beam. One of the difficulties associated with light propagation in multimode fibres is modal dispersion, but it is hard to resolve the effects of modal dispersion in its temporal domain since the fibre length is short in the experiments. However, the effects can be seen due to the inference of the modes in the ultrasonic signal-dependent noise (generated-speckle). The modal dispersion also limits the speed of adjacent pulses that can be delivered without overlapping at which an optical fibre can operate (see section 2.5) [61].

Because of the above reasons the response of the transducer was also assessed in reflection using an alternative experimental setup. A first order ITO (110nm) device (Figure 55 (a)) was fabricated on the tip of a single mode cable patch and tested in reflection mode. However, as shown in Figure 55 the transducer was excited from outside of the transducer while the detection performed in reflection mode through a 2:2 optical fibre coupler (see section 4.2, Figure 33). Figure 55(b) presents result of thickness measurements using the transmitted spectrum of a transducer with gold layers of 25nm thickness and ITO layer of 110nm thickness. As shown in Figure 55(b), the experimental trace (red line) matches well with the modelled trace (blue line). These values are used in the mechanical models to calculate the resonance frequency.

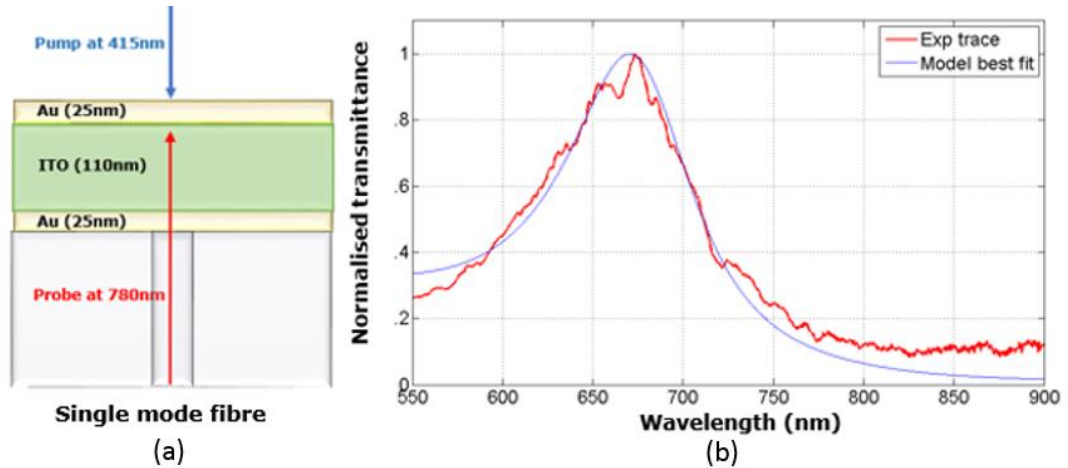


Figure 55: Schematic of a first order fibre-based transducer with thickness of 110nm of ITO and 25nm of gold layers (a). Transmittance comparison between model (blue) and experimental (red) traces for the ITO thickness of 110nm (b).

The device was tested in reflection and the results are shown in Figure 56. As can be seen there is a good agreement between the theoretical (blue) and experimental (red) traces of modulation depth and mechanical response of the for a third order ITO plate transducer with 25-110-25nm of gold-ITO-gold. There is a main frequency signal at $\sim 11\text{GHz}$ associated with the resonance frequency of the transducer. Other high frequency signals are also presented which are due to radiation of waves from the glass substrate ($\sim 22\text{GHz}$) and gold into the transducer.

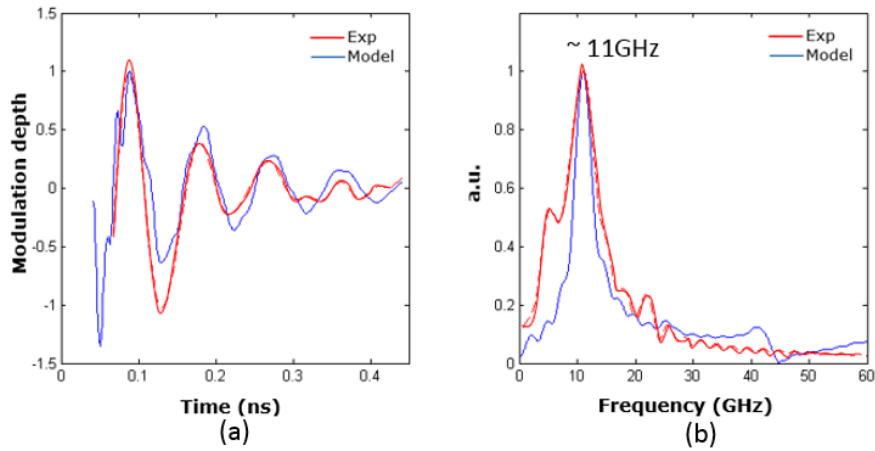


Figure 56: Comparison between experimental (red) and simulated (blue, $\delta R/R$) modulation depth response of a fibre-based transducer with thickness of 110nm of ITO and 25nm of gold layers to a single pump laser pulse. Temporal response (a) and corresponding Fourier transform (b). The detection was performed in reflection.

The response of the transducer was detected in reflection setups (see Figure 30). As expected based on the numerical model, the frequency decreased from $\sim 10\text{GHz}$ to $\sim 3\text{GHz}$ by increasing the size of the device from 144nm to 561nm of the ITO layer. So, all these devices behaved as expected from the numerical model considering multiple source of errors such as variations in the gold layer thickness due to different sputtering cycles. These results suggest that the resonance frequency of the transducers can be predicted accurately using numerical models.

5.3 Results from fabrication of Polystyrene polymer based transducer

The polystyrene-based transducers were fabricated as plate transducers and fibre-based transducers using spin coating and dip coating methods, respectively. The gold

coatings were deposited via a standard sputtering process. The PS thick films (>300nm) were deposited based on the process outlined in section 4.5.2. PS films were spun from toluene solutions with initial solution concentrations of PS ranging from 4% to 20% PS. The layers were spin coated on the top of coated with gold glass substrate and the second gold layer was coated on the top of PS thin films and the transmission spectra from thin films were collected using transmission spectroscopy setup. The thickness calculated as previously described in Chapter 3 and the average of PS film thicknesses as a function of spin speed are presented in Figure 57. Due to low sample size for each concentration, the error has not been calculated in this case.

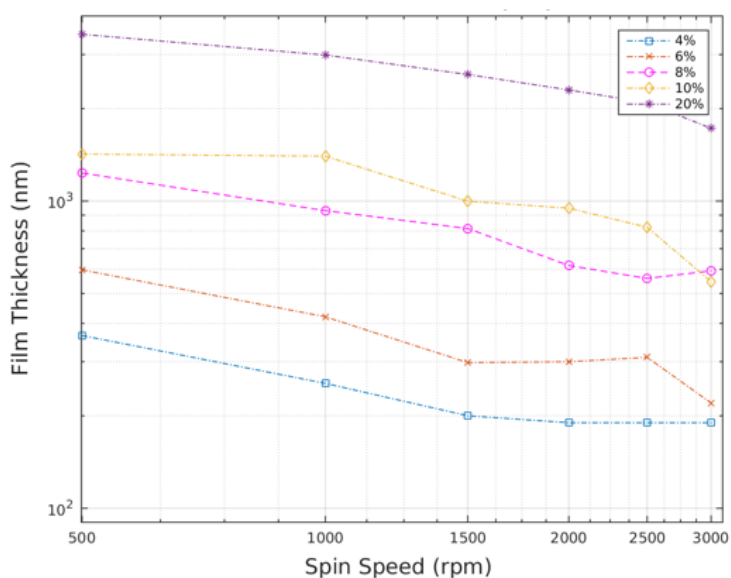


Figure 57: Film thickness as a function of spin speed and initial polymer solution concentration for PS in toluene: 4, 6, 8, 10, and 20 wt./vol (%). The thin films were deposited at 500, 1000, 1500, 2000, 2500, and 3000 rpm.

Film thicknesses ranged from 365nm to 1725nm for the PS films. Thicker thin films were obtained at high initial PS concentration ($\geq 10\%$), and the thicknesses decrease by increasing the spin speed (Figure 57). As expected (see section 2.8.2), by increasing the spin speed, the layer thickness decreased. It was also observed that the higher concentration PS solutions gave rise to thicker films. However, at lower initial PS concentration less than 10% there are some unexpected changes in thickness observed at 3000 rpm using 8% PS solution where it was expected to obtain

lower thickness, a thicker thin film was obtained. This is maybe because of human error during performing the procedure.

As previously discussed in Chapter 3, there is a correlation between film thickness and spin speed ($d \propto \omega^{-b}$). The value of the exponent "b" is dependent on evaporation rate of solvents in spin-coating fluids. The value of "b" is equal to 1 when the evaporation rate is too low. For solvents with constant evaporation rate, the value of "b" is -2/3 [95]. The solvent evaporation varies with the square root of the angular velocity ($\omega^{-1/2}$) [95] (see section 3.6.3). There is a correlation between the constant evaporation rates of solvents with film thickness as well as spin speed which is due to moving air about the substrate as the stage rotates. A higher rotation speed in spin coating, a faster evaporation of the solvents and thus a thinner film in the thickness will be obtained. However, in mixed solvents (acetone mixed with DMSO), the evaporation rate can vary since acetone has high evaporation rate and DMSO has lower evaporation rate. The thickness is also proportional to solution concentration, angular velocity and sample preparation. Any error in these parameters leads to unexpected results in fabrication. Also due to high evaporation rate of toluene it seems by using the spin coating method to deposit polymer solution is more suitable for fabrication of thicker ($\geq 1\mu\text{m}$) films, while the sputter coating method is mostly used for depositing thinner ($1\mu\text{m} \leq$) films.

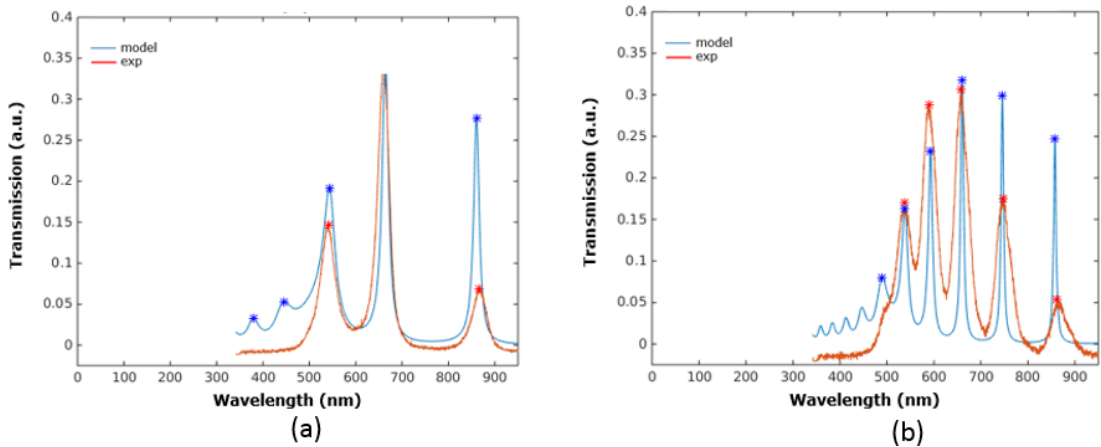


Figure 58: Transmittance comparison between model (blue) and experiment (red). The experimental trace fits well to a second and third order PS devices with thickness of 593nm (a) and 1234nm (b), respectively.

Figure 58 shows an example of thickness measurements of P-based thin films using transmitted spectrum of a transducer with gold layers of 30nm and PS layers of 593 (a) and 1234nm (blue curves) and experimental measurements (red curves). As can be seen, the experimental traces match with the theoretical output. More peaks appear in thicker transducers and the peaks are slightly shifted to the left (see section 2.3.2.2). In this case, the thickness measurement for a thicker device will be more challenging. In order to measure the thickness of higher order device, along with number of peaks other factors were also considered, such as the locations and the width of peaks.

5.3.1 Validation of optical and mechanical models for the plate transducers

Figure 59 shows the results of the optical model in terms of sensitivity for a transducer with wide range of gold (5-70nm) and PS (100-600nm) thicknesses. The sensitivity ($\delta R/\delta H$ (nm⁻¹)) for the reflected beam is presented and is measured for probe of 780nm. As can be seen the sensitivity for reflection case is still high even with increasing the thickness (0.080 nm⁻¹) for the 780nm of the probe where in the ITO case the sensitivity is decreased with increasing the thickness (see section 3.3).

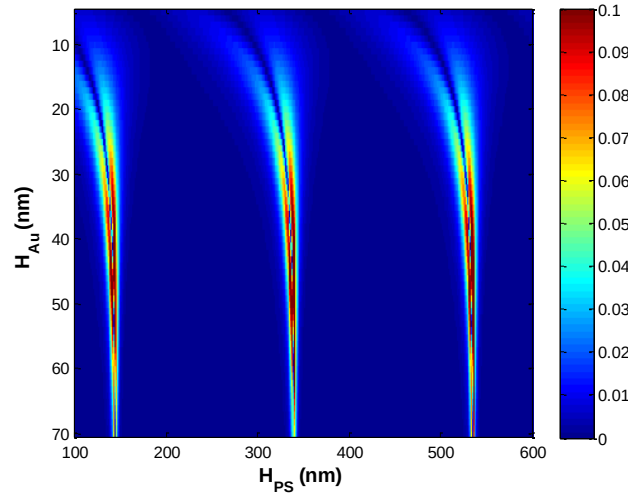


Figure 59: Transducer sensitivity $\delta R/\delta H$ at λ_{probe} (780 nm) for the PS transducer with a wide range of thicknesses from 100 to 700nm. The colour bar presents the sensitivity.

Also the acoustic frequency is lowering with increasing thickness of PS (see section 2.4, equation 28) from ~ 1.3 to 0.8 and 0.6GHz for the first order (140nm), second order(330nm), and third order (530nm) PS device, respectively. Whereas in ITO transducers (see section 5.2.1, Figure 2) the highest order device with 560nm thickness produced ~ 3.3 GHz frequency.

Two fabricated plate transducers with thicknesses of 560nm and 930nm of PS layers were validated optically and mechanically. The results from optical and mechanical validation of these two devices are shown in Table 3. As can be seen the sensitivity of the PS devices are 0.083 and 0.081 nm^{-1} for probe of 817nm and 789nm, respectively. The sensitivity is high and in the same range for both transducers for the given thickness (560nm and 931nm), whereas in ITO transducers case the sensitivity is decreased by increasing the thickness. It has been well documented that ITO shows a high optical transmittance in the UV region and it is found to be almost independent of film thicknesses. But the transmission in the NIR region decreases with increase in film thickness [80] due to the free carrier absorption. This phenomenon is common in all transparent conductors having high carrier concentration [81]. The frequency also is lowering with increasing thickness, from 550MHz to 430MHz for 560nm and 931nm thickness, respectively.

Table 3: The sensitivity ($\delta R/\delta H$) of the transducers at probe wavelength and frequency response at pump wavelength for the given thickness.

Plate PS transducers			
Order of device	Thickness of samples (nm)	Sensitivity ($\delta R/\delta H$) (nm^{-1}) at $\lambda_{\text{(probe)}}$ (nm)	frequency (GHz) at $\lambda_{\text{(pump)}}$ 415 nm
Third order	30-560-33	0.083 at 817	0.55
Higher order	32-931-33	0.081 at 789	0.43

The thickness measurement results of third and higher order PS devices are shown in Figure 60. As it can be seen there is a good agreement between the experimental trace and the theoretical trace for 560nm (second order) and 931nm (higher order) PS transducers. The evaluated sensitivity in reflection mode is 0.050 (nm^{-1}) for the probe at 822nm of a second order PS device whereas for a higher order device the sensitivity is 0.049 nm^{-1} for the probe at 792nm.

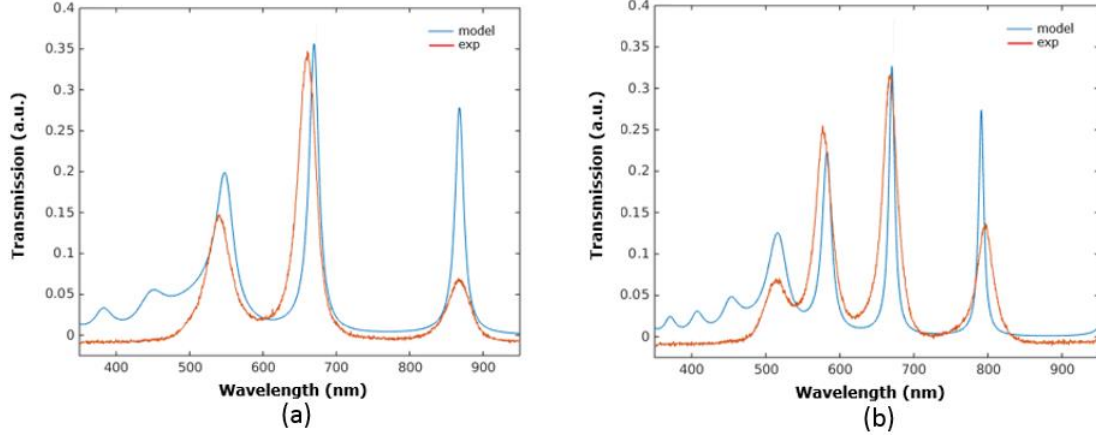


Figure 60: Transmittance comparison between model (blue) and experiment (red). The experimental trace fits well to a third and fourth order PS devices with thickness of 560nm (a) and 931nm (b), respectively.

5.3.2 Comparing the experimental and numerical traces for the plate transducers

The thicker (third-order) PS device with 560nm thickness was tested in reflection mode in the free-space experimental setup (Figure 31). Figure 61 shows the comparison between the theoretical (blue line) and the (red line) traces of the modulation depth and mechanical response of the device. There is a main frequency signal at $\sim 0.55\text{GHz}$ associated with resonance frequency of the transducer. There is an agreement between the modelled and measured variations in amplitude in frequency response where the variation in reflection is slightly offset in comparison with the theoretical trace.

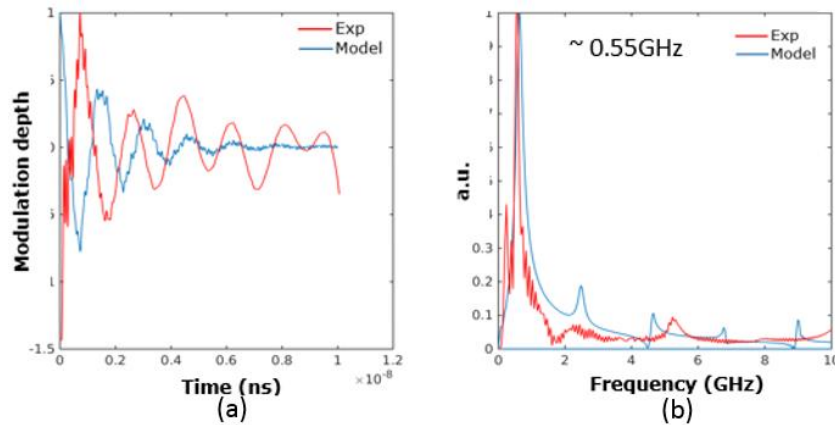


Figure 61: Comparison between experimental (red) and simulated (blue, $\delta R/R$) modulation depth response of a second order transducer with 560nm thickness of polystyrene and 30nm of gold layers. Temporal response (a) and corresponding Fourier transform (b).

As was previously discussed, there are some difficulties in regards to the detection of lower frequency in MHz range ($\geq 800\text{MHz}$) using the initial laser system due to its pulse duration. It seems that oscillations in the higher order transducers lasts longer than 12ns where the transducer is excited in this 12ns time window (see section 2.1.3). It is due to the viscoelasticity behaviour of polymers. Viscoelastic materials exhibit characteristics of a viscous liquid as well as of an elastic solid. When a polymeric material is exposed to a stress, it is deformed and can return to its original shape due to elastic behaviour of the polymer, but this is slow and it can coincide with the next cycle of vibration [96].

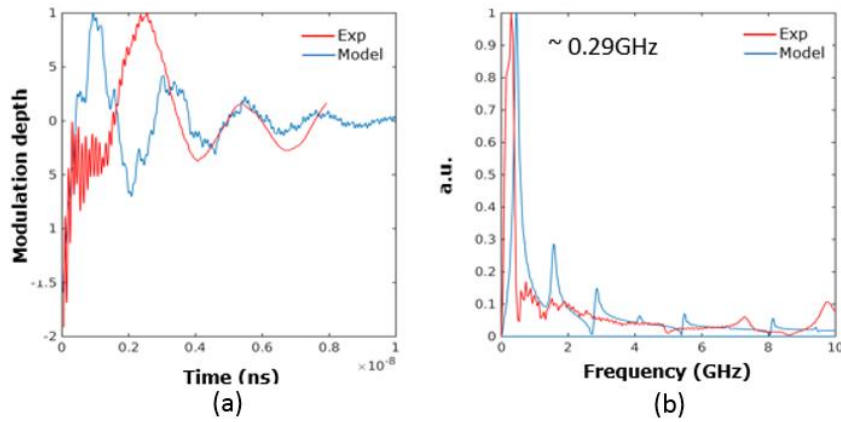


Figure 62: Comparison between experimental (red) and simulated (blue, $\delta R/R$) modulation depth response of a third order transducer with thickness of 931nm of polystyrene and 31nm of gold layers to a single pump laser pulse. Temporal response (a) and corresponding Fourier transform (b).

In this case, the pulse duration is increased to more than 12ns in the mechanical model. Extending the laser pulse duration depends on the duration of signal oscillations. The modulation depth ($\delta R/R$) trace was then evaluated for the extended oscillation time and the trace was divided into the equal parts. The frequency response was calculated from the averaged modulation depth ($\delta R/R$) trace. The experimental trace was then compared with the average of theoretical modulation depth. Also in the higher order PS device with 931nm thickness a main frequency signal at $\sim 0.29\text{GHz}$ is observed, which is associated with the resonance frequency of the transducer (Figure 62). Despite the agreement of intensity-frequency profile, the variation in reflection is offset in time in comparison with theoretical trace.

These results suggest that, as it is expected, by changing the material from ITO to PS, as well as increasing the film thickness, the frequency decreases. This is due to lower Young's modulus (E) and bulk modulus (B) of the polymers (see section 2.2, equations 28 and 29). The materials with lower E and B are counted as viscoelastic materials and they exhibit a resistance against wave propagation [97]. The material's density is another parameter which affects the frequency, amplitude and speed of waves in materials.

5.3.3 Comparing the experimental and numerical traces for fibre-based transducers

As was previously mentioned, the PS devices were also fabricated on the tip of single mode fibres. The immersion time parameter in the dip coating method was applied to increase the thickness on the tip of optical fibre. In this experiment an 8% PS solution was used to fabricate six transducers with different thicknesses. The experiment was repeated twice (batch 1 and batch 2).

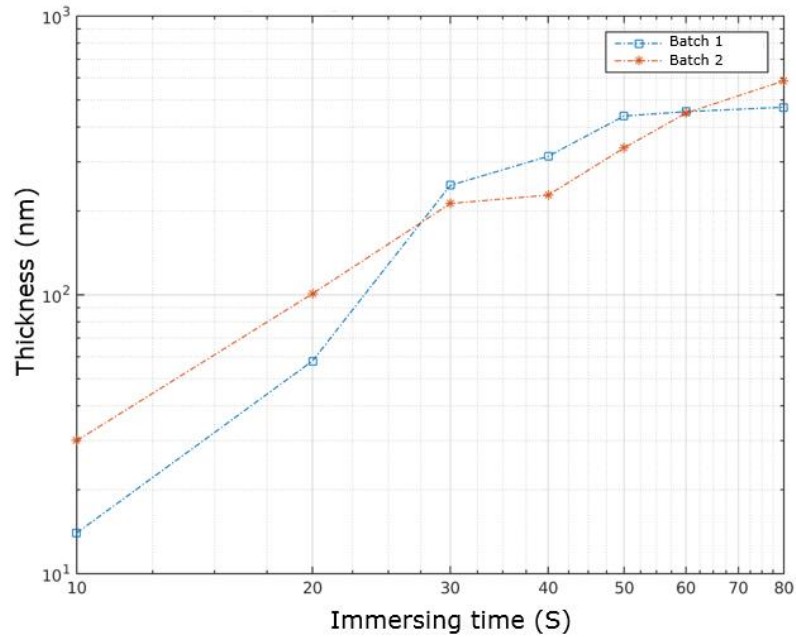


Figure 63: Film thickness as a function of immersion time for initial polystyrene polymer solution concentration for of 8wt./vol (%) in batch1 and batch2. The thin films were deposited in increasing order of immersion time from 10 to 80 seconds.

Film thicknesses ranged from 14.5nm (in batch 1) to 585nm (in batch 2) for PS-based films with initial PS concentration of 8%. The immersion time was in increasing order from 10 to 80 seconds across both batches. The thickness of each sample was calculated and the results are presented in Figure 63 as a function of immersion time. As can be seen, thicker thin films are obtained with longer immersion time and the thickness is increased exponentially by increasing the immersing time (see section 4.7). Immersion time is an effective parameter for interaction of precursors in solution with the substrate surface [98]. However there are some unexpected changes in thickness of batch1 observed at 50, 60, and 80 second of immersion time, where it was expected to increase the film thickness by increasing the immersion time. It was maybe due to human error in conducting experiment or evaporation rate of the solvents at different time points. A thicker fibre-based transducer with a 585nm PS layer thickness was validated optically and mechanically. The results from the optical and mechanical validation of the device are shown in Table 4. For the 585nm PS device the optical sensitivity is 0.086 for the probe at 847nm and a frequency of 0.55GHz with amplitude of 1.23×10^{-6} can be produced by the device.

Table 4: The sensitivity ($\delta R/\delta H$) of the transducer at probe wavelength and frequency response at pump wavelength for the give thickness. The initial PS concentration is 8% in this case.

Fabricated PS device on the tip of a single mode fibre			
Immersing time (S)	Thickness of samples (nm)	Sensitivity (nm^{-1}) ($\delta R/\delta H$) at $\lambda_{\text{(probe)}}$ (nm)	frequency (GHz) at $\lambda_{\text{(pump)}}$ 415 nm
70	26-585-30	0.086 at 847	0.44

Figure 64(a) presents a transducer fabricated on the tip of a single mode cable and tested in reflection mode. The transducer was excited externally while the detection was performed through the fibre (Figure 33). Figure 64(b) presents result of thickness measurements using the transmitted spectrum of a transducer with gold layers of $\sim 30\text{nm}$ and PS layer of 585nm (blue curves) and the experimental measurements (red lines). The experimental trace shows a poor fit to its modelled trace. It may be due to the variation of thickness of gold layers at the top and bottom of optical cavity where it is assumed the thickness of the gold layers are the same in the optical model. These values are used in the mechanical models to calculate the resonance frequency.

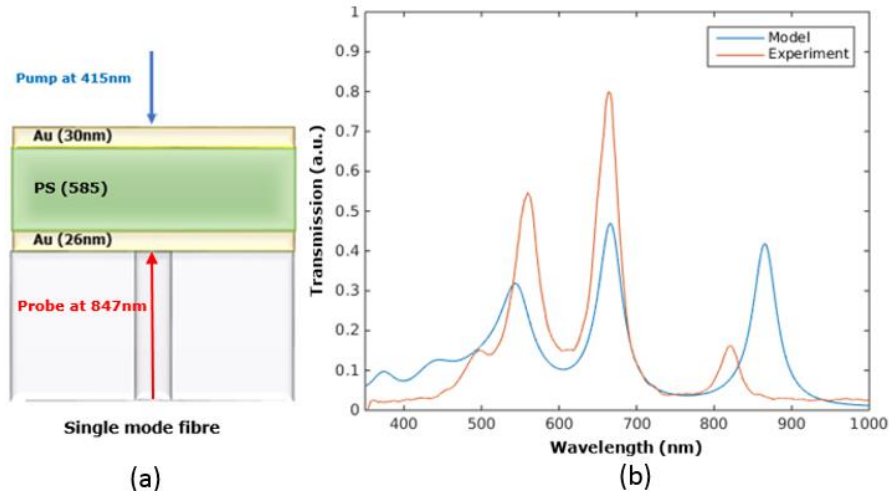


Figure 64: Schematic of a second order fibre-based transducer with thickness of 585nm of polystyrene and ~ 30 nm of gold layers (a). Transmittance comparison between model (blue line) and experimental (red line) traces for the polystyrene thickness of 585nm are presented (b). The detection was performed in reflection.

The device was tested in reflection mode in a fibre-based experimental setup (Figure 33). Figure 65 shows the comparison between theoretical (red) and experimental (blue) traces of modulation depth and mechanical response of the device. The measured variation in amplitude is slightly offset in time in comparison with the theoretical trace but a good match between the experimental frequency peak and the theoretical peak. The reason was explained in previous section (section 5.3.2).

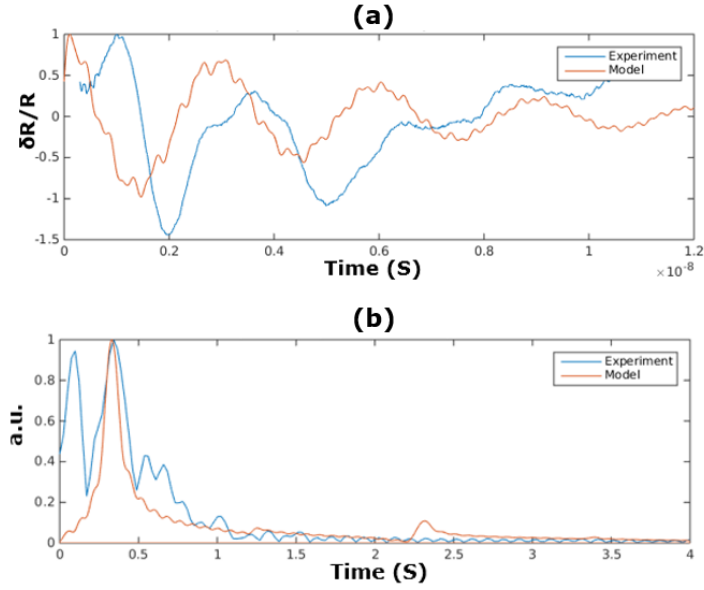


Figure 65: Comparison between experimental (red) and simulated (blue, $\delta R/R$) modulation depth response of a second order fibre-based transducer with thickness of 585nm of polystyrene and 30nm of gold layers to a single pump laser pulse at the wavelength of 415nm. Temporal response (a) and corresponding Fourier transform (b) are presented.

There is a main frequency signal at ~ 0.4 GHz associated with resonance frequency of the transducer. There is also a lower frequency peak before the main peak at 0.4GHz. There are a couple of cycles of oscillations before the oscillations have died out completely and it seems the transducer is still ringing when the next pulse excites the transducer. The experimental traces are compared with the average of the theoretical trace as it explained in section 4.3.2.

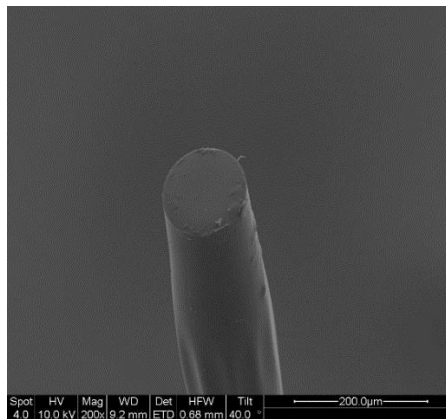


Figure 66: The SEM image of a single mode fibre device coated with PS (930 nm thickness).

The SEM image of a PS transducer fabricated on the tip of a single-mode fibre is shown in Figure 66. The image was taken in order to observe whether a plane flat transducer has been fabricated on the tip of fibre. It should be noted that the SEM measurements were done with gold metallic coating owing to the conductivity of the thin film at 10kV acceleration voltage and 200x magnification. The image shows a flat surface free of contamination without any broken edge and it seems the PS medium covers the tip.

5.4 Results from polymer and dye characterization

At this point, another polymer such as CA was tested to find the best mixture of the APC dye dissolved in a polymer either using a solvent or a mixture of solvents suitable to dissolve both of them.

5.4.1 Solvent selection for both polymer and dye

In this part of study, the compatibility of different solvents to dissolve both host polymer (i.e. CA) and APC dye as a guest substance was investigated. Also the compatibility of a solvent to mix CA polymer with APC dye was investigated. According to the results, the dye was dissolved in DMSO and no undissolved materials were observed in solution with naked eyes.

The plotted spectra from the dissolved dye in DMSO showed a high absorption at ~740 nm in its dried state (Figure 67(a)). The experimental spectra from the dissolved dye in DMSO and acetone are only presented here in Figure 67. The spectra was collected in wet state in order to examine whether the absorption peak around the desired wavelength of 740nm shifts after drying.

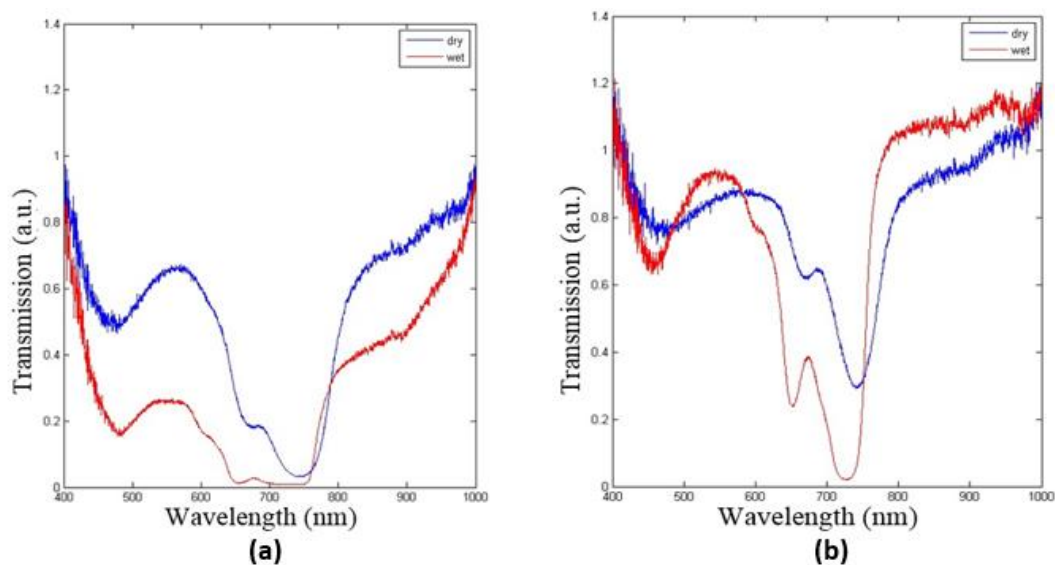


Figure 67: Example normalised transmission spectra for dye dissolved in (a) DMSO and (b) acetone solvents in both dry and wet states.

Based on the results obtained for the solubility of APC dye in different solvents it was concluded that the solubility of the dye in used solvents is decreased in the following order:

DMSO > Acetone > Chloroform > Ethanol > Toluene > water

Following the conduction of a series of experiments, acetone and DMSO were considered as the most suitable solvents for CA, the APC dye showed limited solubility in acetone. To dissolve CA polymer, a mixture of DMSO and acetone solvents was also considered. The APC dye was successfully mixed with CA polymer since Cellulose acetate is a sugar-based polymer and a suitable material for the supramolecular attachment of dyes due to the large number of polar substituent which enable the formation of dipolar interactions with an adequate substance [99].

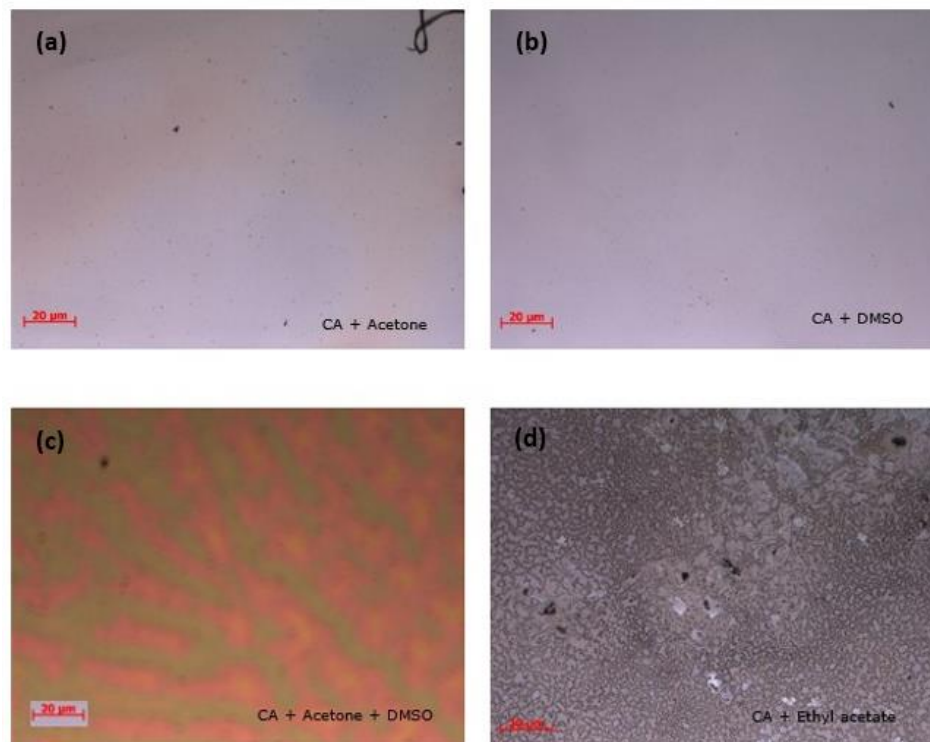


Figure 68: Light microscopy images of the CA-based thin films dissolved in different solvents.

Figure 68 images show smooth thin films prepared from dissolving CA in DMSO (a) and acetone (b) where a wavy pattern can be seen in a thin film prepared by dissolving CA in a mixture of acetone and DMSO (c). A thin film with fractured pattern (Figure 68 (d)) was obtained from dissolving CA in ethyl acetate that shows this solvent is not suitable to dissolve the CA. Based on the results it is suggested that acetone and DMSO can be used either in a mixture form of the solvents or single solvents for CA.

5.4.2 Compatibility of dye on cellulose acetate

The image in Figure 69 shows a dye-CA based thin film with a smooth texture and some undissolved APC dye pieces in the film. Based on the results obtained from the investigation of dye-CA thin films, it is concluded that the APC dye can be mixed with CA polymer properly.

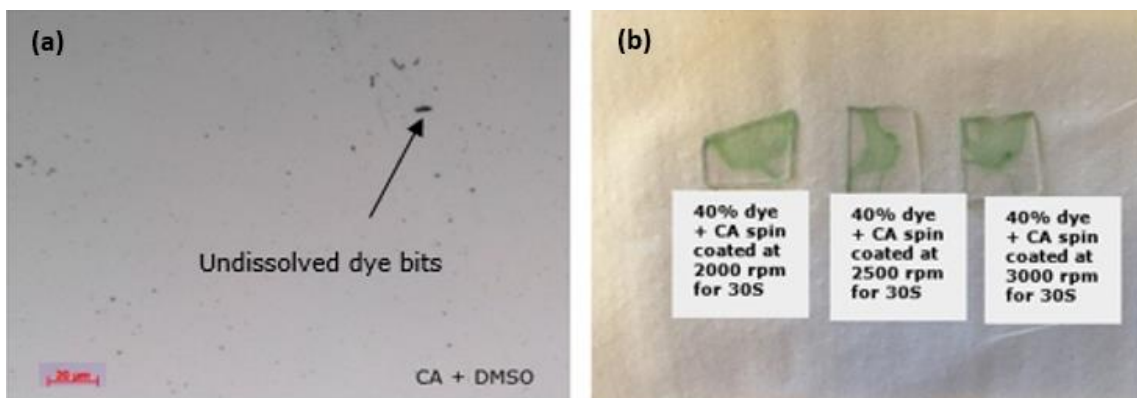


Figure 69: Light microscopy image of a CA-based thin film dissolved in DMSO (a) and prepared samples (b).

The spectra show an absorption band about 730 nm without any shift to the lower wavelength or higher wavelength both in wet and dried samples, however a decrease in intensity of transmitted spectra can be seen around 730nm in dried state of samples. The results showed that the APC dye and CA polymer are compatible in a mixture and there is no shift in spectra after evaporation of solvent. In addition, the results showed that the morphological texture of a dye-CA based thin film is a stable mixture.

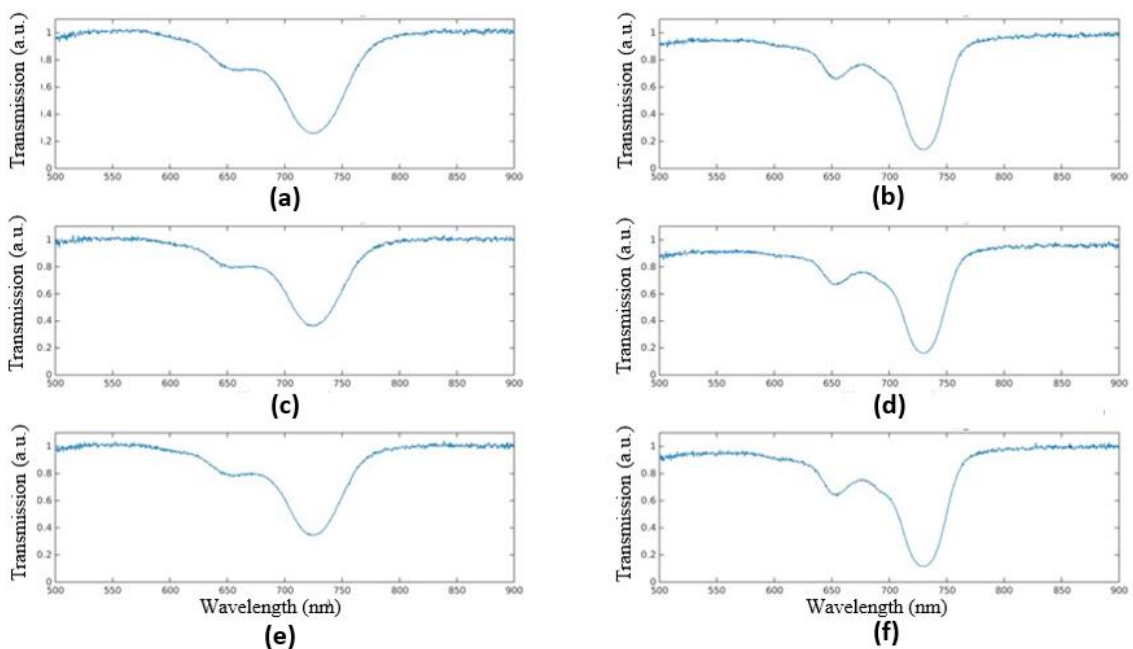


Figure 70: Normalized transmission spectra of dye-CA thin films dissolved in DMSO. The thin films were deposited at the speed of (a, dry state) (b, wet state) 2000, (c, dry) (d, wet state) 2500, (e, dry state) (f, wet state) 3000.

5.4.3 Thickness analysis of thin films using different concentration

Table 5 presents the thicknesses of CA-based thin films prepared by dissolving CA in different concentrations of DMSO and acetone, individually. Unfortunately, the data from thicker films could not be collected as they were too thick to be measured by ellipsometry. Also, it was noted that working with 50% concentration of CA solution is challenging using the spin coating. This is due to acetone's high evaporation rate resulting in a viscous solution within only 2-3 mins. On the other hand, to fabricate a thicker CA-based film using DMSO as a single solvent is difficult to use the spin coating technique. This is because in high concentration at 3000 rpm even after 1 minute spin coating the coated polymer on the glass substrate tends to come to the centre of sample rather than staying in stationary state due to surface tension of DMSO. The results indicate that polymeric solutions prepared in acetone yielded films thicker than those prepared in DMSO. As mentioned previously, spectroscopic ellipsometry technique is typically used for films whose thickness ranges from sub-nanometres to a few microns [92].

Table 5: The collected data from CA-based thin films using a spectroscopic ellipsometer.

Solvents	Thickness at 10% concentration (nm)	Thickness at 20% concentration (nm)	Thickness at 30% concentration (nm)	Thickness at 40% concentration (nm)	Thickness at 50% concentration (nm)
Acetone	50	103	130	-	-
DMSO	22	30	80	-	-

The changes observed in the surface roughness suggest that variation in thickness increases with increasing thickness which makes it difficult to measure the thickness of thin films properly. Therefore, it can be concluded that increasing concentration is not an appropriate way to increase thickness; rather, to obtain a uniform thickness, it is needed to find the optimal concentration and modify other factors such as solvents to achieve the desirable thickness.

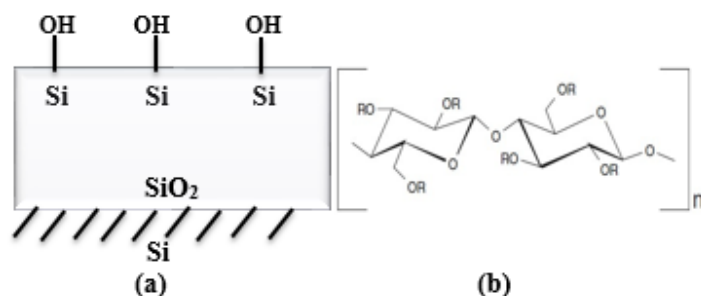


Figure 71: Schematic (a) (b) representation of chemical structures of (a) substrate and (b) cellulose ester, where R refers to the acyl groups, with COCH_3 . The figure was adapted and modified from Amim et al. [103].

Cellulose esters are rich in polar groups such as hydroxyl and carbonyl groups, which are expected to form hydrogen bonding with silanol groups on the Si in glass substrate. In the spin coating process, the solvent flows toward the edges of the substrate and then evaporates. The transition from thinning to evaporation is rapid [100]. Since acetone with the higher vapour pressure than DMSO, it evaporates faster and the film produced from acetone remains thicker than in the case of DMSO [101]. When the solvent evaporates slowly, the polymer solution resides longer on the solid substrate, allowing more uniform liquid film formation prior to evaporation. Undoubtedly the evaporation rate of the solvent plays a very important role on the spin-coated film uniformity. Nevertheless, the interactions balance in the systems formed by substrate, polymer and solvent cannot be neglected [102].

5.4.4 Effect of thickness of dye-polymer films on transmission spectra

It is expected that by increasing angular velocities, the film thickness will decrease. The Figure 72 shows normalized absorption spectra measured for films with different thickness (decreasing order). Each spectrum shows an absorption onset at about 730nm without any shift. It was concluded that the absorption is decreased by decreasing the film thickness.

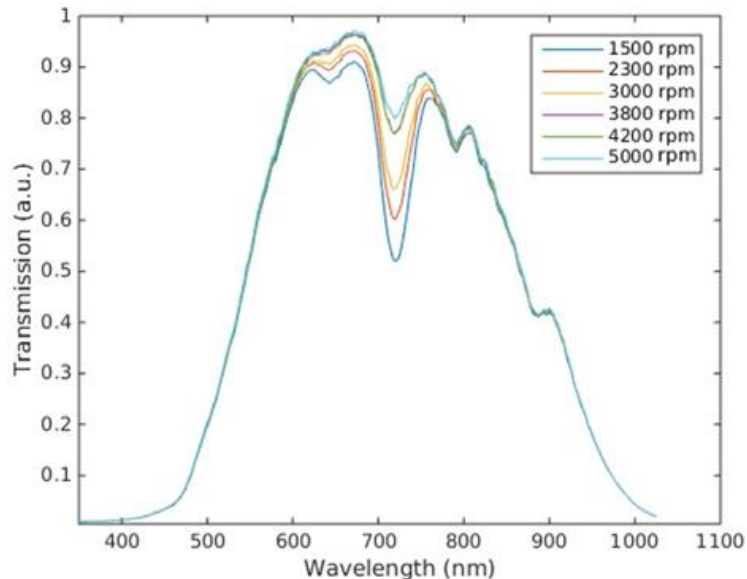


Figure 72: The normalised transmission spectra of APC dye-CA polymer thin films using a modified solvent (acetone and DMSO). The spectra were normalised by colourless CA control thin films.

5.4.5 Thickness dependent optical constants of dye-polymer thin films

In order to measure the optical constants such as the absorption coefficient (α) and the extinction coefficient (κ) of dye-polymer thin films, the measured values from experiments are used such as film thicknesses, and the transmission spectra from both from reference (uncoloured thin films) and sample. This can be accomplished by measuring the Reflectance (R) and/or Transmittance (T), of the film; solving Beer-Lambert-Bouguer [104] equations can give the optical constants (see Appendix A.1). In order to determine the optical properties, 4 curves are built up: raw spectra, normalised spectra, absorption coefficient (α) vs. wavelength and extinction coefficient (κ) vs. wavelength [105] [106]. In this part of study the optical constant α and κ (nm^{-1}) of different concentrations of the APC dye (10, 15, and 20%) at 30% CA solution were calculated through the transmitted spectra of thin films deposited at different spin speed. The samples from previous experiments (section 5.4.4) were used in this section to calculate the optical constants of different dye concentrations in CA thin films. The thickness of each sample is shown in Table 6.

Table 6: The thickness of thin films spin coated at different spin speeds (rpm) using 30% CA solution with different concentrations of the APC dye. In this case the CA was dissolved in a mixture of DMSO and acetone where the dye was only dissolved in DMSO.

Dye concentration (%) @ 30% CA solution	Thickness at 1500 rpm (nm)	Thickness at 2500 rpm (nm)	Thickness at 3000 rpm (nm)
20	845	476.5	380
15	745.3	464.8	380.3
10	840	456	370

As shown in Table 6, the thicknesses of dye-CA thin films are decreased by increasing the spin speed as it expected. However, a fluctuation in the thicknesses of thin films can be seen at the spin speed of 1500 rpm where this fluctuation improves at the spin speeds of 2500 rpm and 3000 rpm. Based on the results from the previous experiment (see section 5.4.3, Table 5) using a lower spin speed is not a recommended method to increase the thickness using CA solutions with higher concentrations. For this reason, in the next experiments higher spin speed and CA solutions with a lower concentration were used to deposit thicker films by sputter assisted spin-coating method. Note, the gold layers were deposited in order to calculate the thicknesses of dye-CA thin film.

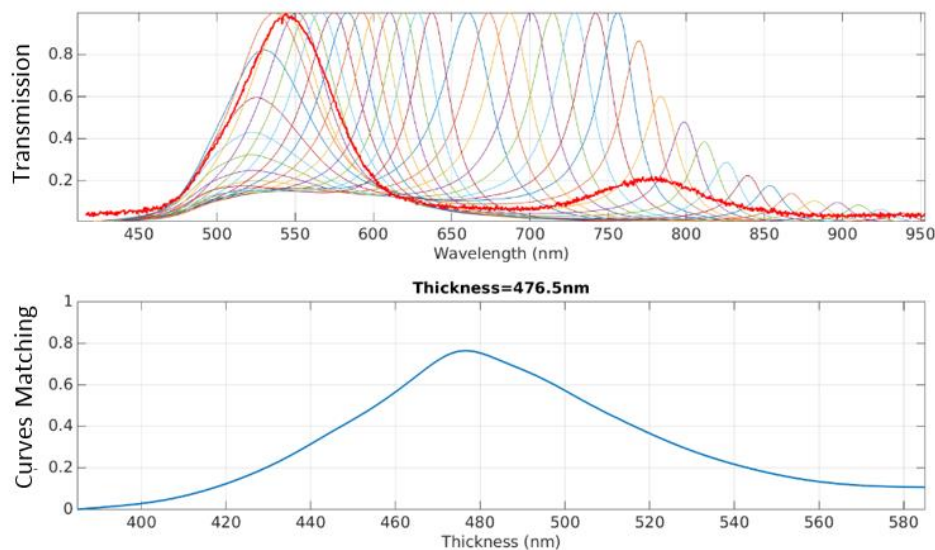


Figure 73: Example of transmittance comparison between model and experiment (bold red in top graph). The top graph presents the model spectra of different CA layers of different thicknesses and the lower graph presents the highest score related to the thickness of CA layer. The experimental trace fits to a 476.5nm CA layer with 24 nm thicknesses of gold layers.

Figure 73 shows the evaluated transmitted spectrum of a transducer with gold layers of about 24nm thicknesses, CA layers of different thickness and an experimental measurement (red bold line curve). The experimental trace of a CA-based thin film fits well to a 476.5 nm thickness. This value and other calculated thicknesses were used to calculate the optical constants (i.e. α and κ) of different dye concentrations.

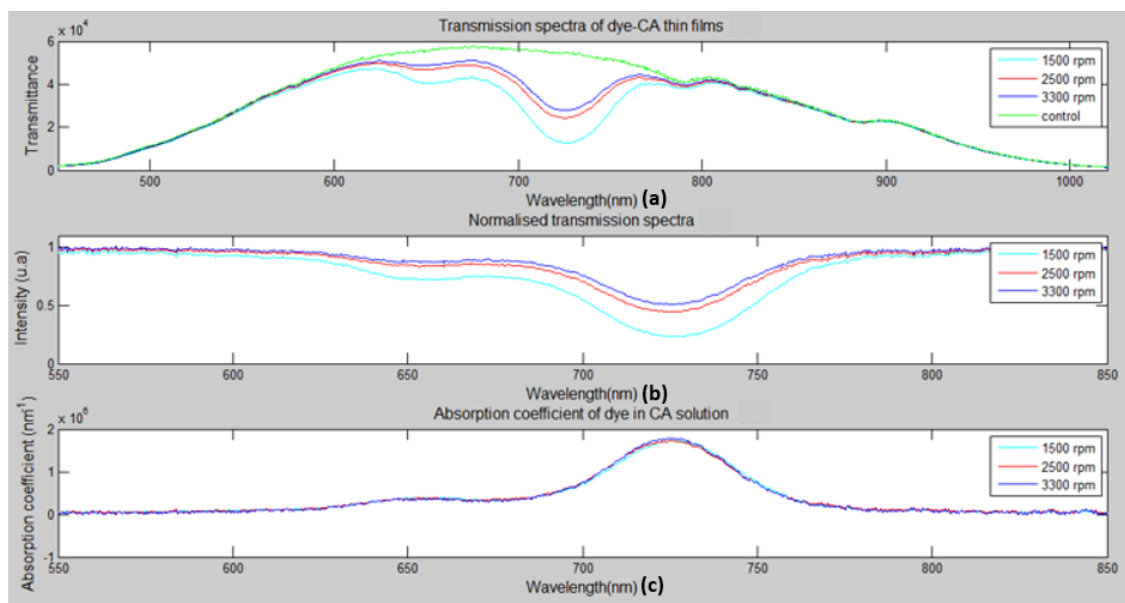


Figure 74: Transmission and Absorption spectra versus the wavelength (nm) of dye-CA thin film with thicknesses of 845 (cyan line), 460 (red line), and 380 nm (blue line): (a) the transmission spectra of dye-CA thin films, (b) normalized transmission spectra, (c) the absorption coefficient (α (nm⁻¹)). The concentration of dye in CA medium is 20%

Figure 74 shows spectral distributions of the transmittance (T) of the dye in CA thin films with concentration 20% dye in 30% CA solution. Figure 74 (a) shows typical transmittance spectra of three CA thin films of thickness of 845, 460, and 380nm. It can be observed that, with decreasing thicknesses of the samples, their transmission is increased. The cyan curve is associated with a thicker film (845 nm) whereas the red and blue curves are associated with thinner films with thicknesses of 476.5 and 380 nm, respectively. These graphs indicate that a peak at ~ 726 nm has been created in the visible region after adding the APC dye in the CA polymer.

The optical absorption coefficient of dye-CA thin films with the thicknesses of 845, 746.5, and 380 nm were evaluated from the transmittance data and are presented in

Figure 74 (b). The normalised absorption spectra measured for the thin films with mentioned thicknesses is shown in Figure 74(b). Each spectrum shows an absorption onset at 726 nm. Figure 74 shows the absorption coefficient (nm^{-1}) of 20% dye concentration in thin films with the mentioned thicknesses. It is evident that with increasing dye concentration leads to significant increase in the value of α . It can be explained by the fact that with higher concentration of the dye in CA polymer, more molecules are deposited in thin films.

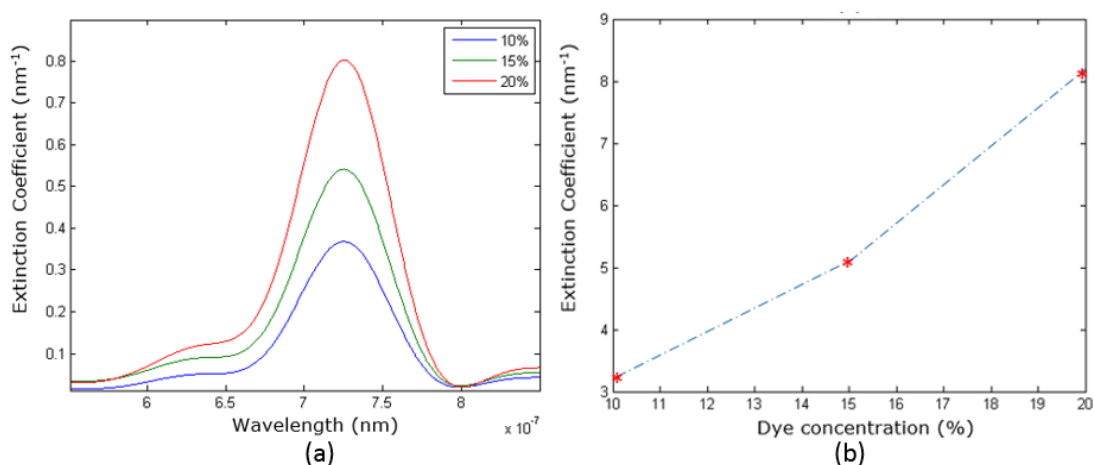


Figure 75: The evaluated extinction coefficients (a) and the graph of the extinction coefficient as a function of dye concentration (b). In this case three different concentrations of the APC dye, 10, 15, and 20% used in 30% CA polymer solution.

The calculated absorption coefficient values for different APC dye concentrations were used to evaluate the extinction coefficient values of corresponding dye concentrations (see Appendix, A.1). As shown in Figure 75 (a), the extinction coefficient begins to rise at $\sim 600\text{nm}$ wavelength with increasing dye concentration in the CA-based thin films and starts to decrease at NIR region ($\sim 800\text{nm}$). The variation of the extinction coefficients as a function of different dye concentrations was plotted in Figure 75 (b).

In this experimental section the effect of APC dye concentration, thickness and optical properties of dye-CA thin films were investigated. Using the spectra, the optical constants κ and α were evaluated. The evaluated extinction coefficients for a wide range of wavelengths were collected and used in the optical model.

5.4.6 Thinner dye-polymer films

The aim of this experiment was to fabricate a thinner transducer (first order device using CA) with absorptive APC dye in CA medium. In other experiments performed using ITO and PS, the absorption and ultimately the generation of acoustic waves takes place in the first gold layer where, by adding absorptive dye into CA medium, it was suggested to tailor the transducer for an available wavelength for the pump laser beam as well as to generate acoustic waves through cavity rather than the first deposited gold layer in a FPI-based transducer. In this part of study the surface variation is investigated using a light microscope in reflection mode.

4.4.6.1 Observation of surface variation

A wide variation in the surface of a thin film can be seen in Figure 76 that has been taken from dye-CA thin film at 5x magnification. This wavy form is known as the striation form, which is created as a result of variations in the solvents' adhesion at different points caused by changes in the evaporation rate of the solvents [107].

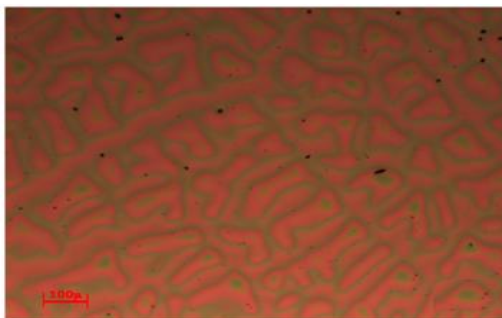


Figure 76: Light microscopy image of a dye-CA thin film in reflection mode at 5x magnification.

This result is not unusual when a mixture of solvents with different evaporation rates is used [107]. In this case the solvent with high evaporation rate (acetone) in solution evaporates quicker than the solvent with lower evaporation rate (DMSO) and produces a thin film with a poor uniformity thickness. This form is often seen in a solution that has a solvent with a high evaporation rate. In this case the solution loses the solvent without having enough time to become thin and spread uniformly. The images prove

a high variation in thickness, therefore high variation in mechanical frequency response across the sample [107].

4.4.6.2 Laser damage threshold test

Figure 77 shows damage threshold testing and the damage to the surface induced by a pulsed laser irradiation at the wavelength of 720nm at different power levels. Points P1-P4 are for power levels of 1.5mW, 1mW, 0.5mW and 0.3mW, respectively.

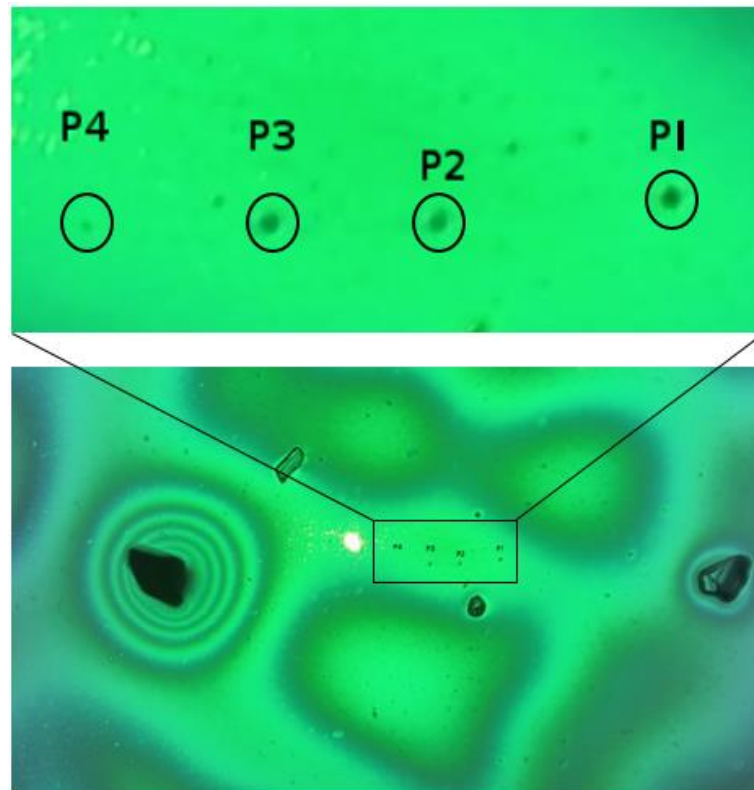


Figure 77: Light microscopy images (20x magnification) of a dye-CA thin film after a laser-induced damage threshold test on the sample. Pulsed-laser damage sites (P1, P2, P3, and P4) caused by different laser power levels at 720nm.

Damage sites can be seen in the image (Figure 77) after a laser-induced damage threshold test on a dye-CA thin film. In this experiment the main determining factor in damage is the absorption characteristics of defects of thin films. As it can be seen in Figure 77, the damage propagation initiated in site P1 and decreased in site P2. The damage does not propagate in site P4 and is relatively small in site P3. Eventually

a pump power was found in the safe level by which a dye-CA thin film can be tested with a lower risk of damaging. This level was approximately 0.3mW at the input to the final stage of the microscope and was therefore $\sim 0.2\text{mW}$ at the sample (x20 objective lens, 2.5pJ per pulse or $\sim 1\text{Wm}^{-2}$). The probe was $\sim 1\text{mW}$ at the sample (x20 objective lens, 12.5pJ per pulse or $\sim 2\text{Wm}^{-2}$) (see section 2.3.1).

This is a significantly lower power level than usually applied in experiments with ITO- and PS-based devices. It is not clear why the power handling of these films is so low. Based on one study reported by Rowen et al., the spectra of cellulose acetate of different thickness were observed and several well defined absorption bands in different wavelength regions were visible. The bands occur at 280, 340, 570, 720, 810, and 950 nm due to O-H, C-H stretching vibrations, ester carbonyl group, C-CH₃ group and C-O vibration, respectively. The strong band at 950nm is also present in all the infrared absorption spectra and is probably due to the C- O vibration [64]. It was observed that even a colourless CA thin film shows an absorptive band around 720nm where the maximum absorption for our dye is also in the same range about 730nm. This is expected to have higher absorption in the region of the wavelength where we intend to manipulate light absorption for the pump laser.

5.4.6.3 Optical model of dye-polymer thin film

Reflectance for λ_{pump} (415nm and 720nm) and λ_{probe} (780nm and $\geq 800\text{ nm}$) and sensitivity of λ_{probe} can be evaluated using the optical model. The absorption of a generation transducer is particularly important for acoustic generation because it predicts how effective the transducer is for generation of acoustic waves when driven by particular wavelengths of pump laser light. At the same time, the transmittance of the detection transducer dictates the detection signal level at the detector. The optical and mechanical model presented in sections 3.3 and 3.4 are also used to roughly predict the optical sensitivity ($\delta R/\delta H$ (nm^{-1})) and the mechanical resonance frequency for the range of devices presented in this work. The data in this experimental section pertains to the CA only device using the pump and probe laser pulses at the wavelengths of 415nm and 780nm, respectively. The evaluated κ for 15% APC dye in CA solution was used in the optical model to produce modelled spectra for calculating the thickness of dye-CA thin films.

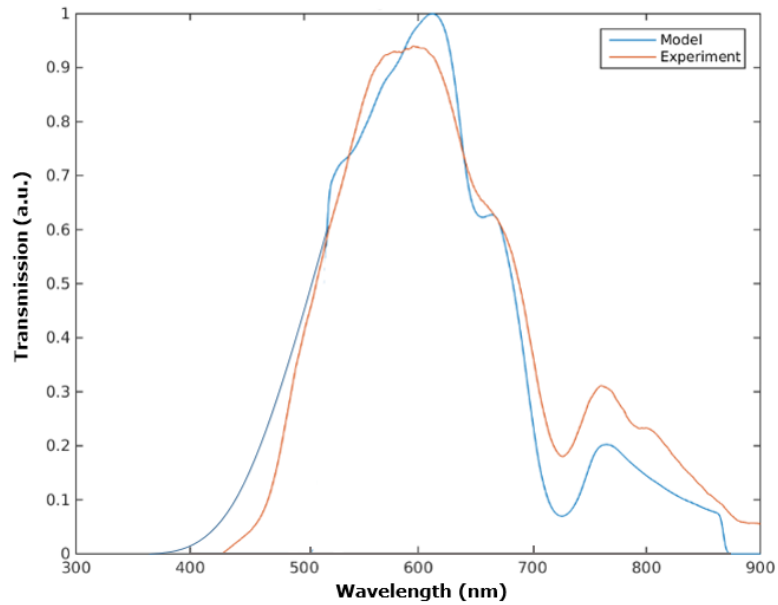


Figure 78: Transmittance spectra comparison between model (blue) and experiment (red). The blue trace represents transmission spectra calculated by Fresnel code with the experimentally calculated extinction coefficients for a 15% APC dye concentration. The experimental trace fits well to a 150nm layer of dye-CA thin film and 22nm of gold layers.

Figure 78 shows the dye-CA thickness measurements produced by the optical model. The new evaluated κ for 15% concentration of APC dye was used in the code to produce new model spectra. As it can be seen from model spectrum (blue curve), there is deep lower transmission spectra around 726nm due to dye absorption. The thickness of the dye-CA thin film was measured to be about 150nm and 22nm of the gold layers thicknesses. There is an acceptable agreement between the theoretical trace and experimental trace. These values were then used in the mechanical model to evaluate the resonance frequency of the fabricated device.

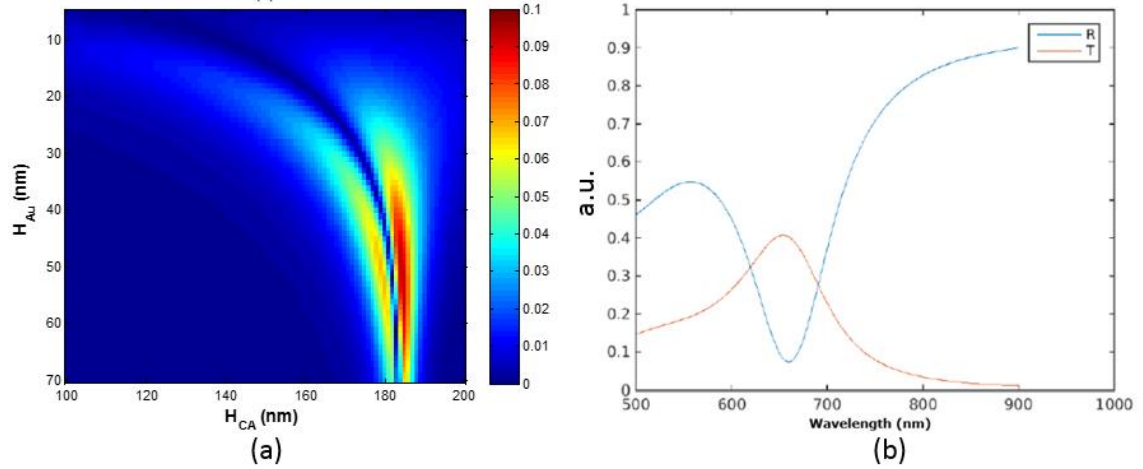


Figure 79: Transducer sensitivity $\delta R/\delta H$ at λ_{probe} (700 nm) (a), and transmission and reflection spectra (b) for the transducer with the layer thicknesses of 150nm for CA and 22nm gold layers at the top and bottom layers. The colour bar presents the sensitivity.

Figure 79 shows the results of the optical model for a transducer with wide range of gold (5-70nm) and CA (100-200) thicknesses. The sensitivity for the reflected ($\delta R/\delta H$) beam is presented. As shown in Figure 79(a), the sensitivity for the reflection case reaches a maximum of 0.023 nm^{-1} for the transducer with dimension of 22nm of gold layers and 150nm of CA layer as spacer. The model shows that a CA device with 150nm thickness is optically sensitive for 700nm rather than 780nm as desired but it still has some sensitivity at 780nm. The modelled response looks similar (both in terms of model and measured spectra) for ITO and PS devices of similar sizes.

5.4.6.4 Mechanical model

The mechanical model of the transducer was applied to predict the resonance frequency of the transducers depending on their layer thickness. To validate those results, experiments were performed in reflection rather than transmission for a practical reason as it explained in section 4.3, there is greater sensitivity to the resonance for the reflectivity case compared to the transmission case. Devices with $\sim 22\text{nm}$ thickness of the gold layers and CA layer thickness of 150 nm were fabricated, aiming for high sensitivity in the reflectivity case at 700nm, but as it mentioned before it still has some sensitivity at 780nm. The model geometry and output can be seen in

Figure 80 where the pump at 415nm and probe at 780nm were both incident on the surface of the device externally.

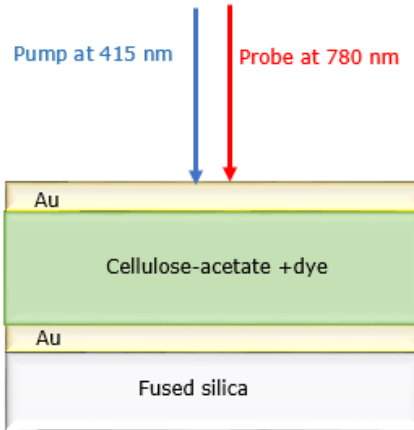


Figure 80: Schematic of a CA-based device (with and without dye) with a 150nm layer of CA and 22nm gold layers. The device fabricated on top of the glass substrate, and excited with 415nm and 720nm (for dyed medium) of pump laser pulses. The detection is in reflection at 780nm and ≥ 800 nm (for dyed medium) of the laser probe.

Figure 81(a) shows an example of the change of H_{CA} with respect to time. The transducer was tested in air. The Fourier transform shows a resonant frequency of approximately 1.7GHz, Figure 81(b). The high frequency components can be seen in Figure 81(a) due to the motion of H_{Au} that radiates waves into the transducer. In this case, the generation takes place in the first gold layer and because of this reason the absorption mostly happens in the first gold layer. The modelled generation in Figure 81(c) shows a temperature distribution in the thickness direction irradiated with a laser pulse (415nm) of 150fs duration, repetition rate of 80 MHz, average power of 1 mW, energy of 12pJ, and spot size of $4.5\mu\text{m}$ (see section 2.3.1). It is assumed that the temperature distributes homogeneously as it exponentially decays in the thickness direction by a single laser pulse incident on the transducer. As it is shown in Figure 81(c), the induced-temperature distribution increases at a distance of about 22nm (gold thickness) and drops dramatically in CA layer (150nm). The induced-temperature distribution at a distance of about 170nm (second gold layer) is similar to the temperature change at a distance of 22nm but it is relatively small compared to the changes in the first gold layer. In other words, the temperature is affected only within the gold layers by the pump laser incident.

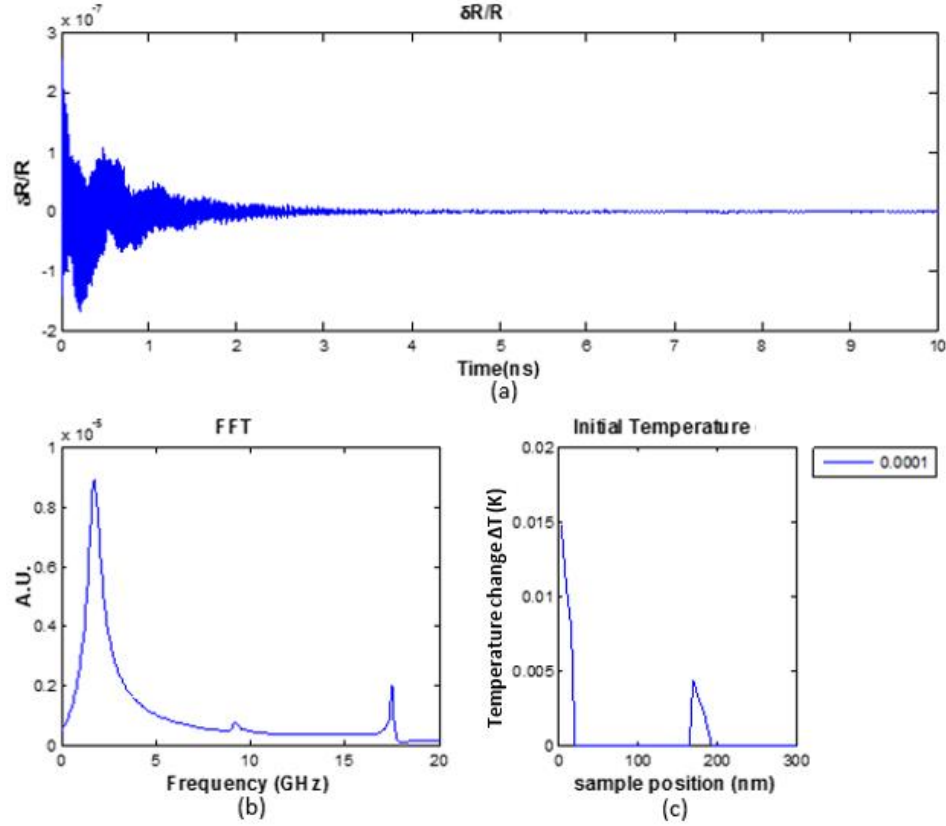


Figure 81: Simulation of the mechanical motion of a transducer with 22nm gold layers and 150nm CA layer. Modulation depth as a function of time (a), its Fourier transform (b), and temperature distribution in thickness direction (c). The CA is colourless with 0.0001 κ (nm $^{-1}$).

The averaged traces after truncations (a), thermal removal (b), filtering, and a Fourier transform (c) (80 GHz cut off) are shown in Figure 82. There is a string of low frequency signals at around 0.5 GHz and another signal around 9GHz for the Transmission case. Also, several peaks can be seen around 5GHz, 2.5GHz and 1.75GHz. It is suggested that those high frequency signals are noise on the laser system. The low frequency components are lower than expected for a first order CA device with the thickness of 150nm. It is suggested that the CA thin film surface variation means the average of many cavity peaks, and the more surface area is taken the measurement over the flatter the response is obtained due to the peaks averaging out.

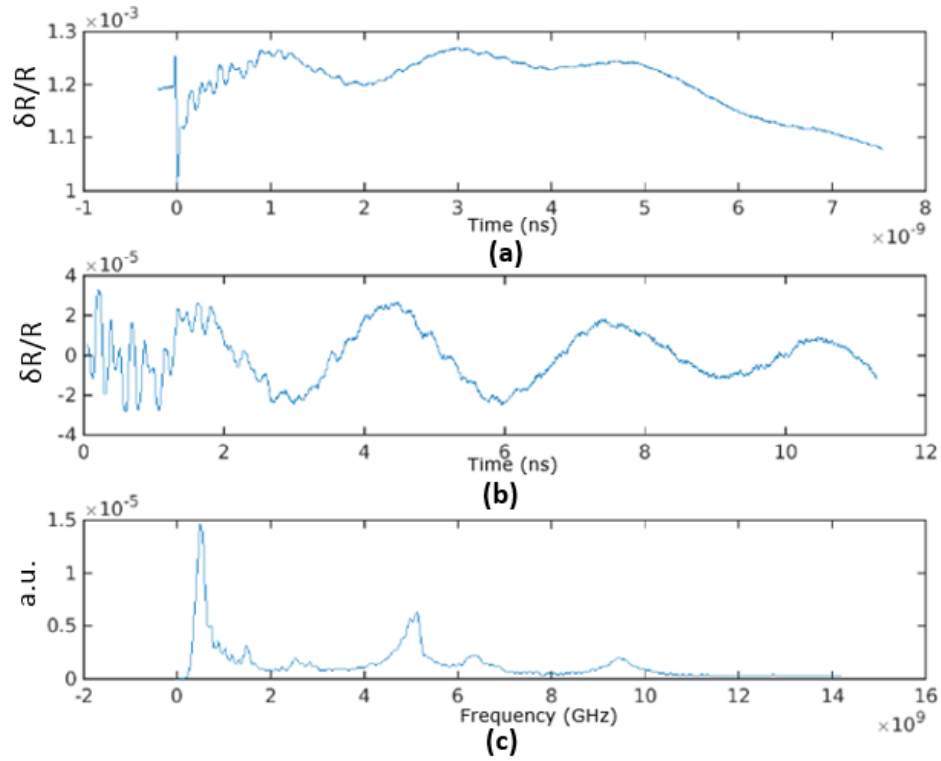


Figure 82: Average of all traces for 0.3mW case performed in reflection. The graphs present raw trace (a), temporal response or processed trace (b), and corresponding Fourier transform (c). A low frequency component at ~0.5 GHz and other peaks present.

The experiments in this section were performed with a pump laser pulse of 730nm and a probe of 830nm using the same experimental setup (Figure 31). A 0.3 mW of the pump laser power level was also used in this experiment. Figure 83 shows an example of the change of reflection with respect to time (a). The transducer was tested in air. The Fourier transform shows (Figure 83 (b)) a resonant frequency of approximately 1.7GHz with lower amplitude than tested with a pump laser pulse of 415nm (0.6 mW). The high frequency components seen in Figure 83(a) due to motion of H_{Au} is not present as the absorption of gold was removed in the mechanical model. This might hypothesise that the absorption occurred in the cavity due to absorptive dye rather than in gold layers. The modelled acoustic generation in Figure 83(c) shows the induced-temperature distribution increases within the cavity and decreases exponentially in the thickness direction by the incident pump power of 1mW and wavelength of 720nm. This means, the temperature change is affected only with the

region of dye-CA layer. It is mainly caused by the high optical absorption of dye-CA due to the added absorptive dye in CA medium.

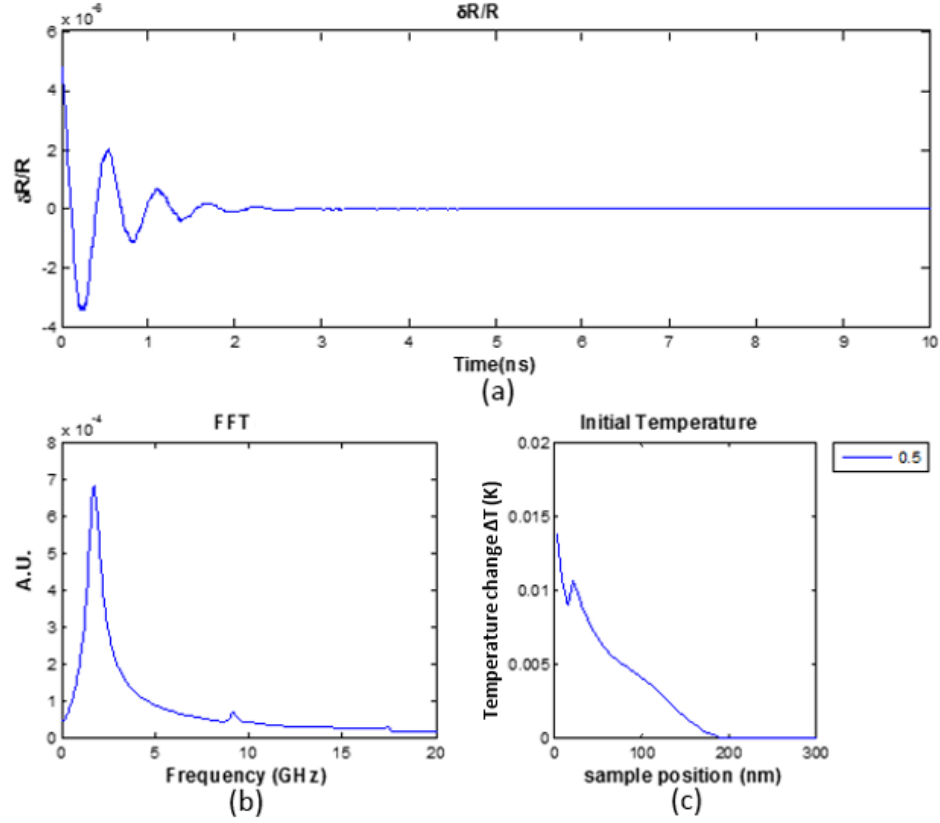


Figure 83: Simulation of the mechanical motion of a transducer with 22nm gold layers and 150nm dye-CA layer. The graphs present modulation depth as a function of time (a), its Fourier transform (b), and a temperature distribution in the dye-CA layer (c). The extinction coefficient of the CA medium is 0.5 nm^{-1} in this case.

Figure 84 shows the averaged traces after truncation (a), thermal removal (b), and (c) filtering and performing an FFT (80GHz cut off). Due to high thin film thickness variation, the sample was moved around until a noticeable signal was found on the oscilloscope. There is a strong low frequency signal at around 0.55 GHz and another signal around 8GHz for the transmission case. The peaks at 5GHz, 2.5GHz and 1.75GHz can also be seen as the peaks which were observed in Figure 81(b).

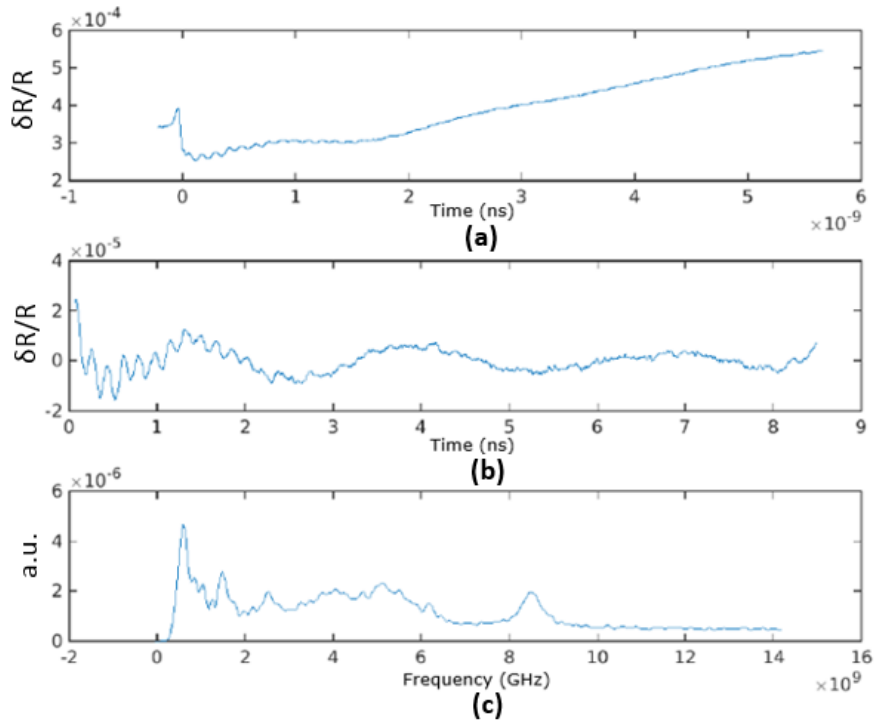


Figure 84: Average of all traces for 0.3mW case performed in reflection. The graphs present raw trace (a), a temporal response or processed trace (b), and corresponding Fourier transform (c). A low frequency component at ~ 0.5 GHz and other peaks present.

The same experiment was performed with the same power level (0.3 mW) of the pump laser pulse but in transmission mode (Figure 32, but in free-space configuration). Figure 85 shows the signals produced by the pump laser pulse at 730nm with power level of 10mW, and the probe at 830nm with power level of 1.4mW. The transmission data in this case is actually now reflection data because the probe is fed from the top (and *vice versa*). The reflection data is very similar to that obtained using 415nm of the pump and 830nm of the probe. The signal is around 0.55 GHz in Figure 85(c) and there is another peak attributable to the sample around 8GHz. The slight shift in the peak location is likely due to being in very different areas of the sample.

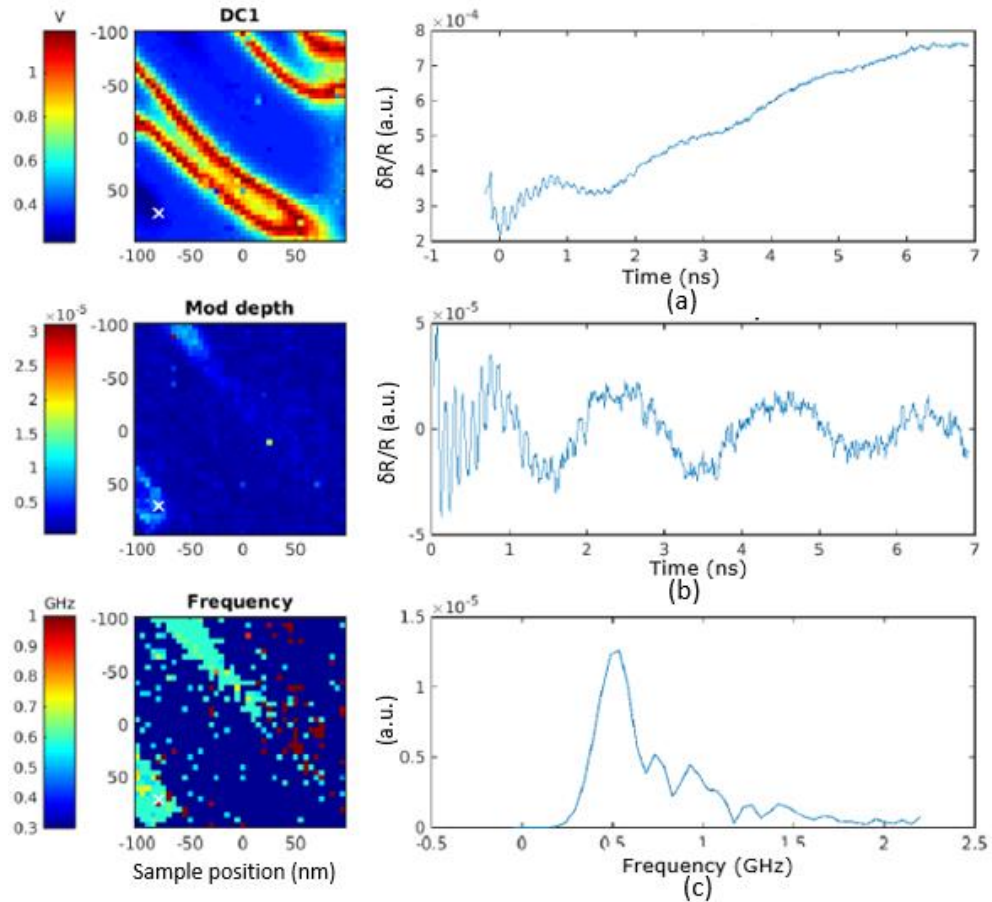


Figure 85: Imaging scan of a large area showing big swings in DC light levels and small regions of good signal levels. The graphs present raw trace (a), a temporal response or processed trace (b), and corresponding Fourier transform (c). A low frequency component at ~ 0.59 GHz and other peaks present.

As a summary, from thinner CA-based devices with and without absorptive dye, it is suggested that relatively good low frequency measurements can be obtained from poly plate devices. However, the devices get damaged easily, which might need more treatment. The surface variation is still a big problem both in terms of thin-film thickness measurement and the generation of ultra-high frequency, tuneable acoustic waves. Also a relatively good low frequency measurement from dyed-poly plate devices can be achieved using the pump laser pulses at blue and IR regions. In this case, also the devices are damaged easily with UV but seem to tolerate high IR levels, even with the dye.

5.4.7 Higher order dye-polymer thin films

As the next step in the experiments, the aim was to fabricate thicker transducers (higher order devices) with the same dimensions with and without absorptive dye in CA medium for generation enhancement. The wavy pattern in Figure 86 shows an inhomogeneous morphology and high surface roughness in the sample.

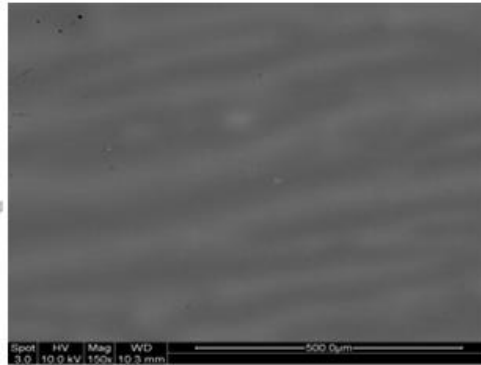


Figure 86: An image from Scanning Electron Microscopy of 10% dye in 40% CA solution dissolved in a mixture of solvents (acetone and DMSO).

5.4.7.1 Optical model

Table 7 presents the evaluated data from optical and mechanical models. It is expected that the frequency will be decreased as the dimensions of the transducer are increased.

Table 7: The sensitivity ($\delta R/\delta H$) at probe wavelength and frequency at pump wavelength are evaluated for thin films with different thickness with and without dye in medium.

Deposited thin films with 10% dye in 40% CA solution			
Spin speed (rpm)	Thickness of samples (nm)	Sensitivity (nm^{-1}) ($\delta R/\delta H$) at $\lambda_{\text{(probe)}}$ (nm)	frequency (GHz) at $\lambda_{\text{(pump)}}$ 720 nm
1500	905	0.052 at 712	0.65
2500	795	0.078 at 820	0.6
3000	695	0.040 at 737	0.5
Deposited thin films with colourless 40% CA solution			
Spin speed (rpm)	Thickness of samples (nm)	Sensitivity (nm^{-1}) ($\delta R/\delta H$) at $\lambda_{\text{(probe)}}$ (nm)	frequency (GHz) at $\lambda_{\text{(pump)}}$ 415 nm
1500	890	0.031 at 710	0.75
2500	790	0.045 at 830	0.60
3000	550	0.030 at 880	0.55

Transducers with approximately 27nm of gold and different CA layer thicknesses were fabricated with 10% dye in 40% CA solution and also without adding dye to 40% CA solution were used in this section of experiment. Thicker CA films were made to calculate the extinction coefficient of dye at the higher concentrations. The κ of 10% dye concentration is 0.3 (nm^{-1}) based on the results from previous experiment in section 5.4.5 in Figure 75. The optical spectrum (in transmittance) of a single device, whose CA layer thickness is unknown, was measured and compared with its modelled counterpart. Figure 87 shows the result of thickness measurements using the transmitted spectrum of a transducer with gold layers of 27nm and CA layers of 790nm (a) and 795nm (b). In the transducer with 795 nm thickness of dye-CA layer, absorptive APC dye was added to the medium. These thicknesses were chosen as the devices with lower sensitivity cannot be characterised as their signals are not detectable (see section 3.3). There is a good agreement between theoretical curves (blue line) and experimental data (red line). These values are used in the mechanical models to calculate the resonance frequency.

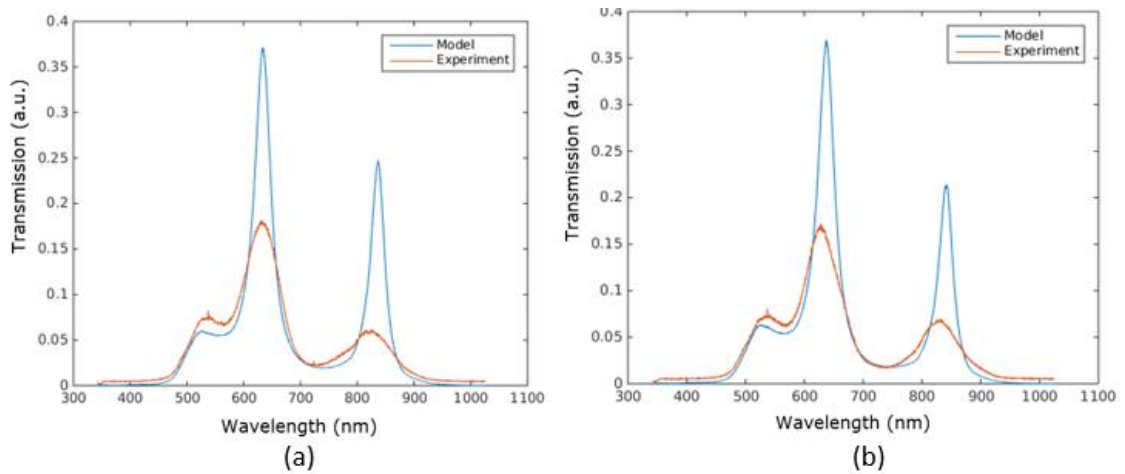


Figure 87: Example of transmittance comparison between model (blue) and experiment (red). The blue traces represent different CA layers 790 nm (a) and 795 nm (b) with adding dye the CA medium.

Figure 88 shows the results of the optical model for the transducer with a wide range of gold (5-70nm) and CA (750-850nm) thicknesses. The sensitivity for the reflected ($\delta R/\delta H$ (nm^{-1})) beam is presented. It can be seen that the sensitivity for the reflection case Figure 88(a) reaches a maximum of 0.059 nm^{-1} for the transducer with

dimension of 790 nm of 27 nm of gold layers and 790nm of CA layer where in transmission case Figure 88(b) it is about 0.02 (nm^{-1}). This means that the reflection case is twice as good for detecting incoming waves. Maximum reflectance for thick gold layers (35-40nm) can be as high as 0.07(nm^{-1}), which would be suitable for detection in reflection. Similar sensitivity is expected for the transducer with 795nm CA with dye in medium.

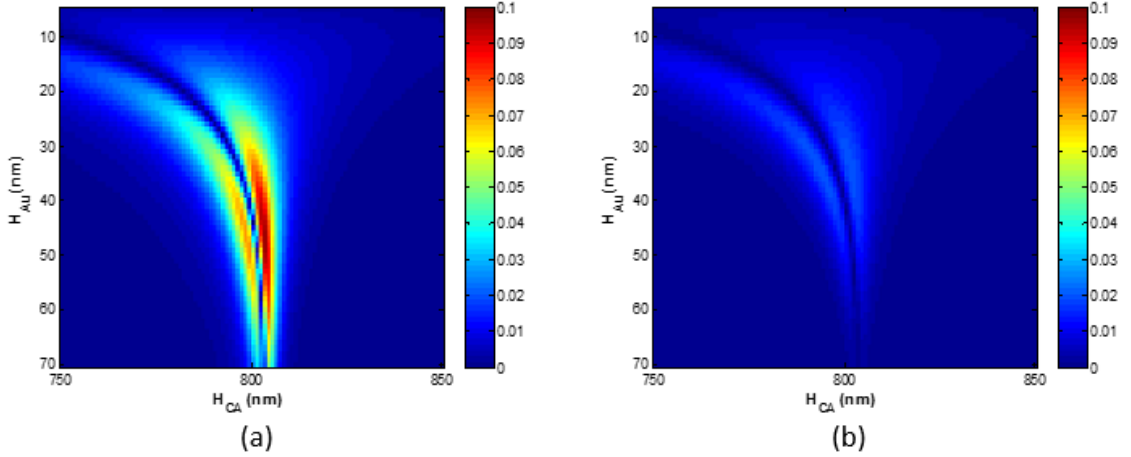


Figure 88: Transducer sensitivity $\delta R / \delta H$ (a) $\delta T / \delta H$ at λ_{probe} (831 nm) for the transducer with the layer thicknesses of 790nm for CA. The colour bars in both images present the sensitivity (nm^{-1}).

5.4.7.2 Mechanical model

The mechanical model of the transducer was applied to predict the resonance frequency of the transducers depending on their layer thickness. To validate those results, the experiments were performed in reflection rather than transmission for a practical reason as in can be seen from Figure 89 there is greater sensitivity to the resonance for the reflectivity case compared to the transmission case. Devices with ~ 27 nm thickness of the gold layer and different CA layer thicknesses (790 and 795 nm) where fabricated aiming for high sensitivity on the reflectivity case.

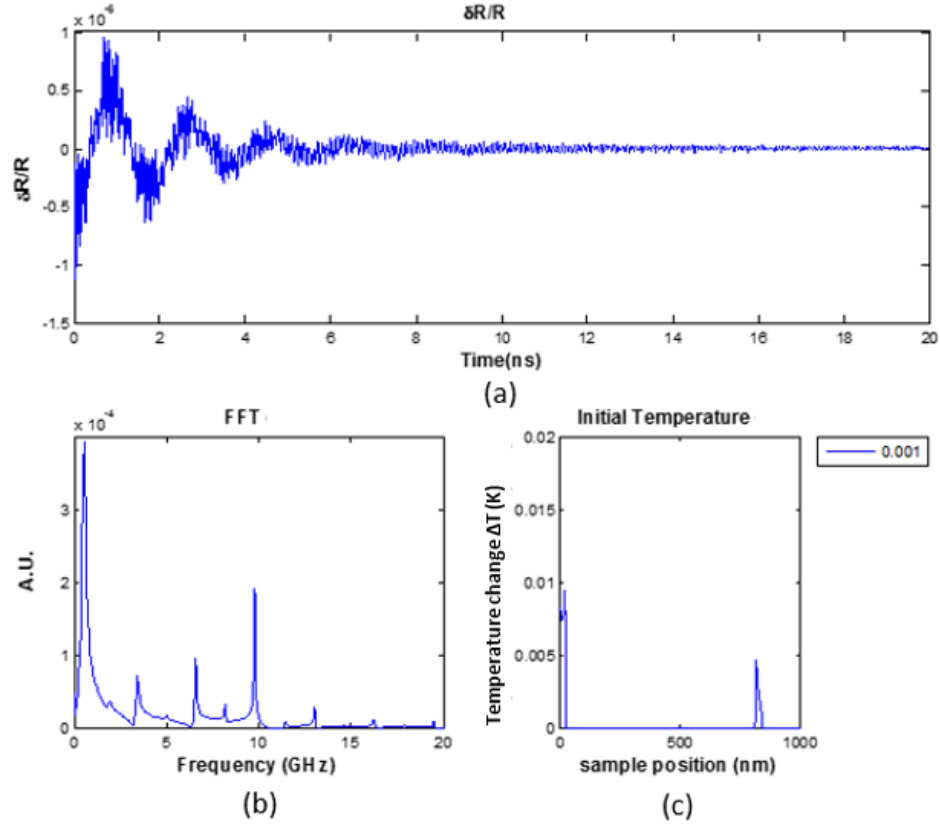


Figure 89: Simulation of the mechanical motion of a transducer with 22nm gold layers and 790nm CA layer. The graphs show modulation depth as a function of time (a), its Fourier transform (b), and temperature distribution in thickness direction (c). The spacer colourless with 0.001 (nm^{-1}) extinction coefficient in this case.

Figure 89 shows an example of the change of H_{CA} with respect to time (a). The traces are simulations of the effect induced by a laser pulse (λ_{pump}) incident at 415nm wavelength on the transducer. The modelled generation transducer based on gold-CA-gold topology with the dimensions of 27-790-29 nm, and attached to the glass substrate. The Fourier transform shows a resonant frequency of approximately 0.6 GHz. The high frequency components can be seen in Figure 89 (a) from the motion of H_{Au} that radiates waves into the transducer. In this case, the generation takes place in the first gold layer and because of this reason the absorption mostly happens in the first gold layer. As shown in Figure 89(c), the temperature change is only affected within the gold layers by the pump laser incident at the wavelength of 415nm and an average power of 1mW. The induced-temperature distribution drops dramatically at

a distance between 27nm and 820nm in CA layer, which shows there is no absorption occurs in the cavity.

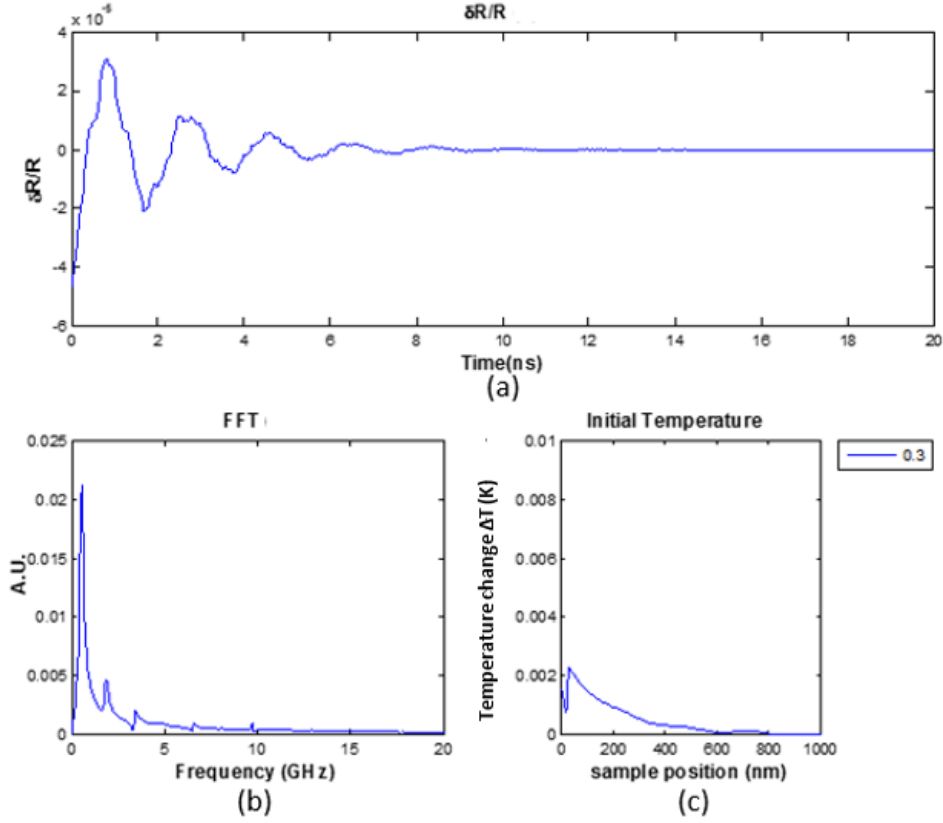


Figure 90: Simulation of the mechanical motion of a transducer with 29nm gold layers and 790nm CA layer. The graphs show modulation depth as a function of time (a) and its Fourier transform (b), induced-temperature distribution in thickness direction (c). The spacer is a dye-CA medium with $0.3 \kappa \text{ (nm}^{-1}\text{)}$.

Figure 90 shows an example of the change of H_{CA} with respect to time (a). The traces are simulations of the effect induced by a laser pulse (λ_{pump}) at 726 nm on the transducer where the absorption takes place in cavity due to addition of dye absorptive dye in CA medium. The modelled generation transducer based on gold-CA-gold topology with the dimensions of 29-795-27 nm, and attached to the glass substrate. Figure 90(c) it can be seen that the temperature change is affected only with the region of dye-CA layer due to the absorptive dye. The induced-temperature distribution increases within the cavity and decreases exponentially in the thickness direction by the incident pump power of 1mW at wavelength of 720nm.

Since the absorption occurs in the cavity rather than using the first metallic reflector layer alone. It is assumed that the temperature is distributed equally in the cavity and is exponentially decaying in the CA thickness direction. The κ of 10% dye concentration is $0.3 \text{ (nm}^{-1}\text{)}$ based on the results from previous experiment (see section 5.4.5) and it was used in this study to investigate the mechanical response of the transducer. The transducer is in air and attached to a glass substrate. The Fourier transform shows a resonant frequency of approximately 0.6GHz, which is similar to the frequency produced with the transducers without dye in CA medium. The high frequency components seen in Figure 89 (a) are not observed in Figure 90(a) since the absorption occurred in the cavity due to added absorptive dye in the CA medium rather than in gold layers.

5.4.8 Increasing thickness by sputter coating assisted spin coating method

The aim of this part of experiment was to improve the uniformity and increase the thickness of CA thin films. The simple multi-layered deposited whole stack is shown in schematic diagram in Figure 91. Compared with single spin coating process to fabricate deposited film on top of the glass substrate, this is another efficient method and applicable to produce multilayer deposited films from the same material, in successive different layers. It was concluded from previous experiments (see section 5.4.3) that increasing concentration of solution, even decreasing the spin speed is not an appropriate way to increase thickness. Using high concentration of solution results in a wavy surface where the thickness of each layer needs to be in an exact size in order to optimize the sensitivity of the transducer.

The transparent ITO layers are very thin in dimension ($\sim 3\text{nm}$) and in comparison with the probe wavelength ($< 780\text{nm}$) the presence of the ITO layers can be neglected. From previous study (section 5.4.3), a uniform thickness with low roughness was obtained using DMSO as a single solvent to dissolve both CA and APC dye. The same protocol was used to study the fabrication of CA thin films dissolved in single solvents (acetone or/and DMSO) and increase the thickness by depositing thin films layer by layer method. Several samples were prepared with 10% dye mixed in 30% CA solution using acetone or DMSO as a single solvent. The thickness, roughness, and

refractive index of each thin film were measured using ellipsometry and the results are presented in Table 8.

Table 8: The data from ellipsometry measurements for samples fabricated on the top of the glass substrate.

Deposited thin films with 10% dye in 30% CA solution dissolved in acetone				
Samples	Spin speed (rpm)	Thickness (nm)	Roughness (nm)	Refractive index (n) at 630nm
CA	1000	120	3.59	1.47
CA+dye	1000	134	0.17	1.46
CA	2500	103	0.93	1.47
CA+dye	2500	105	0.33	1.47
Deposited thin films with 10% dye in 30% CA solution dissolved in DMSO				
Samples	Spin speed (rpm)	Thickness (nm)	Roughness (nm)	Refractive index (n)
CA	1000	105	2.63	1.47
CA+dye	1000	108	7.91	1.46
CA	2500	99.5	0.70	1.46
CA+dye	2500	102	0.42	1.46

By using the data from Table 8, a whole stack was fabricated using layer by layer deposition method. As the procedure was explained in Chapter 3, the thin films were spin coated with 10% dye mixed with 30% CA solution using DMSO, spin coated at 2500 rpm. A control sample was also fabricated individually at each step on the top of a glass substrate. The thickness of each layer in whole stack was calculated using control samples by spectroscopic ellipsometry and the results are shown in Table 9. The transmission spectra were measured in each step and plotted on top of each other in order to observe the spectrum as the thickness is increased.

Table 9: The data from ellipsometry measurements for each control sample to calculate the thickness of each layer (1-5) in whole stack individually.

Number of layer	Thickness (nm)
1	103
2	93
3	102
4	114
5	105

It can be seen from Figure 91 that the transmission spectra of different layers show almost similar absorption band pattern around 730nm. Increasing the number of layers reduces the peak in-band transmission at 730nm. It is likely that a thin film layer of ITO protects the deposited CA thin film from being dissolved by deposition of another CA thin film on top. The absorbance band does not increase in proportion to the film thickness in two first layers, but increasing the thickness produces a corresponding increase in blocking the wavelength of interest. This infers that the thin film layers were successfully added and protected by an ITO layer.

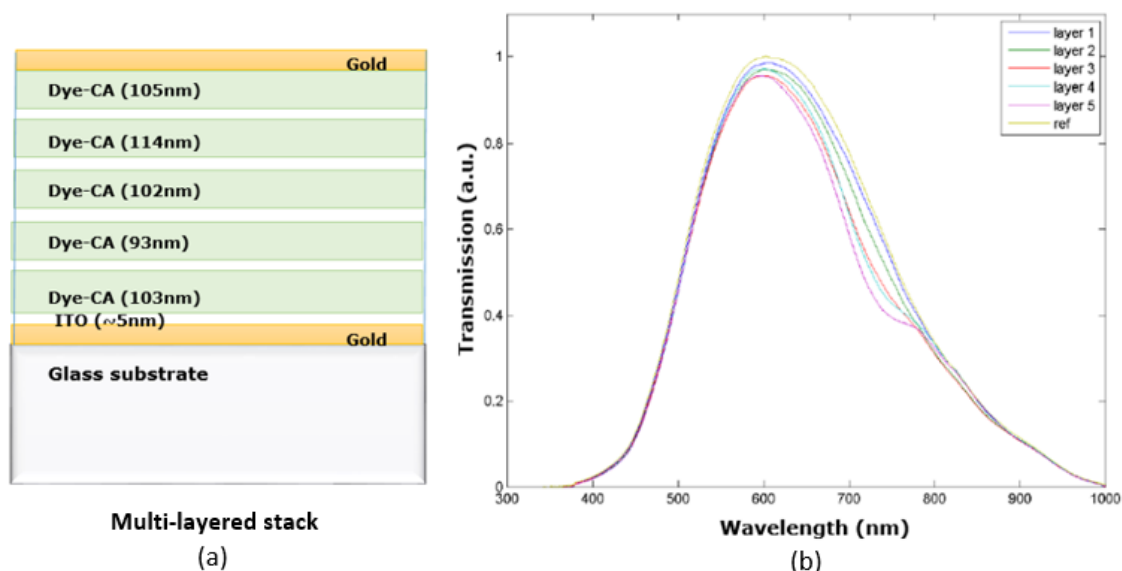


Figure 91: schematic of multi-layered CA stack (a) and measured thickness of each layer individually. The transmission spectra of each associated layers (b).

The images from SEM were taken at different magnifications and are shown in Figure 92. The non-uniform pattern that was seen from thin film in previous studies, has been improved significantly. More non-uniform surface can be seen around those undissolved dye pieces on the surface.

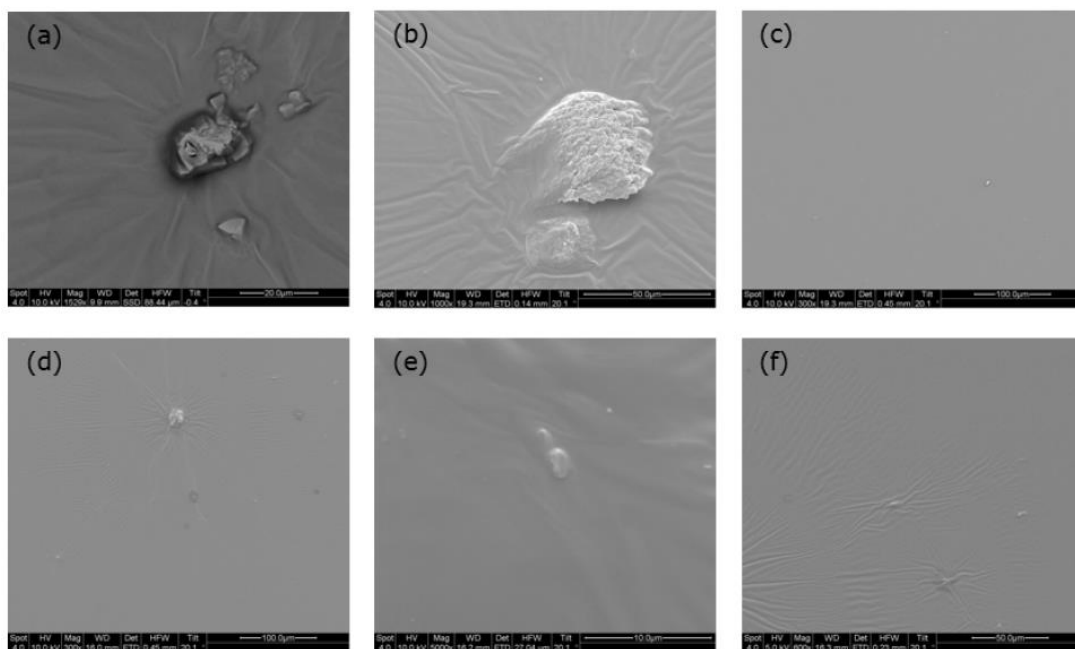


Figure 92: The images from SEM with magnification of 1500x and 20 μ m scale bar (a), 1000x and 50 μ m scale bar (b), 300x and 100 μ m scale bar (c), 300x and 100 μ m scale bar (d), 5000x 10 μ m scale bar (e) 500x and 50 μ m scale bar (f). The images have been taken from different part of the same sample.

Figure 93 (a) presents results of thickness measurements using transmission spectroscopy of a transducer with gold layers of ~ 30 nm and multi-layered CA stack of 550nm (red curves) and theoretical trace (blue curves). The experimental trace shows an agreement to its modelled trace. It was expected to obtain ~ 500 nm CA thickness from five layers. By adding ITO thicknesses which is $\sim 5-7$ nm for each layer, a 530nm CA thin film was expected to be obtained.

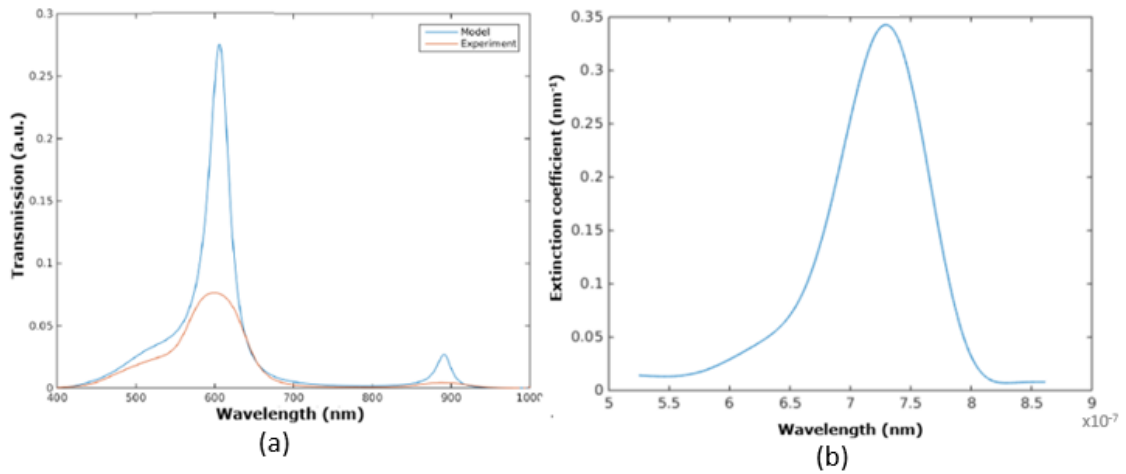


Figure 93: Transmittance comparison between model (blue) and experimental (red) traces for the multi-layered CA stack thickness of 550nm and ~ 30 nm gold layers (a). Calculated extinction coefficient (κ) for the measured thickness with 10% dye (b).

Figure 93 (b) shows the plot of the extinction coefficient (nm^{-1}) as a function of the wavelength of the dye-CA thin film. The maximum κ was found about 0.35 nm^{-1} for 30% of APC dye mixed with CA solution at 728 nm. The concentration of dye in this solution was 10%. Based on previous study, 0.35 nm^{-1} of κ was expected for a 10% APC dye solution and the graph shows the same value for the κ .

Table 10: The sensitivity ($\delta R/\delta H$) at probe wavelength and frequency at pump wavelength are evaluated for the multi-layered CA stack.

Deposited CA thin films with 10% dye in 30% CA solution				
Spin speed (rpm)	Thickness (nm): Au:CA:Au	Sensitivity (nm^{-1}) ($\delta R/\delta H$) at λ (probe)(nm)	frequency (GHz) at λ (pump) 415 nm	Extinction coefficient (nm^{-1})
2500	27-550-35	0.084 at 884	0.75	0.35

The results from optical and mechanical validation of the device with thickness of 550nm of CA layer are shown in Table 10. For the 550nm CA device the optical sensitivity in reflection mode is $0.084 (\text{nm}^{-1})$ at 884 nm of probe and the frequency of 0.75 GHz produced by the device.

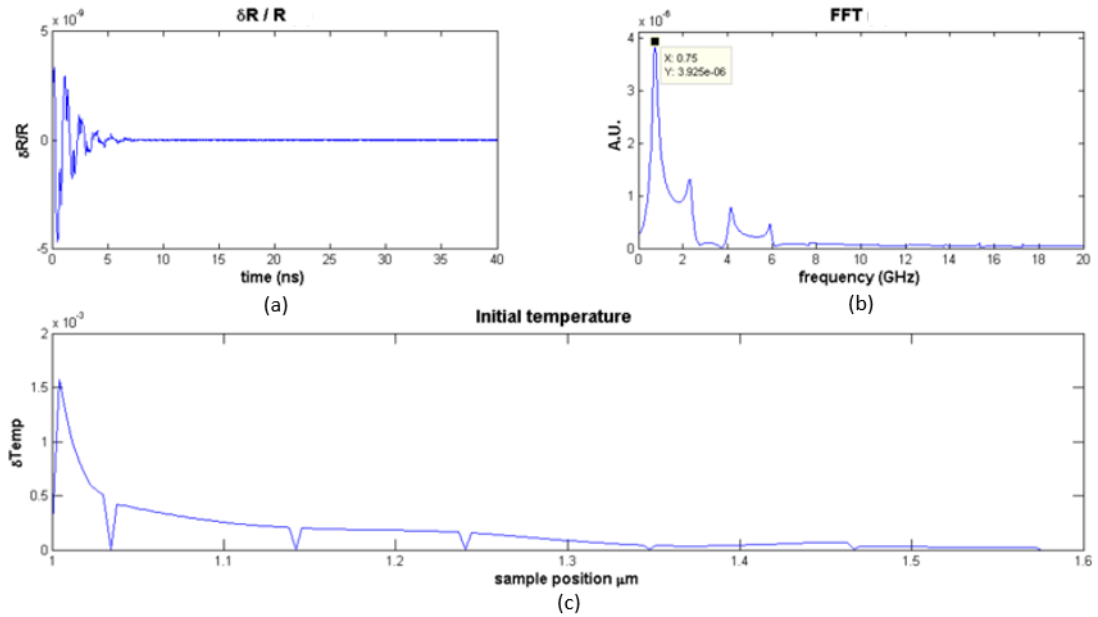


Figure 94: Simulation of the mechanical motion of a transducer with 27nm and 35nm gold layers and 550nm multi-layered CA stack. The graphs present modulation depth as a function of time (a), its Fourier transform (b), and induced-temperature change in a multi-layered CA stack (c). The extinction coefficient is 0.35 nm^{-1} in this case. The dips present ITO layers.

The mechanical model is stimulated for the given thickness and the results are shown in Figure 94. As can be seen, the duration of oscillation for a 550nm CA device mixed with dye ($0.35 \text{ nm}^{-1} \text{ k}$) is $\geq 15 \text{ ns}$ where for higher order PS device the long duration of oscillation caused some difficulties to measure the lower frequency signals. The main frequency is about 750MHz but higher frequency is also observed. From (c) it can be seen that the temperature is distributed between the layers of CA stack and it decreases exponentially in thickness direction. This infers that in order to obtain a thick device using layer by layer deposition method will be effective method to control the temperature distribution within the stack. Unfortunately, these results were not be validated by experimental results due to an unexpected personal problem that occurred during my study and I had to finish experimental practice at this point.

Chapter 6

Future work

6.1 Introduction

In the latest experiment, it was demonstrated that a sputter coater assisted spin-coating method to increase the thickness of the thin films. In this method a very thin layers of ITO (2-3nm) were deposited between CA polymeric thin film layers. Improvement of fabrication method can offer many advantages such as flexibility in tuning the absorption behaviour of film layers in multi-layered stack by adjusting system parameters such as dye solutions concentrations in each layer or the thickness of layers individually.

In this study, it was proposed and partially demonstrated that the acoustic waves can be generated more efficiently by matching the optical properties of dielectric thin films such as absorption coefficient (α) with the wavelength of pump laser beam. The absorption behaviour at specific wavelength of a dielectric thin film can be tuned by adding absorptive dye. This method has several advantages over the current approach (i.e. metallic layers in FPI structure) for generation of acoustic waves. Another advantage is the possibility of generation of acoustic waves through the optical cavity. Additionally, the generation of acoustic waves can be conducted by the new configuration of FPI where the generation can take place outside of reflectors at a tunable absorbing nanoscale thin-film system. In this Chapter, future experiments are discussed that can help to improve both fabrication methods and new configurations of the transducers. The possible applications of the transducers are also discussed.

6.2 Improvement in fabrication of thin films on the tip of single mode fibre using sputter coating assisted dip coating method

Based on the results from the last experiment (see section 5.4.8), we could increase the thickness of the transducer spacer deposited onto the microscopic glass substrate

using layer by layer deposition method or sputter assisted spin coating method. This experiment was conducted because the DMSO was suggested as the suitable solvent for spin coating of the layers of dye-CA polymer in plate transducers, whereas fabricated thin films using a mixture of solvents acetone and DMSO were not smooth as compared to the ones prepared out of weakly volatile solvents. The influence of solvents on the formation of uniform thin films can be observed in all the performed experiments using the spin coating method. Multi-layered sputter assisted spin coating was initially studied experimentally on plate transducers to improve the uniformity or increase the thickness of thin films. Multi-layered sputter assisted dip-coating method can also be studied to fabricate a multi-layered transducer on the tip of the single-mode fibre. Figure 95 shows a schematic of multi-layered thin film fabricated on the tip of a single-mode optical fibre using sputter coater assisted dip-coating method.

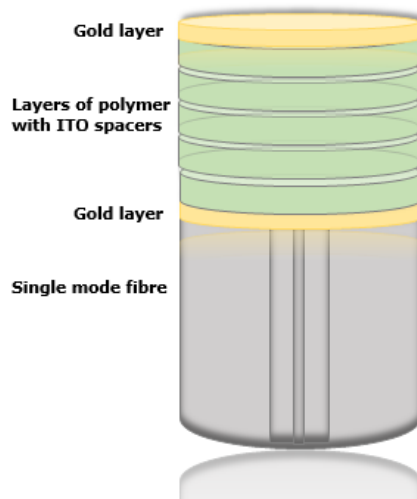


Figure 95: Multi-layered transducer fabricated on the tip of a single mode fibre.

The process is defined in two steps procedure to increase the thin film thickness by deposition of material layer by layer. The fibre is dipped into a coating solution and after drying step, it is sputter coated with a very thin layer of ITO ($\geq 3\text{nm}$) to protect the coated layer with solution from dissolving with another dipping process. Compared with single dip-coating process to fabricate polymeric film on the tip of single mode fibre, the repetition of dip-coating is more efficient and applicable to increase and control the thickness of thin films as a multilayer deposited film. This method of dip

coating also can be effective to fabricate a planar transducer where the dip coating with high concentration can produce a round shaped transducer on the tip of an optical fibre as shown in Figure 96.

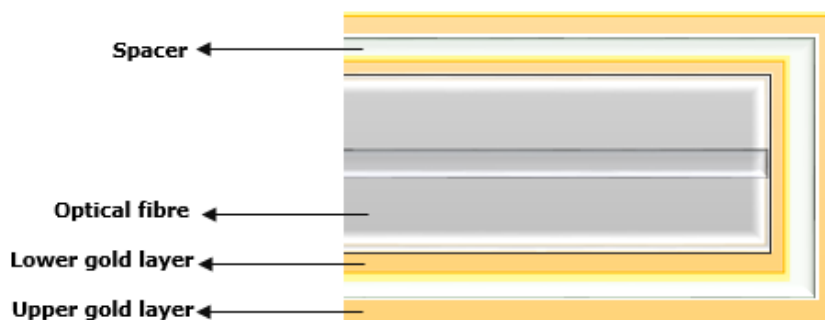


Figure 96: Schematic of a planar transducer.

This method of depositing also offers a great advantage over conventional dip coating method. In conventional method, different thickness of thin films are produced by changing different parameters in dip coating such as withdrawal speed or solution concentration where in the new proposed technique, by using only one withdrawal speed and concentration of medium we can increase the thickness.

However, it is still needed to investigate the suitable solvent and medium concentration to fabricate multi-layered transducer on the tip of a single mode fibre. In addition to solvent influence, other parameters such as drying methods and medium concentration are also effective in fabrication of thin films. In the next part of this study, it will be mainly concentrated on the mechanism, recently developed for fabrication of multi-layered transducer using dip coating instead of spin coating. The emphasis will be made here to present practical basis of dip coating film deposition using layer by layer method especially, to increase the thickness with high uniformity based on various processing parameters. The multi-layered dip-coating method in this experiment can be introduced as a modified dip coating method to deposit thin films as a sputter coating-assisted method.

6.3 Controlling temperature distribution in thick dielectric coating

Compared with the repeated dip-coating process of single coating solutions, multi-layered dip-coating for different coating solutions show more advantages considering the synergistic effect of deposited layers. The multi-layered dip-coating is different from layer-by-layer assembly. In multi-layered dip coating, different layers from same solution or different polymeric solution can be deposited layer after layers by depositing a protective layer such as ITO between each layer whereas in layer by layer assembly deposition, the layers are fabricated by alternately dipping a charged substrate into aqueous solutions of oppositely charged materials with intermediate steps of rinsing in water.

The sputter assisted dip coating deposition method can contribute to increase uniformity as well as temperature distribution in the higher order transducers of this study. As it previously described, the temperature is increased where the pump laser light is absorbed. Instantaneous temperature rise in thickness direction (δ Temperature (K)) causes thermal stress and thermal expansion within the region irritated by the incident pump pulse. The strain components caused by the thermal expansion. The strain distribution in the thin coating can be divided into two parts: a static part due to the thermal expansion near the free surface and a propagating part with high frequency content up to several GHz. The stress, strain, and temperature are strongly associated within the structure in order produce acoustic waves. The Figure 97 shows the temperature profile of a 550nm thick optical cavity. The pump laser beam at 415nm with spot size of 4 μ m in diameter was used to irradiate the transducer and induce the temperature in 30nm of gold layer in a three layer FPI.

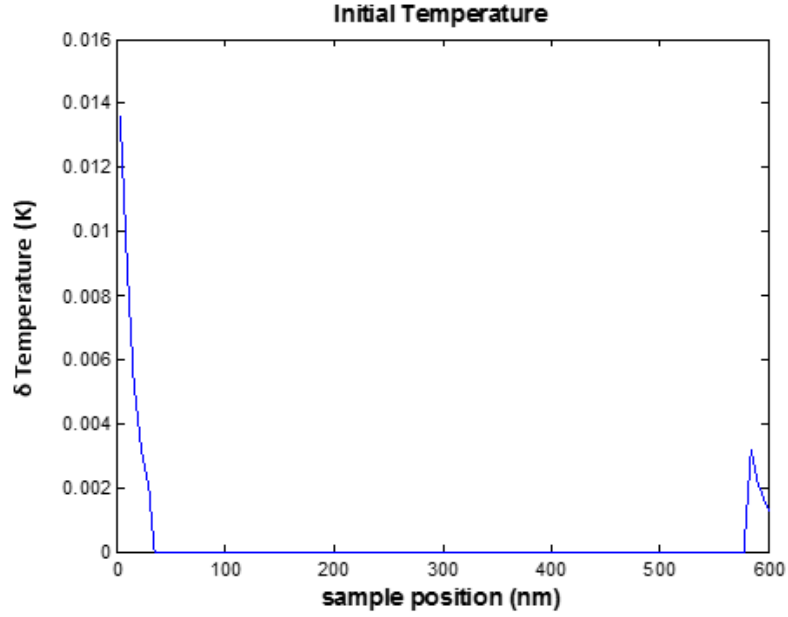


Figure 97: Illustration of the induced temperature change in thickness direction in gold thin film of a FPI.

The temperature rise is mainly caused by the high optical absorptivity of gold. The area of the specimen irradiated by the laser pulse is large compared to the film thickness d and the absorption length ξ . The temperature is assumed to be homogeneously distributed in the plane and is exponentially decaying in the thickness direction with $\delta T = \epsilon T / C$ (see section 2.3.1). Therefore the excitation of the acoustic waves is considered in the z direction only. By an efficient coupling between molecular absorption and Fabry-Perot resonances in the thin-film cavity, a proper excitation condition of FPI-based transducer can be uniquely set to generate acoustic waves through the cavity.

For 415nm of pump laser beam the skin depth is about 34nm and it is comparable with the thickness of gold which is usually about 20-30nm in conventional FPI in our work. For 728nm of pump laser beam, and 0.3nm^{-1} the skin depth is $\sim 342\text{nm}$ and if higher order of thickness ($1\mu\text{m} \leq$), the skin depth is not comparable and the temperature will not be distributed as it was in gold.

The amplitude at the Brillouin frequency is another parameter in the designing a transducer as an acoustic wave generator. This parameter is considered for optimization of Brillouin detection for each set of thicknesses. The amplitude of frequency determines the amount of energy is available for the detection of Brillouin oscillations. By evaluating the amplitude of Brillouin frequency before fabrication of the transducers, we can determine the most suitable design parameters.

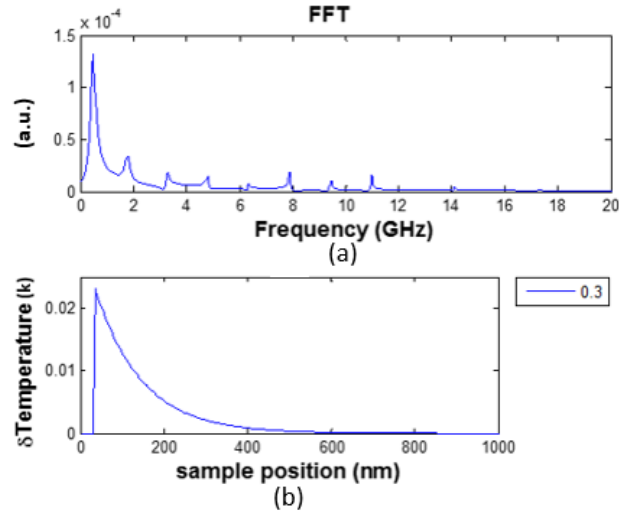


Figure 98: Mechanical simulation of 540nm of CA and 30nm of gold layers with 0.3 (nm^{-1}) κ . The graphs present mechanical response (a) and induced-temperature change in thickness direction (b).

As can be seen from Figure 98(b), the temperature is mainly affected in the near surface region where the pump laser beam is mostly absorbed by this region of a cavity. Thus, the amount of light that can be transmitted within the cavity is decreased. This is because the most of light is absorbed initially where the light enters the cavity. The highest amplitude of the frequency 0.625 (GHz) is about 1.3×10^{-4} (Figure 98(a)) in this case.

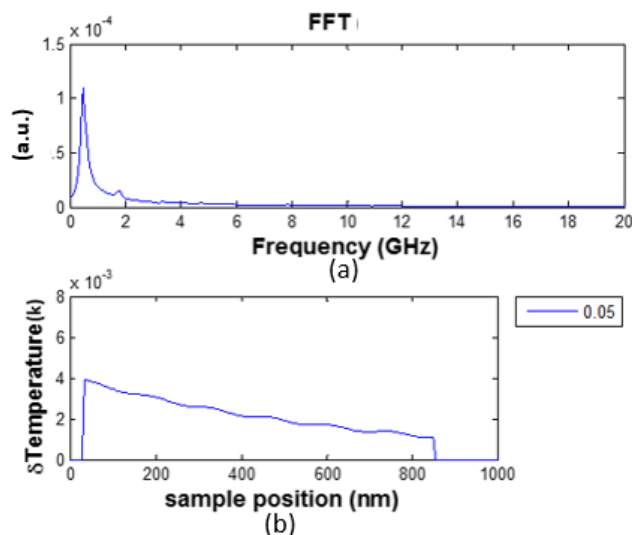


Figure 99: Mechanical simulation of 540nm of CA and 30nm of gold layers with $0.05 \text{ (nm}^{-1}) \kappa$. The graphs present mechanical response (a) and induced-temperature change in thickness direction (b).

As shown in Figure 99(b), by decreasing the κ from 0.3 to $0.05 \text{ (nm}^{-1})$, the amplitude of frequency at 0.625 GHz (Figure 99(a)) decreases to $\sim 0.12 \times 10^{-4}$ where the induced-temperature change also effects the far end of the optical cavity and the temperature is assumed to be distributed inside the cavity. There is also a high frequency peak observable around 2 GHz in comparison with the sample using higher concentrated dye in CA medium. As can be seen, the temperature can be equally distributed within the cavity in order to obtain a frequency of interest.

Various coating solutions can be used to coat the layers in a multi-layered stack. This is mainly achieved by the functional superposition of several deposited film. By changing the number of coating cycles and the concentration of various coating solutions, the thickness is not the only parameter that can be well controlled, but the optical constants can be controlled as well [77]. This fabrication method is also simple and durable to fabricate a multi-functional synergy of each deposited layer, and this multi-layer sputter assisted dip-coating process broaden benefits.

The mechanical model is simulated for a multi-layered stack with 540nm thickness by superposition of five layers of CA thin films with the thickness of 105nm plus five

layers of ITO between each layer of CA with thickness of 3nm. The pump of 728nm and probe of 860nm were set in the simulation. The results are shown in Figure 100. The extinction coefficient in all layers is $0.1 \text{ (nm}^{-1}\text{)}$ which produces a frequency of $\sim 0.65 \text{ GHz}$ with amplitude of ~ 0.042 (Figure 100(b)). However high frequency signals also can be observed and the oscillation decays after a couple of cycles, and vanishes before 15ns (Figure 100(a)). The temperature is also distributed within the optical cavity (Figure 100(d)). In addition a very strong strain is observed in the cavity (Figure 100(c)) which can be counted as a limitation in the process.

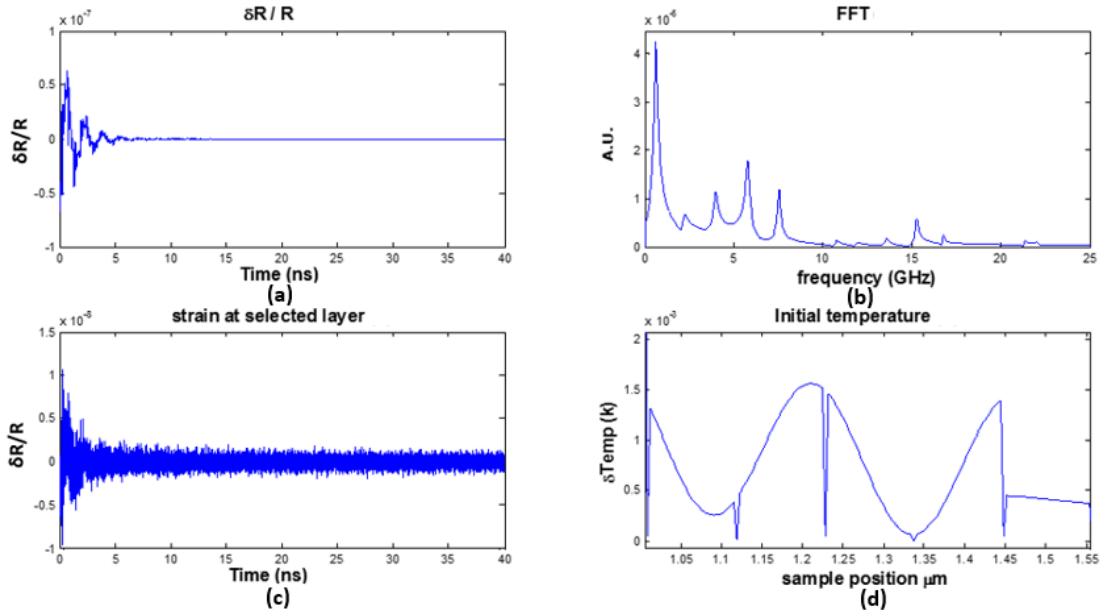


Figure 100: The graphs present mechanical simulation of 540nm of multilayer CA with 3nm of ITO protective layers (all together 553nm), modulation depth (a), FFT of the modulation depth (b), strain in selected layer as a function of time(c), and the temperature change of the selected layers (d).

It should be noted here that the x-axis of the initial temperature graph in Figure 100(d) is in micrometre (μm) unlike other graphs presenting the sample position in thickness direction. This is because in this case $1\mu\text{m}$ was counted as the substrate thickness in the numerical model and the transducer is positioned on the top of the substrate as shown in Figure 101. However, the position of substrate has been eliminated for displaying purposes in the graph as we are interested in the temperature distribution within the transducer.

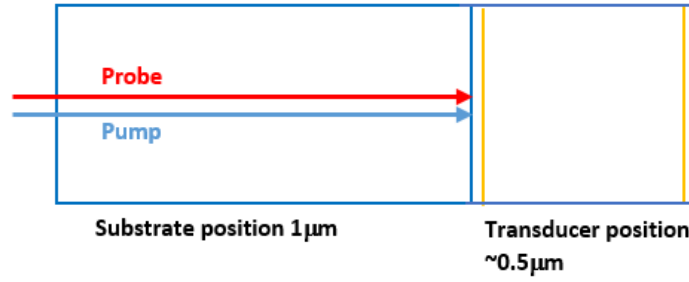


Figure 101: The position of the transducer in thickness direction after substrate position.

Figure 102 shows the mechanical model simulation of the same multi-layered stack but the thickness of ITO thin films are decreased from 3nm to 2nm. The optical constant κ is only changed for the last layer of the stack from 0.1 to 0.3 (nm^{-1}). The amplitude of the calculated frequency (0.65 GHz) (Figure 102(b)) is significantly increased from 0.042 to 0.08. This infers that the transducer will ring for a longer time (≥ 40) (Figure 102(a)) and higher frequencies are observable. The pattern of temperature distribution (Figure 102(d)) is relatively the same with the previous case (Figure 100(d)). A strong strain also can be seen in the cavity (Figure 102(c)).

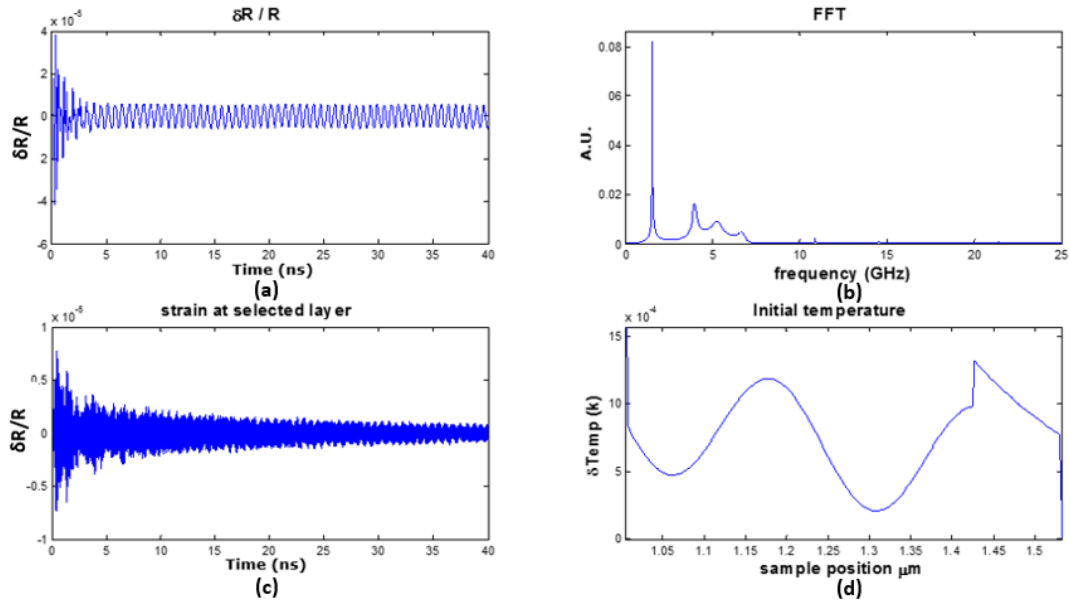


Figure 102: The graphs present the mechanical simulation of 540nm of multilayer CA with 2nm of ITO protective layers, modulation depth (a), FFT of the modulation depth (b), strain in selected layer as a function of time(c), and the temperature change of the selected layer (d).

As a summary, from the simulated mechanical models, it is possible to produce a desired frequency and control the amplitude of the frequency by changing several parameters such as thickness of layers or/and protective layers (ITO) as well as optical constant of the layers in the multi-layered stack. However, a continuous wave (CW) laser beam is needed in order to detect δR since in some cases the transducers are ringing for longer than the pulse duration of the current laser system (12ns) (Figure 102(a)).

6.3 Hybrid device

In 2014, Ding and the group reported that by increasing the concentration of absorptive dye and decreasing the thickness of thin films to $dn \approx \lambda / 8$, the doped polymer thin films showed a high (>90%) and identical absorption magnitude [78]. They used polyvinyl alcohol (PVA) thin films doped with Rhodamine 6G (R6G) molecules coated on opaque silver (Ag) substrates [76][78]. They suggested that the thin film system can be used to flexibly tune the spectral and directional range of absorption by doping with proper absorbing media and setting appropriate excitation conditions of FP resonances [78].

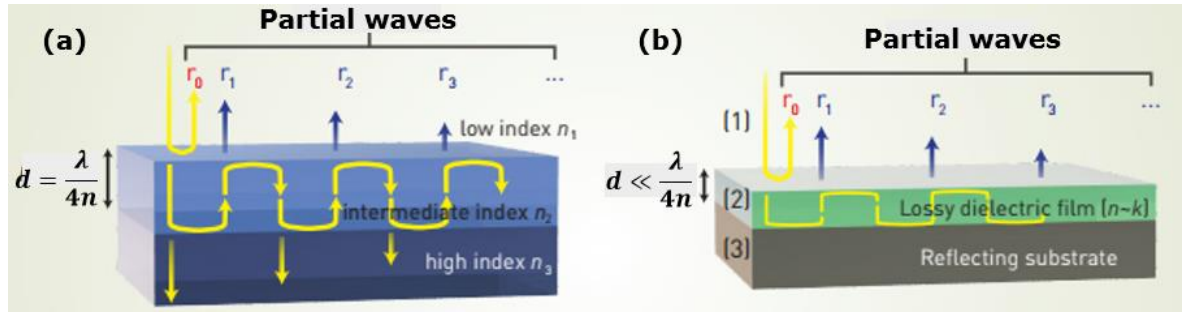


Figure 103: Schematics and phasor (Tischler et al., 2006 [79]) for (a) a conventional quarter-wave anti-reflection coating, and (b) an absorber using an ultra-thin lossy layer on a reflecting substrate. The figure has been.

An ultra-thin absorber fabricated on top of a reflecting substrate can be considered as an absorbing anti-reflection (AR) coating. Therefore, a basic operating principle of a simple AR coating is instructive for these type of coatings, by which R is minimized and T is maximized at normal incidence, given that both the substrate and superstrate

are transparent: a quarter-wave film with $n_2 = \sqrt{n_1 n_3}$ and $d = \lambda/4n_2$ [77]. In this AR coating, there are no losses of light, and the film is designed such that destructive interference within the structure causes the suppression of reflection. This can be understood by decomposing the incident light into partial waves (Figure 103(a)), plotting the amplitude and phase of the reflected partial waves r_0, r_1, r_n in complex space (Figure 103 (b)). The destructive interference occurs due to the 180 degree phase difference. Given a lossless film, substrate, and superstrate, $h = \lambda_0/4n_2$ is the thinnest film that yields a minimum reflectance [78][79].

I proposed the new configuration of FPI in this study where the absorbing dielectric layer works as AR coating as shown in Figure 104(b). The diagram of such a structure is very similar to that of Figure 103(b), with the key difference being that absorption in the film replaces the loss channel of out-coupling via the bottom interface (i.e. transmission). This effect can be through coupling to a resonance of the asymmetric Fabry Perot (FP) cavity formed by the top and bottom interfaces, where the light is gradually absorbed as it circulates.

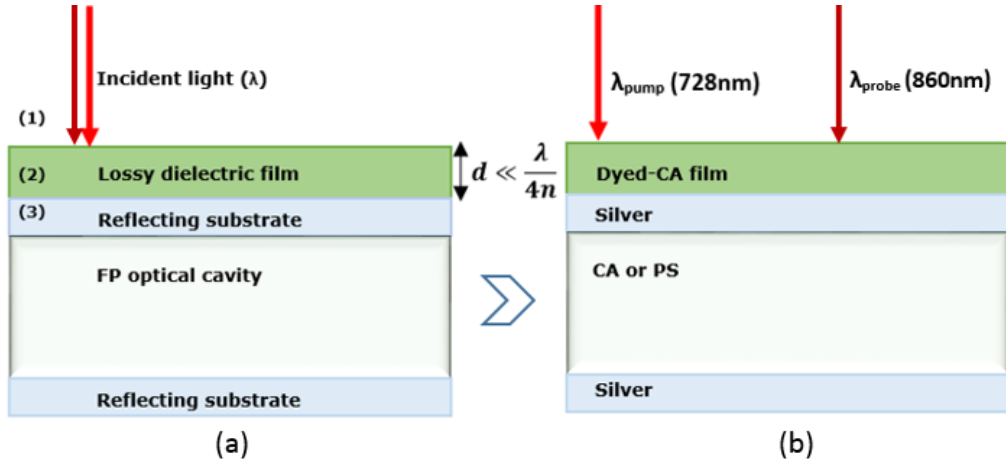


Figure 104: Schematic of a hybrid device with a dielectric absorbing film (dye-CA) with silver reflector layers (a) and an example of such a hybrid device (b). 728nm of pump can be used for excitation and 860nm of probe for detection.

In order to increase the transmission rate at 860nm, another metallic layer can be considered to replace with the gold layer where the absorption at metallic layer is not a case for generation of acoustic waves. For this, the transmission spectra for a

250nm of CA with 20nm of silver and gold layer were stimulated to observe the transmission at 860nm wavelength (the wavelength of interest for the probe).

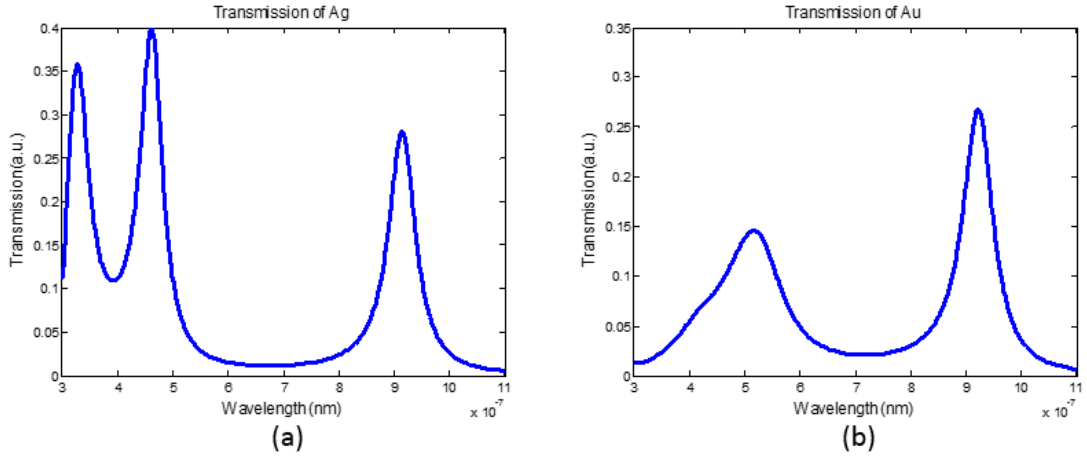


Figure 105: Simulated transmission spectra of silver and gold.

The simulated transmission spectra of gold and silver are shown in Figure 105(a). The corresponding simulated spectra of silver thin film shows a maximum transmission (~ 0.6) spectra at $\sim 860\text{nm}$ (probe wavelength), where the low transmission occurs at 728nm (pump wavelength). Gold thin film shows less transmission around 860nm (~ 0.4) in comparison with silver which makes silver a more suitable metal for the probe laser beam. Silver can be also used in hybrid device as a reflecting substrate due to its high transmission efficiency at the probe wavelength (Figure 105(b)).

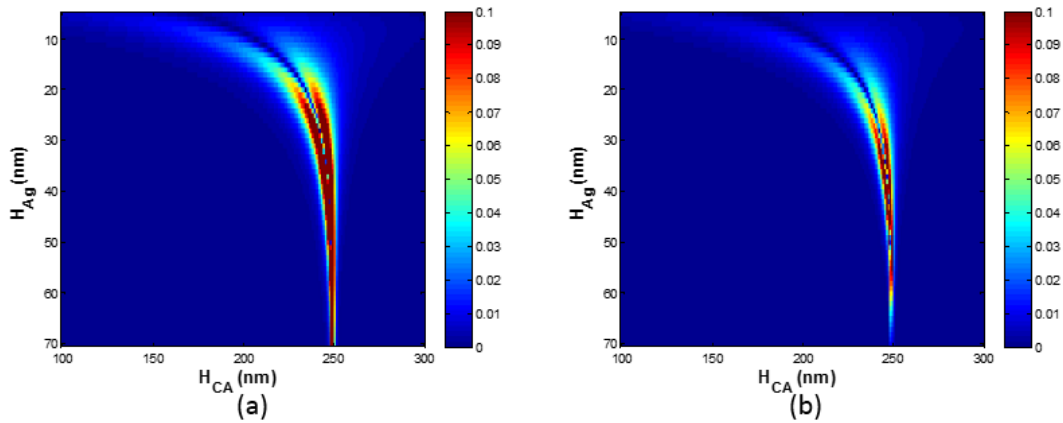


Figure 106: Transducer sensitivity $\delta R/\delta H$ (a) $\delta T/\delta H$ (b) at λ_{probe} for a range of layer thicknesses of CA with silver reflector layers. The colour bars present sensitivity.

Figure 106 shows the optical model for a wide range of silver (5-70nm) and CA (200-300nm) thicknesses to investigate a transducer with 240nm CA layer and 20nm of silver layers. The simulation was performed at 860nm of the probe which shows a relatively high sensitivity both in transmission (Figure 106(a)) and in reflection (Figure 106(b)). The optical model for the mentioned transducer was also simulated to investigate the sensitivity of the transducer at 860nm of the pump in the reflection, transmission, and absorption (Figure 107). The transmission in Figure 107(b) and reflection in Figure 107(c) of the transducer are about 0.40 and 0.34, respectively. As can see in Figure 107 (a), the transducer shows a low absorption rate at 860nm, where a relatively good transmission (Figure 107(b)) occurs within the transducer at this wavelength. Figure 107(c) shows the transducer becomes a partial reflector at 860nm of pump. A maximum absorption at 728nm was considered in all simulations.

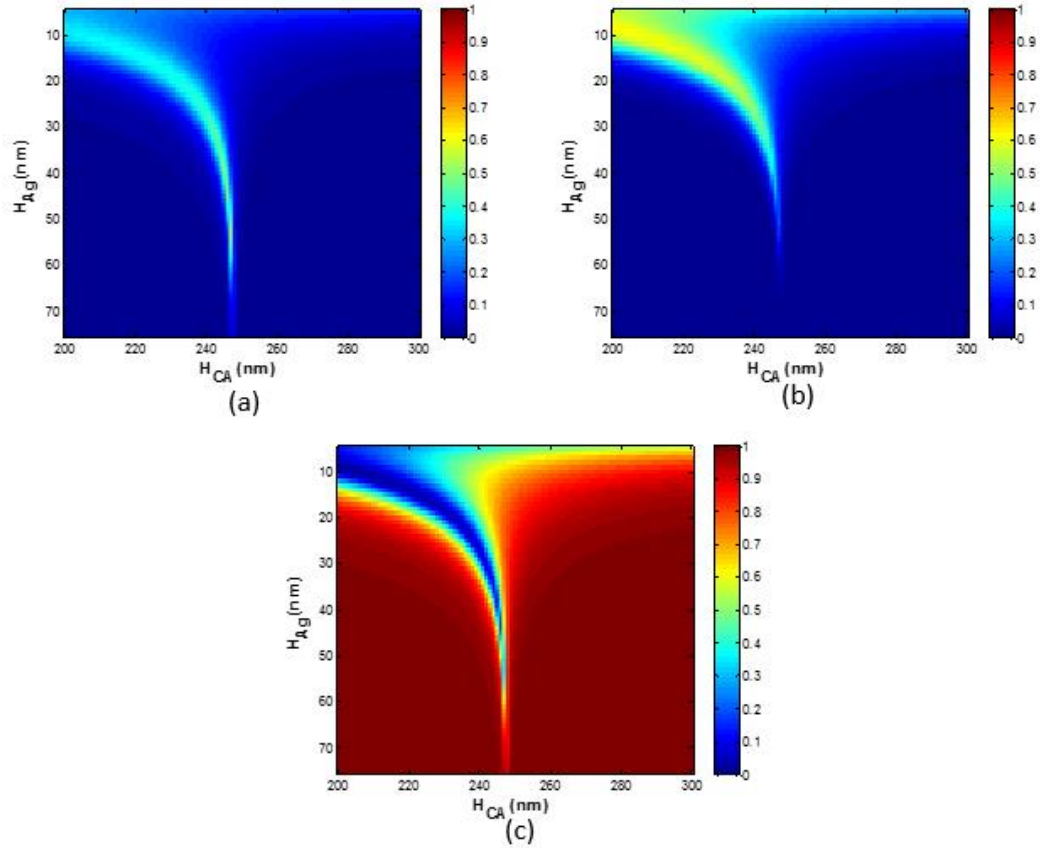


Figure 107: Result of the optical model for the pump wavelength (860nm). The colour bar shows the percentage of light absorbed by the transducer (a), its related transmittance (b), and reflectance (c) at λ_{pump} in a range of thicknesses.

The mechanical simulation for a hybrid device (Figure 108) with a lossy dielectric dye-CA layer before the first reflector layer of silver is shown in Figure 109. The simulation was performed for 728nm of the pump and 860nm of the probe. The thickness of dielectric film is 240nm with 0.3 nm^{-1} absorption coefficient in the hybrid device. The thickness of the silver reflector substrate is 20nm with a CA cavity without absorption at pump wavelength.

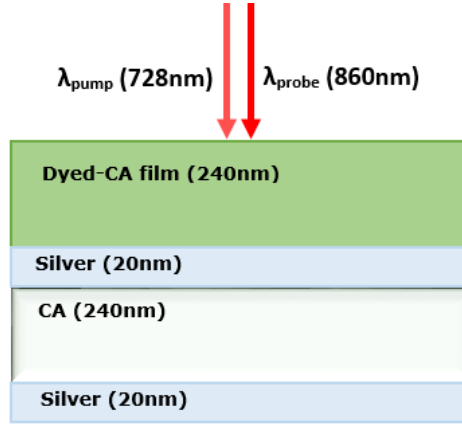


Figure 108: Schematic of a hybrid device with a dielectric absorbing film (dye-CA) in the size of spacer with silver reflector layers. The pump at 728nm and the probe at 860nm of the probe are used for the excitation and detection, respectively.

The frequency of 2.2 GHz with amplitude of 6.5 (mV) (Figure 109 (b)) can be observed and the oscillation is almost completely decayed before 12ns (Figure 109 (a)). There is another high frequency peak at 0.75 GHz with a lower amplitude than main peak which can be the Brillouin oscillation from dye-CA dielectric layer during detection. Figure 109 (c) shows that the temperature is affected only within the dye-CA dielectric layer which is mainly caused by the high optical absorption due to absorptive dye in medium (maximum absorption at 728nm).

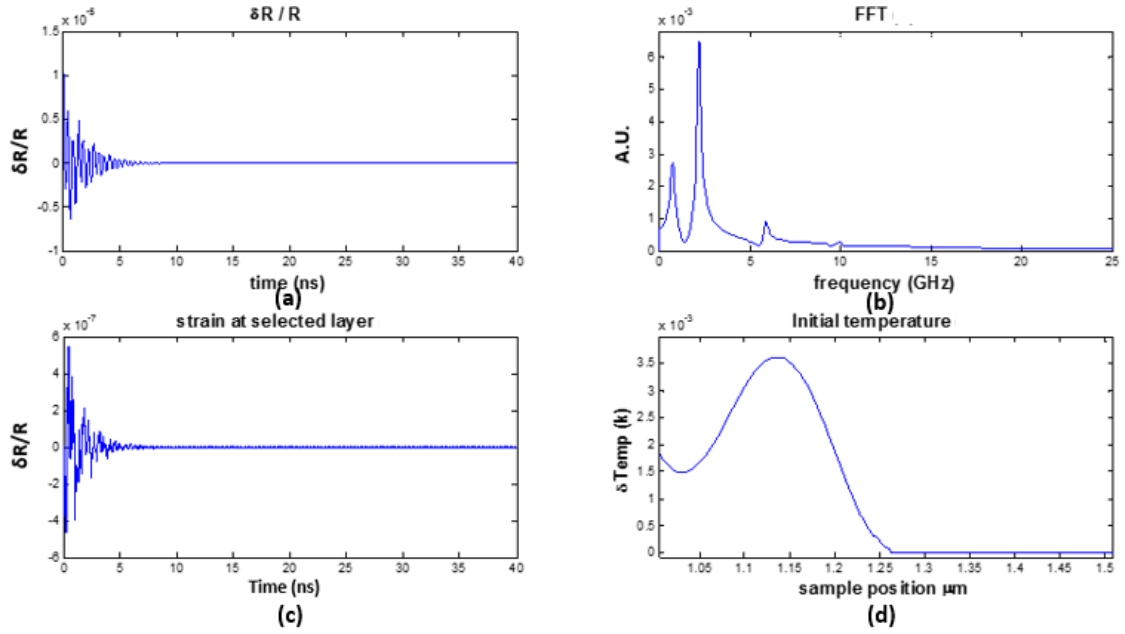


Figure 109: The mechanical simulation of a hybrid device with the dielectric absorbing layer equal to spacer with 240nm thickness. The graphs present modulation depth (a), FFT of the modulation depth (b), strain in selected layer as a function of time(c), and temperature profile of the selected layer (d).

The mechanical simulation for another hybrid device with same configuration (Figure 110) with different thickness of 30nm of a lossy dielectric dye-CA layer is shown in Figure 109. The simulation was performed at 728nm for the pump and 860nm for the probe. The thickness of dielectric film is relatively thinner than the optical cavity with higher extinction coefficient ($0.6 \text{ (nm}^{-1}\text{)}$) in the hybrid device.



Figure 110: Schematic of a hybrid device with a dielectric absorbing dye-CA layer with 30nm thickness and CA-based spacer. The pump at 728nm and the probe at 860nm are used for excitation and detection, respectively.

The thickness of the silver reflector substrate is 20nm with a CA cavity without absorption at pump wavelength. The frequency of 1.2 GHz with amplitude of ~ 2 (Figure 111(b)) can be observed. There are also higher frequency peaks present which can be due to the Brillouin oscillation from dye-CA dielectric film during detection. Since the thickness of the dye-CA dielectric absorbing layer is relatively thin, the temperature is assumed to be distributed in the layer and is exponentially decreasing in the dye-CA layer direction (Figure 111 (d)). The strain is stronger in comparison with the pervious case (Figure 109(c)). The presence of high frequency components can be due to ongoing strains (Figure 111 (b)).

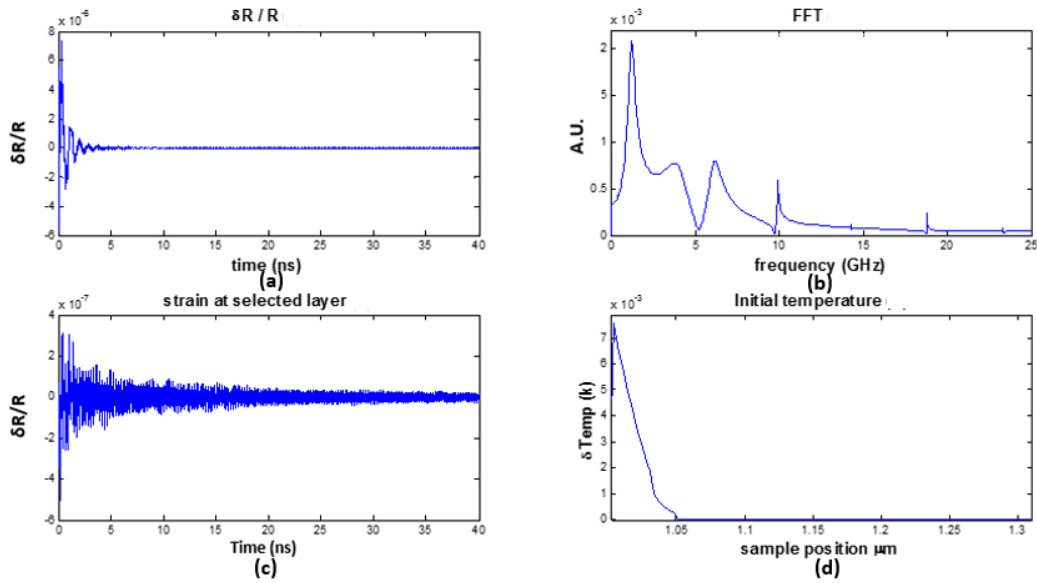


Figure 111: The mechanical simulation of a hybrid device with 30nm dielectric absorbing layer. The graphs present modulation depth (a), FFT of the modulation depth (b), strain in selected layer as a function of time(c), and temperature distribution of the selected layer (d).

As previously described, in multi-layered deposition of thin films using layer by layer method, it is possible to use different coating solutions within the structure of the transducers. One of the options in hybrid device is to use PS in spacer instead of using the same material of the dielectric film. In this case dye-CA polymer thin film is used only for generation of strains within the spacer with $0.3 \text{ (nm}^{-1}\text{)}$ of extinction coefficient optical constant. Also, the thickness of the PS is assumed to be the same as dye-CA dielectric absorbing film (380nm).

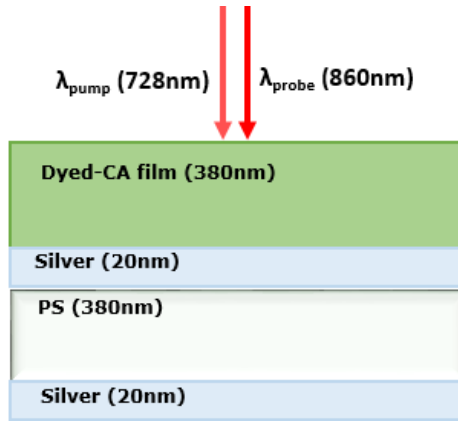


Figure 112: Schematic of a hybrid device with a dielectric absorbing dye-CA layer and a PS-based spacer. The pump at 728nm and the probe at 860nm are used for the excitation and detection, respectively.

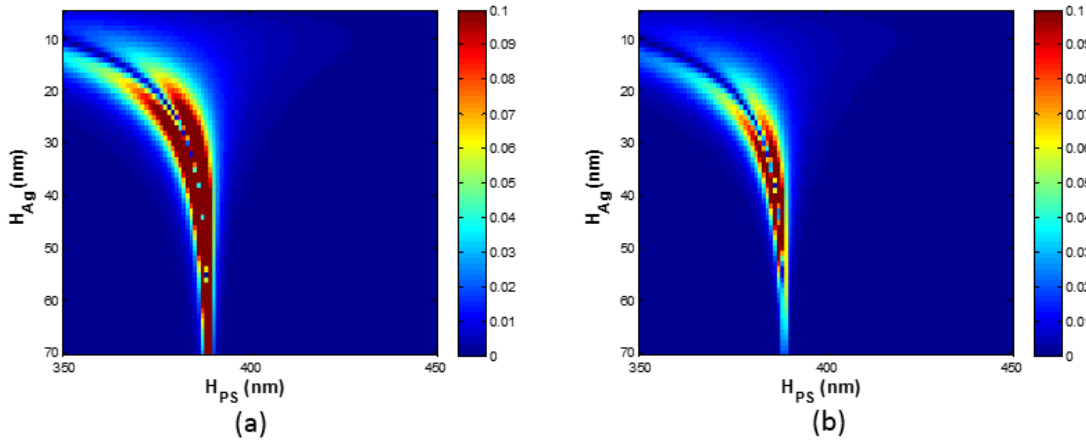


Figure 113: Transducer sensitivity $\delta R/\delta H$ (a) $\delta T/\delta H$ (b) at λ_{probe} for a range of layer thicknesses of PS with silver reflector layers.

The optical simulation of such a device shows a high sensitivity ($0.072 \text{ (nm}^{-1}\text{)}$) at 860nm of probe in both reflection and transmission of 380nm of PS spacer and 20nm of silver layers (Figure 113). The mechanical simulation also shows a frequency of 0.55 GHz with the amplitude of ~ 0.07 can be observed and the high frequencies are negligible. A noisy oscillation can be also observed but it seems after a couple of the cycles, the oscillations start to decays significantly before 15ns.

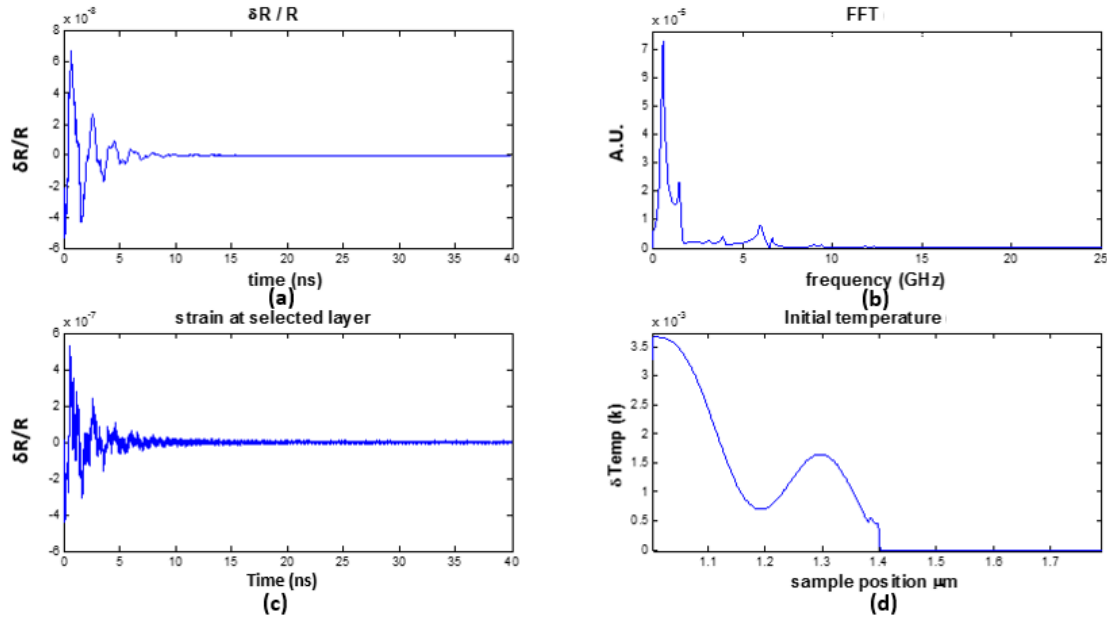


Figure 114: The mechanical simulation of a hybrid device with the dielectric absorbing layer dye-CA and PS spacer with the thickness of 380nm and 20nm of silver layers. The graphs present modulation depth (a), FFT of the modulation depth (b), strain in selected layer as a function of time(c), and temperature distribution of the selected layer (d).

A hybrid device with different configuration can be designed by placing the dielectric absorbing layer after the second reflector layer, but a low amount of light can be transmitted to the far end of the transducer. There is also a chance of interference of acoustic waves generated by this layer with strain in spacer or with light resonance inside the spacer. It seems from the simulated models, the hybrid device with configuration shown in Figure 112 will offer higher amplitude with less unwanted high frequency signals and distributed temperature in the absorptive layer. The cycles of oscillations are also decaying before 15ns. In addition, different materials can be used in this configuration as a spacer. However, it is the first time that such device with this particular configuration has been suggested in generation and detection of acoustic waves using FPI.

6.4 Potential of needle-based ultrasound diagnostic tool in aerospace industry

Perez Cota from Nottingham University in 2015 [109] presented the plate transducers for high resolution real-time imaging at molecular level and my study was the extension of Perez Cota's study to detect Brillouin scattering in reflection technique.

A needle-based diagnostic imaging tool can be used for inspection of cooling holes in a turbine blade of jet engine (Figure 115). Film cooling in turbine blade keeps the temperature of the jet engine turbines at an acceptable level. The cooling holes in the turbine blade discharge cooling air from the interior structure of the turbine blade onto the surface [110].

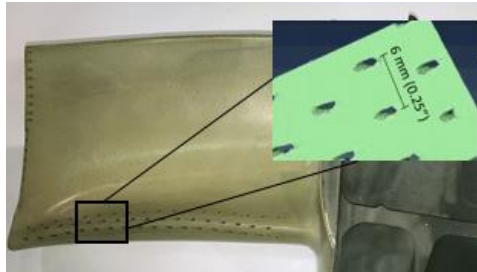


Figure 115: The cooling holes in a jet-engine turbine blade [110].

Any blockage of the holes in a turbine blade causes a serious damage to its material due to raising temperature above the melting point of the blade. The cooling holes must be inspected both in the manufacturing process and during engine reconditioning. The diagnostic imaging tool at micron-level inspection offers significant value and capabilities to both the aerospace and power generation industries. The small-size of the transducer fabricated on the tip of an optical fibre can be used to inspect and observe the film cooling of the interior surface of a blade.

Chapter 7

Conclusions

Designing extremely small-sized photoacoustic transducers can be important in the medical field in order to obtain information that is hard to access externally and requires major invasive procedures. The device can be used *in vivo* biopsy in order to prevent of unnecessary biopsies which can help to reduce the cost of healthcare. It also can be used to monitor cancer therapy, study the progression of disease in vivo as well as microsurgical guidance.

Acoustic waves with frequencies from several GHz to hundreds of MHz can be optically detected using FPI. This method of detection provides an alternative to broadband piezoelectric transducers. The sensing element requires a very small-sized, thin film spacer surrounded by two optically reflective metallic coatings. In a planar sensing structure, the lateral dimensions must be on a sub-millimetre scale in order to work with acoustic waves at MHz frequencies and above. The fundamental advantage of this type of acoustic detection transducer is high sensitivity and significantly smaller structure of the sensors in comparison with piezoelectric receivers. The intention of this study was to fabricate these transducers on the tip of an optical fibre in order to optically generate as well as detect acoustic waves radiated from the same fabricated transducer.

7.1 Summary of thesis

Development of transducers

Three layer thin film FPI transducers were fabricated both as fibre-based and as the plate transducers. In my study, I demonstrated several approaches to produce a wide range of frequencies in the prospect of bandwidths extending from several GHz to hundreds MHz frequency range by using different materials, and increasing the spacer thickness which require different fabrication methods with fabrication tolerance of $\pm 5\text{nm}$. The characterizations of the transducers were carried out by fibre-based and free-space transmission spectroscopy setups. Brillouin scattering was the main detection mechanism that was used in this work. A sets of thicknesses, both for

reflectors and optical cavity were determined to the best sensitivity in designing a transducer for detection of Brillouin scattering in reflection.

A pump and probe technique was used for excitation and detection of ultrasound. An ultra-short pulse laser unit emits the pump and probe laser pulses synchronously with a 12ns time delay. A maximum 1-1.5 mW output power of the pump (415 nm) and 4 mW average output power of the probe (780 nm) were used to generate and detect acoustic waves in fibre-based ITO. In polymeric transducers the output pump power was reduced to about 0.5 and 0.3 mW to irradiate the PS and CA devices, respectively.

Fabrication of the transducers

The transducers were fabricated using different methods based on materials to deposit the layers in the transducers. The ITO and gold layers in the three layer transducers were deposited using sputter coater. This allowed very fine control over the devices made, easy monitoring of the deposition of the layers in the devices and fine-tuning the design and fabrication process. Spin coating and dip-coating also were used to deposit the polymeric coating solutions for higher order devices (thicker). The transducers were deposited on the glass substrate either on the tip of single-mode fibre (4.6µm core diameter) or on the top of glass microscope slides. The layers in the transducers were characterized using custom made transmission spectroscopy system both as the fibre-based and free-space setups. The experimental transmission spectra were compared to the theoretical spectra. The similarity between each theoretical curve and experimental one is scored from 0 to 1: $\text{Score} = 1 - |T(\lambda)_{\text{model}} - T(\lambda)_{\text{exp}}|$. A good match to the obtained value presented the thickness of the layers.

Experimental results

In this study, the generation and detection of acoustic waves was performed through the same transducer. In Chapter 4, the theoretical and experimental results were presented for the Indium Tin Oxide (ITO), Polystyrene (PS) and Cellulose acetate (CA) based devices. The opto-thermo-mechanically coupled model was used to predict the behavior of the transducer when interacting with the pump and probe

laser beams. This allowed to optimize design for a particular operating frequency range.

The Indium Tin Oxide and Polystyrene transducers

It was demonstrated that by increasing the thickness of ITO spacer in a FPI fabricated on the tip of a single mode fibre from about 150nm to 560nm (as the first and second order ITO), the frequency decreased from ~ 9.8 GHz to ~ 4.9 GHz, respectively. The detection was performed in transmission and reflection mode with different excitation configuration setups. By replacing material from ITO to PS for a third order and higher order device, we demonstrated that frequency reduced from 550 MHz to 430 MHz for a 560nm and 931nm PS devices, respectively. In this case also the detection was performed in reflection mode with a different excitation configuration setup.

Characterization of Cellulose Acetate

As a summary it was concluded from a series of experiments in this section that the solvent used during the preparation of thin polymer films via spin-coating has a high influence on the resulting morphologies. Solvents with low vapour pressure such as DMSO were used in preparation of CA-based thin films, whereas the morphologies give rise to a large surface roughness and nonhomogeneous polymer films in high concentrated of CA solutions at low spin speed using solvents with high vapour pressure such as acetone. Thin films were prepared using a mixture of solvents acetone and DMSO showed a wavy pattern in comparison to the ones prepared out of weakly volatile solvents. This suggested that one way to increase the thickness with even smoother CA polymer films was to use layer by layer deposition method via solvents with low vapour pressure. However, to increase the thickness of a thin CA film several parameters have direct effect such as ambient temperature, medium concentration, and sample preparation as well as using a certain solvent. By changing the number of coating cycles and the concentration of various coating solutions, the thickness and uniformity of the thin films can be well controlled.

Tailoring a transducer based on the available wavelength for the pump laser

A tunable absorbing nanoscale thin-film system was developed experimentally and numerically, comprising of dye molecules mixed in dielectric coatings on reflecting surfaces. In this part of the study, it was demonstrated for the first time the manipulation of light absorption to tailor the wavelength for pump laser beam in order to generate acoustic waves. Specifically, CA thin films mixed with absorptive dye with the maximum absorption at 748nm coated on opaque gold substrate were used. It was demonstrated that the absorption behaviours of dielectric thin films can be flexibly tuned by adjusting the proper system parameter such as dye solution concentrations and the thickness of dielectric thin film layer coating. However more experiment were needed to be conducted in order to generate acoustic waves by this method.

Closing remarks

The significance of this study resides in the demonstration of generation and detection of acoustic waves through the same FPI transducer fabricated on the tip of a single-mode fibre. It is feasible if two laser beam for pump and probe can be delivered in the same fibre. It was also demonstrated that the frequency can be produced in a wide range from several GHz to the hundreds of MHz range by either changing the martial or increasing the size of optical cavity in the FPI transducers. The approach presented in this study has potential to provide new insight in generation of acoustic wave through the optical cavity in the FPI transducers rather than the through the metallic reflector layers. This study has been presented in multiple national and international conferences (AFPAC2016, OPUS2016, LU2016, and IOP2016) and currently there are more works under study by other colleagues in the group.

Appendix

A.1 determination of optical properties of materials: Absorption and Extinction coefficients

The optical properties of materials including absorption coefficient (α) and extinction coefficient (κ) can be obtained by interacting them with electromagnetic radiation. The intensity of light is calculated by Lambert Beer-Bouguer law (1) when a beam of light is incident on a material [13]:

$$I = I_0 \exp(-\alpha d) \quad (33)$$

If absorption in a material takes place, it appears that the optical intensity of the light wave (I), is exponentially reduced (in respect to the cavity thickness) while travelling through the film (Figure 116).

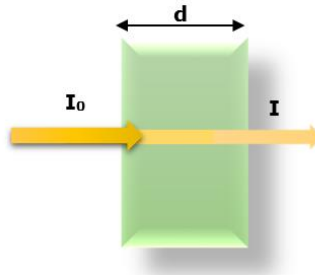


Figure 116: Light intensity directing forward to a media and traveling the distance of media and the intensity is decreased as the light travels through the media.

The equation (41) can be rearranged in order to calculate the absorption coefficient using from the power of light is shone at the film (I_0) and the power of transmitted light (I) and the film thickness (d):

$$\alpha = -\frac{1}{d} \ln (I/I_0) \quad (34)$$

The extinction coefficient (κ) describes the properties of the material with respect to light of a given wavelength and indicates the absorption changes when the electromagnetic wave propagates through the material. The extinction

coefficient is the imaginary part of complex refractive index and can be calculated by the following equation after calculating absorption coefficient [14].

$$k = \frac{\lambda\alpha}{4\pi} \quad (35)$$

Where λ presents the wavelength of the incident light. The evaluated values for extinction coefficient for a wide range of wavelengths from 300nm to 900nm for different dye concentrations in medium were used in optical model.

A.2 Matlab Scripts

A.2.1 Signal processing

```
close all
clear all

%finding my signal from my raw data
addpath '/home/mitra/ultorasonud/2016/optical model of 780 of thick
stacks/experimental data/'
addpath /home/mitra/MATLAB/mydatafile/

%% loading data

[waveformAC,timebase]=loadwf('water2_100k_ac1.wf');% loading data
[waveformDC,timebase]=loadwf('water2_100k_dc1.wf');%

AC=mean(waveformAC);%get the mean from the waveform (100 traces with
2500 points)
DC=mean(waveformDC);

data_load = load('data602.mat');

peak = data_load.peak;
T= data_load.t;
A = data_load.a;

%% diffining dT/T on time domain. The time is converted to the real
time base in this section

mysignal=AC./DC;%is the voltage in y axes-dividing point by point
dT/T
new_timebase=timebase*1.2500e-04;%converting to the reall time bas
```

```

%% removal of thermal expansion. For this, a polyfit is needed to
calculate my polynomials (p1,p2,p3,p4) and a polyval is needed to
calculate my new_y

y=AC(230+20:800);%extracting my signal from raw data which gonna be my
trace cut
n=7; %the order number of best curve fit
N=2^12;%zero pads to have more points
x=new_timebase(230+20:800)*1e9;%new timebase in ns
X=x-x(1);

subplot(3,1,1);
plot(new_timebase,mysignal);
xlabel('Time(ns) ');
ylabel('dT/T');
title('Raw trace ');

%Low pass filter

p=polyfit(x,y,n);%calculating p
tracepoly=polyval(p,x);%calculating new y or our tracepoly
trace_oscillation=y-tracepoly;%remove the thermal expansion

windowSize=5;
b = (1/windowSize)*ones(1,windowSize);
a =1 ;
Y=filter(b,a,trace_oscillation);

% subplot(3,1,1);
% plot(x,trace_oscillation,'b');
% xlabel('Time(ns) ');
% ylabel('dT/T');
% title('Raw trace ');

subplot(3,1,2);
plot(X/1e9,Y./max(Y), 'b');
hold on
plot (T,A./max(A), 'r');
xlabel('Time(ns) ');
ylabel('dT/T');
title('processed trace');
hold off

%% doing fft
trace_f=abs(fftshift(fft(Y,N)));%doing fft on the data and doing
ftta=abs(fftshift(fft(A,N))); % data from theoretical

%% calculating fs for freq axis
newtime=(1:N)*(new_timebase(2)-new_timebase(1)); %new timebase with
zero pads added to tails. This part is diffing a pixel
nt=length(newtime);

```

```

dt=newtime(2)-newtime(1);
df=1./(nt.*dt);
fm=0.5./dt;
fs=-fm;
f=linspace(fs,fm-df,nt); % the f units is Hz in this data

NewTime=(1:N)*(T(2)-T(1)); %new timebase with zero pads added to tails
for theorical data
nT=length(NewTime);
dT=T(2)-T(1);
dF=1./(nT.*dT);
Fm=0.5./dT;
Fs=-Fm;
Fa=linspace(Fs,Fm-dF,nT); % the f units is Hz in this data

% f=xtof(t);

%% windowing filtering my data using low pass filter hann window from
f-cut of 50
p2=find(f>50.*1e9,1); % finding all points smaller than 50 GHz and
gives the first number
p1=find(f>-50.*1e9,1); % finding all points bigger than -50 GHz. Because
the "f" was based on Hz, we need to convert it to GHz in here

P2=find(Fa>50.*1e9,1); % finding all points smaller than 50 GHz and
gives the first number
P1=find(Fa>-50.*1e9,1);

f2=f(p1:p2); % f2 is a defined new frequency for points p1 and p2 "
f2=f(p1:p2)" gives all f between p1 and p2 but if put zero pads instead
of p1, it throw out the first half before 0
trace_f2= trace_f(p1:p2);
hannWid=hann(p2-p1+1);
dataWid=trace_f2.*hann(p2-p1+1).';

F2=Fa(P1:P2); % f2 is a defined new frequency for points p1 and p2 "
f2=f(p1:p2)" gives all f between p1 and p2 but if put zero pads instead
of p1, it throw out the first half before 0
trace_F2= ffta(P1:P2);
hannWid=hann(P2-P1+1);
dataWid=trace_F2.*hann(P2-P1+1).';

f_cut=50;
[dataWid] = Widfunction(f,trace_f,f_cut);
windowSize=5;
b = (1/windowSize)*ones(1,windowSize);
a =1 ;
F=filter(b,a,dataWid
F_cut=50;
[DataWid] = Widfunction(Fa,ffta,F_cut);
windowSize=5;

```

```

b = (1/windowSize)*ones(1,windowSize);
a =1 ;
F1=filter(b,a,DataWid)

subplot(3,1,3);
plot(f2/1e9,F/max(F),'b');
hold on
plot(F2/1e9,F1/max(ffta),'r');
xlabel('Frequency (GHz)');
axis([0 50 0 1]);
ylabel('dT/T');
title('Processed spectrum');

hold off

%print('-dpng','comparison of data for ITO')

```

A.2.2 Absorption and Extinction coefficients calculations

%% in this script, the data are loaded and plotted as a function of wavelength.
 %The spectra then are normalised by bulb or reference. The glass substrate is a reference in this case.
 %The absorption coefficient (a) then calculated by using thickness of thin films. Extinction coefficient then is calculated using absorption coefficient and plotted as a function of wavelength.

```

clear all
close all
addpath /home/mitra/matlab/high_order_device/
% Sample=load('bulb.txt');
addpath E:\matlab\whole_stack
%% loading data and plotting row data
Sample1=load('ref.txt');
Sample2=load('ca_30_20_1500.txt');
Sample3=load('ca_30_20_2500.txt');
Sample4=load('ca_30_20_3300.txt');
%bulb = load('ca_bulb.txt');
sample1=Sample1(:,2);%./max(bulb(:,2));
sample2=Sample2(:,2);%./max(bulb(:,2));
sample3=Sample3(:,2);%./max(bulb(:,2));
sample4=Sample4(:,2);%./max(bulb(:,2));
%sample=Sample(:,2);%./max(bulb(:,2));
%bulb = bulb(:,2);%./max(bulb(:,2));
W=Sample1(:,1);
%fudge factor to get ~unity transmission out of the absorption band -
matt
norm2=01;norm3=1;norm4=1;

```

```

subplot(3,1,1)
plot(W,sample2,'cyan')%*norm2)
hold on
plot(W,sample3*norm3,'r')
plot(W,sample4*norm4,'b')
plot(W,sample1,'green')
hold off
xlabel('Wavelength(nm)', 'fontsize',12,'FontWeight','bold');
ylabel('Transmittance (a.u.)', 'fontsize',12,'FontWeight','bold');
axis([450 1020 0 6*1e4])
title('Transmission spectra of dye-CA thin
films', 'fontsize',12,'FontWeight','bold');
legend ('1500 rpm','2500 rpm','3300 rpm','control')
%% normalising spectra by ref

nor_sample2=sample2./sample1*norm2;
nor_sample3=sample3./sample1*norm3;
nor_sample4=sample4./sample1*norm4;

subplot(3,1,2)
plot(W,nor_sample2,'Cyan')
hold on
plot(W,nor_sample3,'r')
plot(W,nor_sample4,'b')
hold off

xlabel('Wavelength(nm)', 'fontsize',12,'FontWeight','bold');
ylabel('Intensity (u.a)', 'fontsize',12,'FontWeight','bold');
axis([550 850 0 1.1])
title('Normalised transmission
spectra', 'fontsize',12,'FontWeight','bold');
legend ('1500 rpm','2500 rpm','3300 rpm')

%% absoprtion coefficient calculation
t_sample2=845e-9*input('thickness of sample2:');
t_sample3=470e-9*input('thickness of sample3:');
t_sample4=380e-9*input('thickness of sample4:');

abs2=(-1./t_sample2).*(log(nor_sample2));
abs3=(-1./t_sample3).*log(nor_sample3);
abs4=(-1./t_sample4).*log(nor_sample4);
subplot(3,1,3)
plot(W,abs2,'cyan');
hold on
plot(W,abs3,'r');
plot(W,abs4,'b');
hold off
xlabel('Wavelength(nm)', 'fontsize',12,'FontWeight','bold');

```

```

ylabel('Absorption coefficient (nm^{-1})','fontsize',12,'FontWeight','bold');
%axis([550 850 0 7*1e-4])
xlim([550 850])
title('Absorption coefficient of dye in CA solution','fontsize',12,'FontWeight','bold');
legend('1500 rpm','2500 rpm','3300 rpm')

%% calculation of extinction coefficient and using a low pass filter
raw_data_abs=(abs2+abs3+abs4)/3
mean_data_abs=raw_data_abs(300:1500);
W0=W(300:1500);
w=W0*1e-9;
i=1:length(w);
k0(i)=(mean_data_abs.*w)/4*pi;
x=k0;fc=0.1/1e1;t=w
[data_filt]=filt_low(fc,x,t);

k_20=(data_filt)

figure;
plot(w,k_20)
xlabel('Wavelength(nm)');
ylabel('Extinction coefficient(nm^{-1})');
xlim([5.5*1e-7 8.5*1e-7])
%title('Extinction coefficient vs. wavelength','fontsize',10);

```

A.2.3 Low pass filter (butterworth function)

```

function [data_filt]=filt_low(fc,x,t)

nt=length(t);
dt=t(2)-t(1);% is the 1 pixel time
df=1/(dt);% converts time to freq
fm=1/dt;
fs=fm;% calculating sampling rate
% fc=3;% freq cut_off point. Any frequency about 3 is going to be
% filtered
% x=Data;
[a,b]=butter(4,fc*1e9/(fs/2));% lowpass filter
data_filt=filtfilt(a,b,x);

```

A.2.4 Sum of trace for higher order transducers

```

%% This example script shows how to use the simulation
%% to predict the f-response of a low order device
%% the outcome of the model will be lowpass filtered to remove
%% all high frequencies. Since the time window of the pulsed laser

```

```

%% is limited to 12.5 (ns), it is needed to see the data in different
areas
%% of the oscillation time. The code makes it possible to see the
possible echos
%% based on the structure of the devices.

%clear all
close all

%addpath /home/share/matlab/PLU/Model/matt_prop_v0_6/
addpath /home/mitra/matlab/whole_stack/
%addpath /home/mitra/matlab/high_order_devic/

sample.lambda_pump=415e-9;
sample.lambda_probe=780e-9;
sample.materials_list{1}='Au';
sample.materials_list{2}='fused_silica';
sample.materials_list{3}='PS';
% sample.materials_list{4}='Ag';
% sample.materials_list{4}='water';
sample.layers= [18e-9 585e-9 18e-9 1500e-9];
sample.materials=[1 3 1 2];

n=input('Number of areas: ');
fc=input('Enter a cut_off point: ');
simulation.dT=2e-12;
simulation.Tmax=n*12500e-12;
simulation.plot_on=0;

index=1;
filling =585;

for k=1:length(filling);
    sample.layers= [30e-9 (filling(k))*1e-9 30e-9 1500e-9];
    [solution,simulation_out]=matt_prop_engine_v0_6(sample,simulation);
    dR_R(k,:)=solution.dR_R-mean(solution.dR_R);
    T(k,1:length(simulation_out.T_initial))=simulation_out.T_initial;
    index=index+1;
end;
t=simulation_out.time;
zp=2^16;
F = fftshift(xtof((t(2)-t(1))*(1:zp)));
%% lowpass filter is applied to original data
x=dR_R;
[data_filt]=filt_low(fc,x,t); % lowpass fillter is applied to all data
data_array=reshape(data_filt,length(data_filt)/n,n); % rearrange the
data based on "n" numbers
t_array=reshape(t,length(t)/n,n);
freq_data=abs(fft(data_filt',zp))';

t1=t_array(:,1); data1=data_array(:,1);

```

```

t2=t_array(:,2); data2=data_array(:,2);

t3=t_array(:,3); data3=data_array(:,3);

sum_data=([data1 + data2 + data3 ])./n;
fft_sum_data=abs(fft(sum_data,zp));

%save('acoustic_trace_model.mat','t','sum_data','F','data_filt','data1'
,'t1');
%% plotting data
m=8;
subplot(3,2,1);plot(t,data_filt);title('\delta R/R of
au:ps:au(18:585:18)','fontsize',m);xlabel('Time (s)')
subplot(3,2,2);plot(t1,data1);title('\delta R/R the area
1','fontsize',m);xlabel('Time (s)');%xlim([0 1.2])
subplot(3,2,3);plot(t1,data2);title('\delta R/R the area
2','fontsize',m);xlabel('Time (s)');%xlim([0 1.2])
subplot(3,2,4);plot(t1,data3);title('\delta R/R the area
3','fontsize',m);xlabel('Time (s)');%xlim([0 1.2])
subplot(3,2,5);plot(t1,sum_data);title('\delta R/R Sum of the
traces','fontsize',m);xlabel('Time (ns)');%xlim([0 1.2])
subplot(3,2,6);plot(F*1e-9,fft_sum_data);title('FFT
\delta R/R');xlabel('freq GHz','fontsize',m);axis([0 10 0
max(max(fft_sum_data))]));
%
print('-dpng','Modeled data');

```

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