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Real-Time Ellipsometry-Based Transmission Ultrasound Imaging

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LDRD Auspices Statement

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Introduction

Ultrasonic imaging is a valuable tool for non-destructive evaluation and medical diagnosis. Reflection mode is exclusively used for medical imaging, and is most frequently used for non-destructive evaluation (NDE) because of the relative speed of acquisition. Reflection mode imaging is qualitative, yielding little information about material properties, and usually only about material interfaces. Transmission imaging can be used in 3D reconstructions to yield quantitative information: sound speed and attenuation. Unfortunately, traditional scanning methods of acquiring transmission data are very slow, requiring on the order of 20 minutes per image.

The sensing of acoustic pressure fields as optical images can significantly speed data acquisition. An entire 2D acoustic pressure field can be acquired in under a second. The speed of data acquisition for a 2D view makes it feasible to obtain multiple views of an object. With multiple views, 3D reconstruction becomes possible. A fast, compact (no big magnets or accelerators), inexpensive, 3D imaging technology that uses no ionizing radiation could be a boon to the NDE and medical communities. 2D transmission images could be examined in real time to give the ultrasonic equivalent of a fluoroscope, or accumulated in such a way as to acquire phase and amplitude data over multiple views for 3D reconstruction (for breast cancer imaging, for example). Composite panels produced for the aircraft and automobile industries could be inspected in near real time, and inspection of attenuating materials such as ceramics and high explosives would be possible.

There are currently three optical-readout imaging transmission ultrasound technologies available. One is based on frustrated total internal reflection (FTIR) [1,2], one on Fabry-Perot interferometry [3], and another on critical angle modulation [4]. Each of these techniques has its problems. The FTIR based system cannot currently be scaled to large aperture sizes, the Fabry-Perot system has never been fully implemented for area imaging, and the critical angle modulation system is not sensitive enough for medical imaging.

We proposed an entirely new way of using acoustic pressure to modulate a light beam. This new technology should be sensitive enough to be useful for medical imaging and have a large enough aperture to speed acquisition by orders of magnitude over point sampling. Unfortunately, we were unable to bring this technology to fruition.

Background

The sensor is based on two technologies, ellipsometry and aerogel thin film fabrication. Ellipsometry is based on measuring changes in the polarization state of light, encompasses a large number of techniques used to characterize optical thin films, and is capable of detecting thickness variations in a thin film of less than an Angstrom[5]. Comparison ellipsometry would be used to determine the changes in thickness induced in an aerogel thin film as it is deformed by an acoustic pressure wave.

Aerogel Thin Films

Aerogels are porous materials made by creating a gel and replacing the liquid in the gel structure with air (drying the gel)[6]. Aerogels used to have to be made by supercritically drying the gel (to prevent the gel structure from collapsing under surface tension), but in the last decade ambient-dried aerogels have become available [7]. These aerogels can be deposited on an optical substrate by either spin or dip coating. The film characteristics can be manipulated through both the deposition process and the gel chemistry.

Ellipsometry

Ellipsometry is a technique normally used to determine the thickness, refractive index, and absorption of a thin film. The thin film is usually deposited on a substrate and the substrate and film are surrounded by an ambient (such as air). In a null ellipsometer (see figure 1) a monochromatic input beam is polarized, passed through a phase compensator, reflected off of the thin film/substrate system, and then passed through a final polarizer (or analyzer). By adjusting the angles of the polarizer, compensator, and analyzer the light reflected off of the thin film/substrate system is nulled to zero. By measuring the polarization component angles, and knowing the angle of incidence on the film, the optical wavelength, and the substrate parameters, we can deduce the parameters of the thin film.

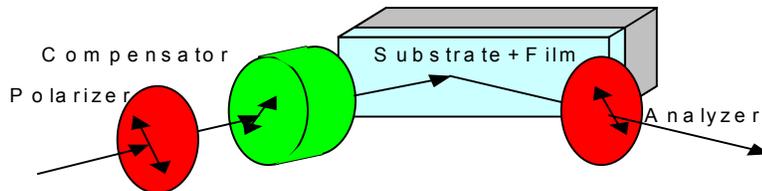


Figure 1. Null Ellipsometer.

Null ellipsometry could be used as an ultrasonic imaging technology, but it has some drawbacks. Compensators are wavelength dependent devices, rarely have a large aperture, and are expensive. These problems restrict the field of view of an imaging ellipsometer to approximately 2 square inches, and restrict the light source to a laser or light that has been passed through a monochromator. A promising alternative is comparison ellipsometry.

Comparison ellipsometry [8] depends on the fact that light polarized parallel to the plane of incidence of the reflection off an isotropic test surface will remain polarized parallel to the plane of incidence after reflection. Likewise, light polarized perpendicular to the plane of incidence will remain polarized perpendicular to the plane of incidence after reflection. Only their phases and amplitudes will change. Suppose we now take two surfaces, a reference and a test surface (see Figure 2), and rotate them relative to one another such that the light polarized parallel to the plane of incidence in the reference is polarized perpendicular to the plane of incidence in the test and the light polarized perpendicular to the plane of incidence in the reference is polarized parallel to the plane of incidence in the test. Then linearly polarized white light reflected from the reference surface will become elliptically polarized, and, if the two surfaces are identical, that elliptically polarized white light, when reflected off the test surface, will become linearly polarized. If the two surfaces differ, the white light reflected from the test surface will be elliptically polarized at some wavelengths, and some of that light will leak through the analyzer.

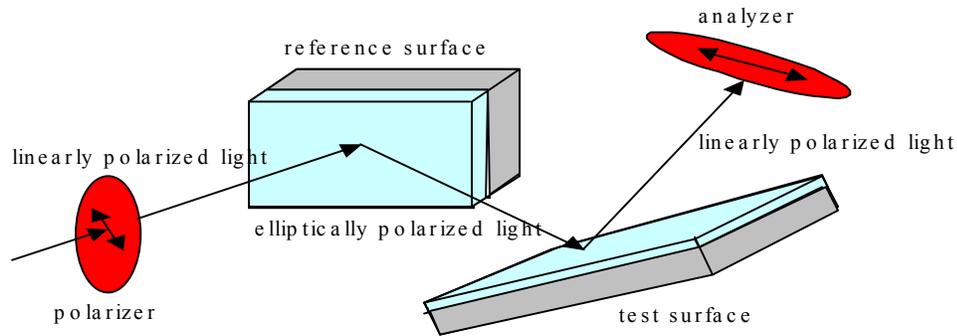


Figure 2. Comparison ellipsometer. Linearly polarized light bounces off the reference surface and becomes elliptically polarized light. If the test surface is identical to the reference surface, the elliptically polarized light bounces off and becomes linearly polarized again.

So far the comparison ellipsometer is not an ultrasound detector, but the ambient does not have to be air, and the reference surface does not have to be solid. We planned to use glass prisms as our ambient, aerogel thin films as our film, and water as our reference surface. Without ultrasound, the surfaces are identical and the light is nulled out. When an ultrasound pressure wave distorts the aerogel film in the test surface, light leaks through the analyzer. This situation is illustrated in Figure 3.

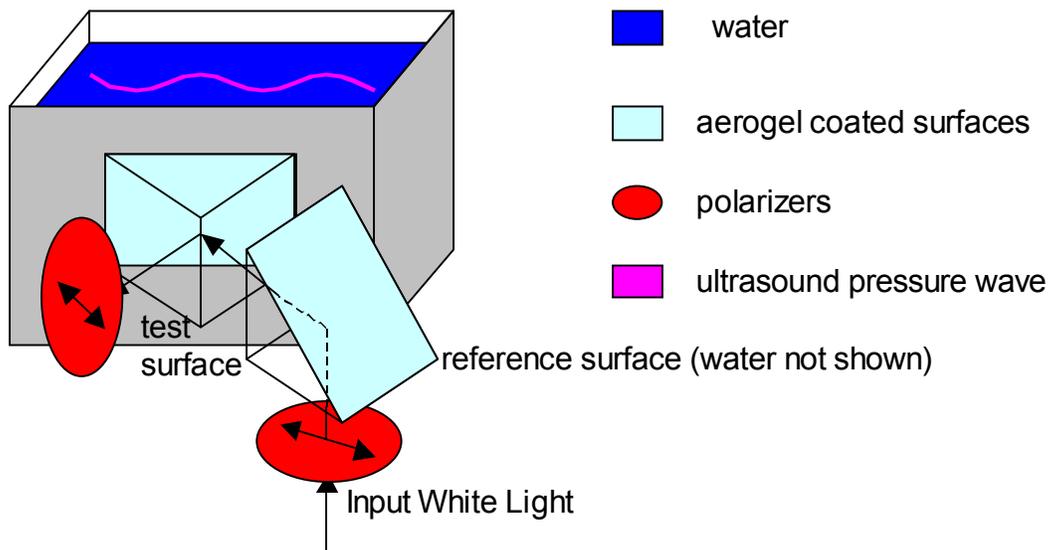


Figure 3. Ultrasound sensor based on comparison ellipsometry. If there is no ultrasound the light is nulled out, but when ultrasound hits the aerogel thin film the test surface deviates from the reference surface, and light leaks through the analyzer.

This comparison ellipsometer makes possible real time imaging of ultrasound intensity (and so is the ultrasonic equivalent of an x-ray fluoroscope). By changing the angle of either the polarizer or analyzer slightly it is possible to use this device away from the null and obtain phase and amplitude data useful for 3D reconstruction.

Such a sensor needs to be supported with acoustic and optical systems. Standard ultrasound transducers can be used to produce the acoustic pressure field, a collimated white light source

can be provided for illumination, and a camera can pick up the sensor response.

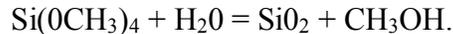
There are a variety of acoustic systems that the sensor can be used with: acoustic holography, imaging with an acoustic lens, and transmission imaging. Acoustic holography can be used to obtain pressure phase and amplitude information needed for 3D reconstruction from a nulled sensor (which normally would not provide these data). The acoustic lens could be used to image a particular plane of interest (ultrasonic tomogram). The direct imaging of the scattered pressure wave can be used to acquire pressure phase and amplitude.

Research Activities

Aerogel Chemistry Research

The properties of an aerogel depend heavily on its chemistry. We have made a number of iterations on the chemistry of our aerogel precursors. The final chemistry and processing procedure are laid out in Figure 4.

Our precursor chemical is Tetramethoxy Silane (TMOS). TMOS is mixed with water and catalyst and the proper amount of diluent to create a sol-gel network. The basic reaction is:



The SiO_2 network formed is a combination of solid particles and open pores. During the drying phase the Methanol (CH_3OH) evaporates condensing the structure and increasing its density (and refractive index). In order to enhance the materials hydrophobicity we add (3,3,3 Trifluoropropyl) tri-methoxysilane. This chemical leaves F1 atoms "exposed" in the structure. The highly electronegative F1 atoms enhance the materials hydrophobicity preventing water's entrance into the porous structure. We use a 35% molar ratio mix of (3,3,3 Trifluoropropyl) tri-methoxysilane to Tetramethoxy Silane (TMOS). An initial mix of Tetramethoxy Silane (TMOS) + (3,3,3 Trifluoropropyl) tri-methoxysilane is hydrolyzed with a stoichiometric addition of H_2O and 20 uL of 37% HCl plus 50 gr. of Methanol as a pre-diluent. This solution is stable and can be used after many months. The spin coating is done with the initial mix diluted with another 50 gr. of Methanol and an appropriate amount of base to yield a gelation time of roughly 1 hour. This sol-gel mixture has a theoretical density of 100mg/cc.

Before the mix gels, we drop the sol-gel solution onto our substrate. We are using a Laurell Technologies Corp. spinner Model # WS-400. Our experiments yielded the optimum results at spin speeds of 3000-6000 rpm's. The spin coating was performed in an atmosphere of alcohol/ NH_4OH . After spin coating, the film/substrate is removed and placed in a small chamber of alcohol/ NH_4OH to enhance gel polymerization. After 20 minutes the film/substrate is transferred to a small container of methanol. The film/substrate is then "treated" with the addition Chloro-trimethyl silane to the methanol. This treatment reduces the aerogel collapse on subsequent drying. The solvent is exchanged with hexane after the chlorosilane treatment and then air dried.

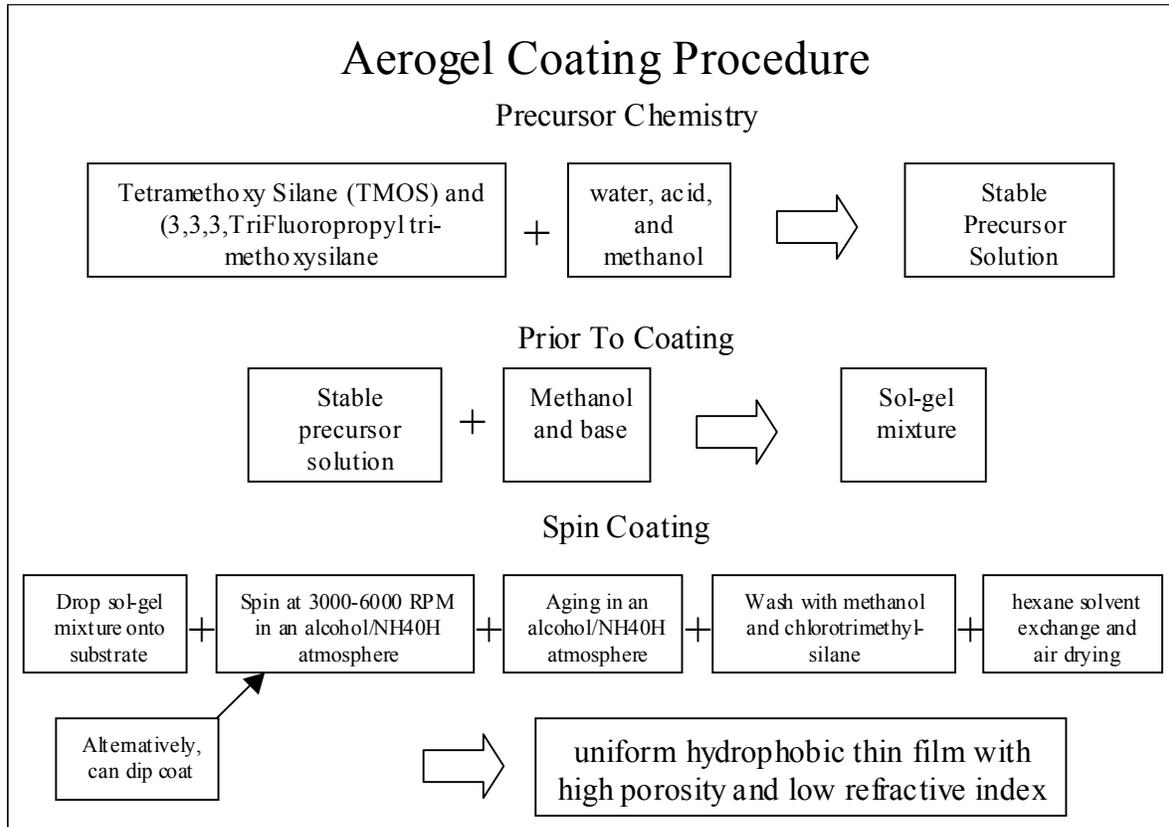


Figure 4. Aerogel chemistry and coating procedure.

Aerogel Metrology Work

In order to determine the utility of our aerogel thin films we have to be able to measure their properties. We need to examine three aspects of any particular film: its index of refraction, its thickness, and its uniformity. We used a Brewster angle measurement system to determine film indices of refraction and a reflectometer to determine film thickness

Index of Refraction

If light specularly reflected from a surface is polarized in the plane of incidence, there is an angle at which there is no reflection. This is the Brewster angle (see Figure 5). We can utilize this fact to determine the index of refraction of a thin film of nonuniform thickness.

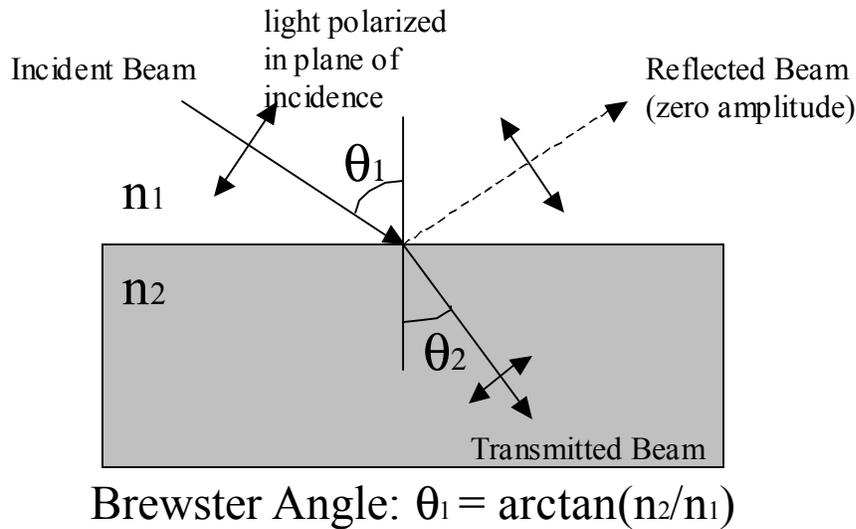


Figure 5. The Brewster angle is the angle of incidence such that there is no specular reflection if the light is polarized in the plane of incidence.

In our early coating research we were not overly concerned with making uniform thin films, so there was a considerable amount of thickness variation (up to 5 micrometers) over the samples we created. These thickness variations were observable in the form of interference fringes when the films were examined by specular reflection of narrow bandwidth light. We built a test stand to examine these fringes under polarized illumination from a red LED so we could determine the Brewster angle for our films and thus their indices of refraction. This test stand is illustrated in Figure 6.

Brewster Angle Test Stand

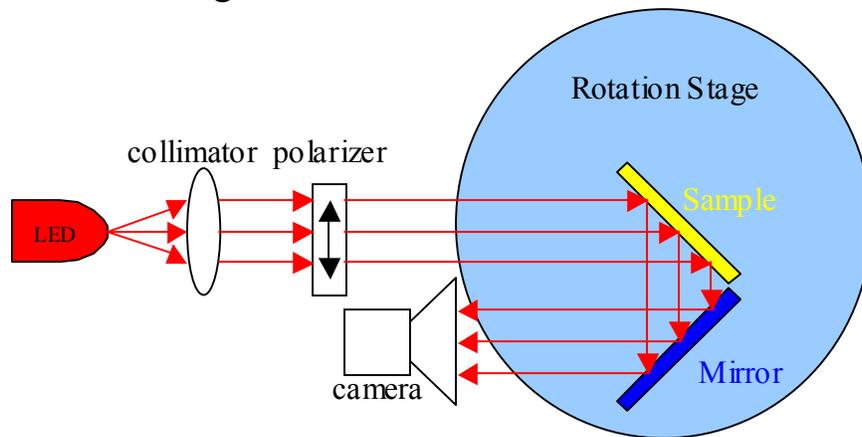


Figure 6. Variations in film thickness on the sample causes fringes in the camera image. Light polarized parallel to the plane of incidence has no reflection from the film at the film's Brewster angle. The fringes disappear from the image in the camera at the film's Brewster angle. As the index of refraction of air is approximately 1, the index of refraction of the film is equal to the tangent of the film's Brewster angle.

Using the Brewster angle as an index of refraction diagnostic we were able to set our film chemistry such that we obtained films with refractive indices on the order of 1.06. The film indices have varied from as low as 1.05 to as high as 1.45.

Thickness

To measure film thickness we used a reflectometer. A reflectometer sends a beam of light toward the sample and determines the spectrum of the reflected light (see Figure 7).

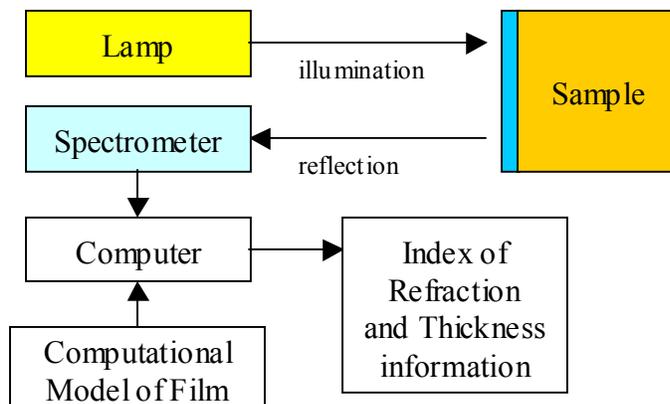


Figure 7. Reflectometry is used to determine the thickness and index of refraction of a thin film on a substrate. The sample is illuminated by a fiber and the light reflected from the sample is collected and passed through a spectrometer. The data is combined with a computational model of the film to extract the film thickness and index of refraction.

Given a spectrum from an uncoated sample and a coated sample, the spectrometer can determine the thickness of the coating. We used a Mikropak NanoCalc 2000 reflectometer. Unfortunately, the software that drives the NanoCalc needs to know the index of refraction of the film before it can determine the thickness. As we were constantly exploring the chemistry of the aerogel precursors at the same time we were trying to measure the thickness and index, we did not have index information to use in the software. We used the reflectometer to obtain raw data and fit the spectrum curves using software we wrote for that purpose.

Our software can model the spectral response of a thin film with wavelength dependent index of refraction and absorption using the matrix formulation for isotropic layered media [9]. In addition it can model the alteration of the response that occurs when a sample has significant film thickness variations. The software is operated in two modes sequentially: the user determines the rough parameters of the film by using the mouse to match the experimental spectrum to a computed spectrum, then the computer is used to refine the parameter estimates (the refinement software uses a downhill simplex method [10] in the following 8 dimensions: base film thickness, film thickness variation, three wavelength dependent refractive index terms, and three wavelength dependent absorption terms). This work flow is illustrated in Figure 8. An actual image from the software user interface is shown in Figure 9. A second image from the software user interface, shown in Figure 10, shows why both the reflectometer and the Brewster angle measurement are needed. For any experimental spectrum obtained from the reflectometer

there are at least two positions in film thickness-index space that could have generated it. The two positions that match the input spectrum shown in Figures 9 and 10 are shown in Table 1.

Figure	Thickness (nanometers)	Index
9	520	1.06
10	405	1.37

Table 1. Film thicknesses and indices of refraction that each match experimental data shown in Figures 9 and 10.

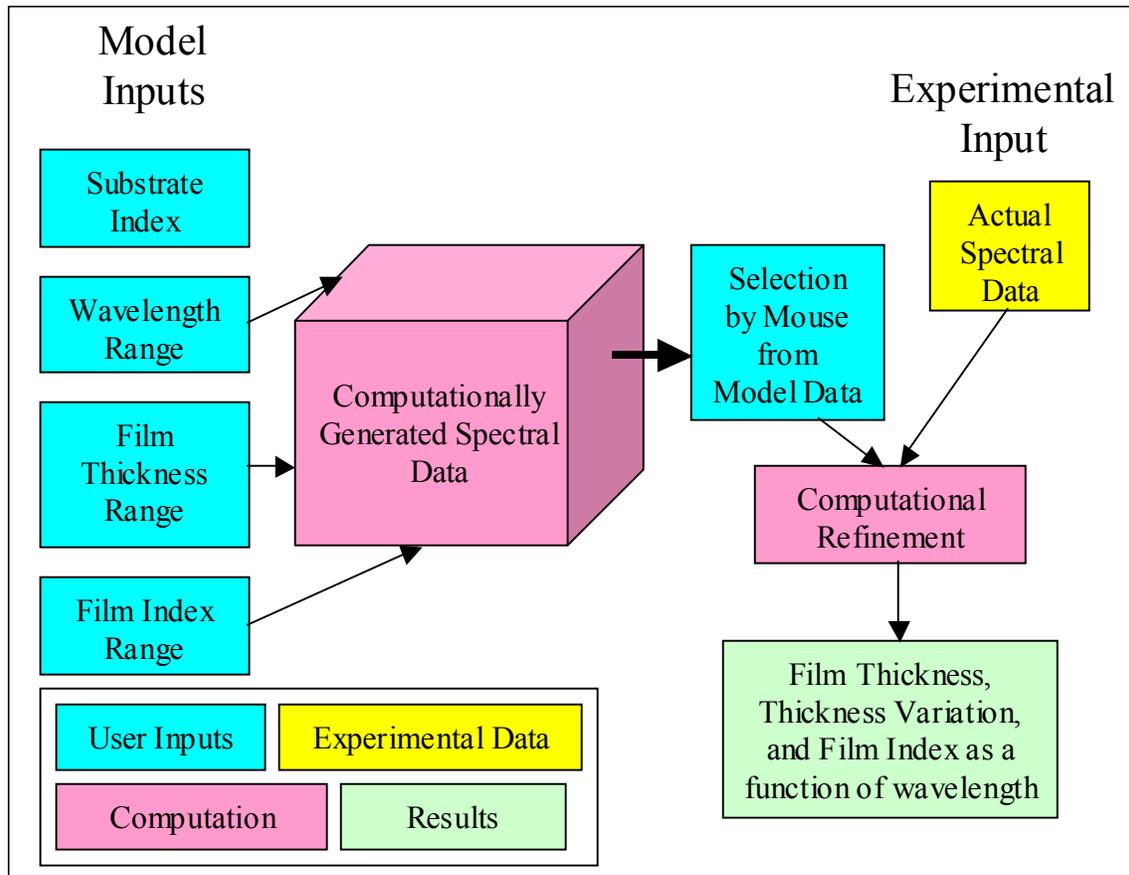


Figure 8. Work flow for determining film thickness, thickness variation, and index as a function of wavelength. The computer generates a cube of spectral data (three axes: film thickness, film index, and wavelength) from which the user selects a spectrum close to the actual spectral data by mouse. This initial guess is refined computationally to determine the actual film parameters.

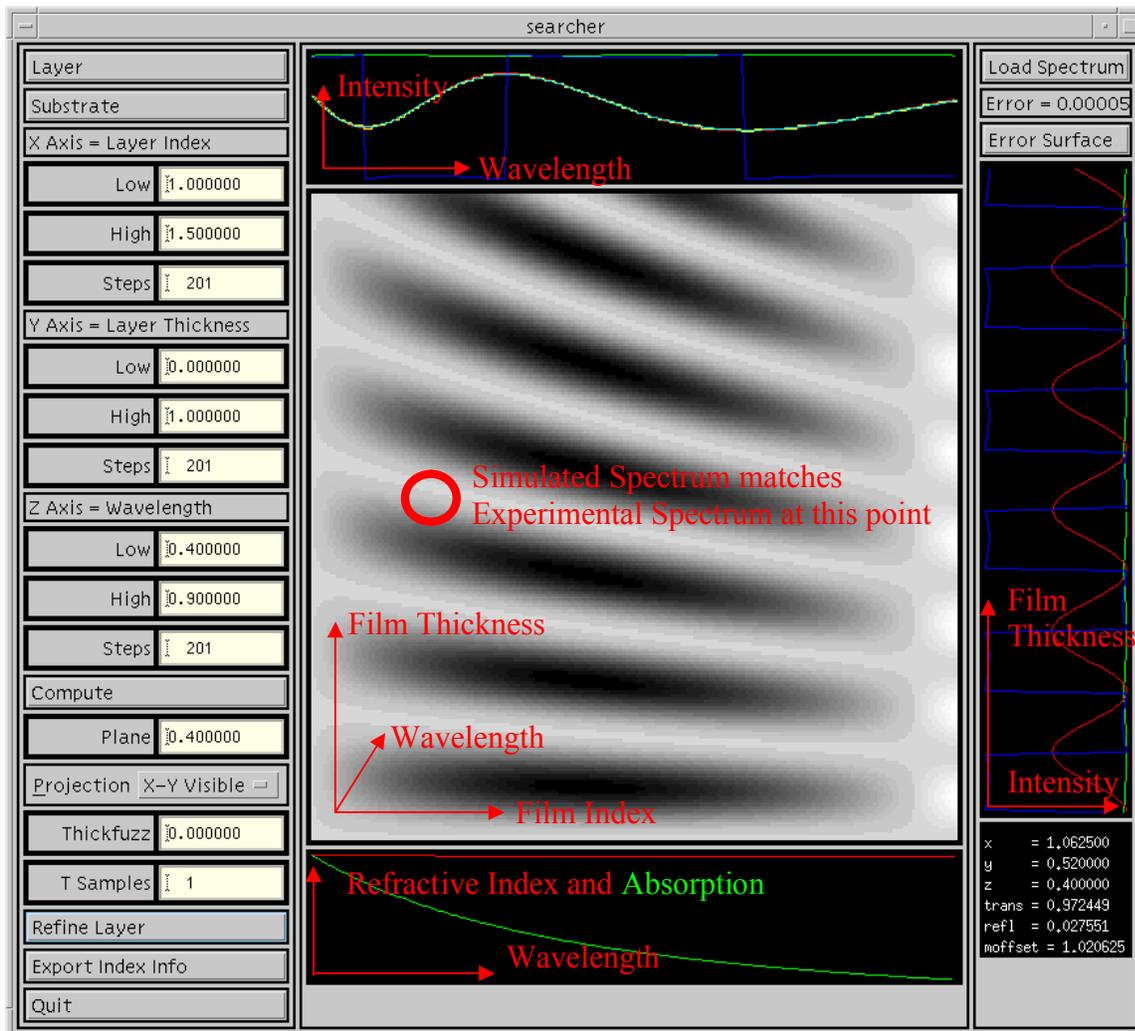


Figure 9. User interface of software described in Figure 8. The operator uses the mouse in the central image to pick one of the 40,000 computed spectra to try and match the experimental spectral data. The graph in the top portion of the window shows the currently selected computed spectrum in red, the experimental spectrum in yellow and the refined computed spectrum in cyan. All three curves overlap. For the spectrum shown, the film is 520 nm thick and has an index of 1.06 (position circled in red).

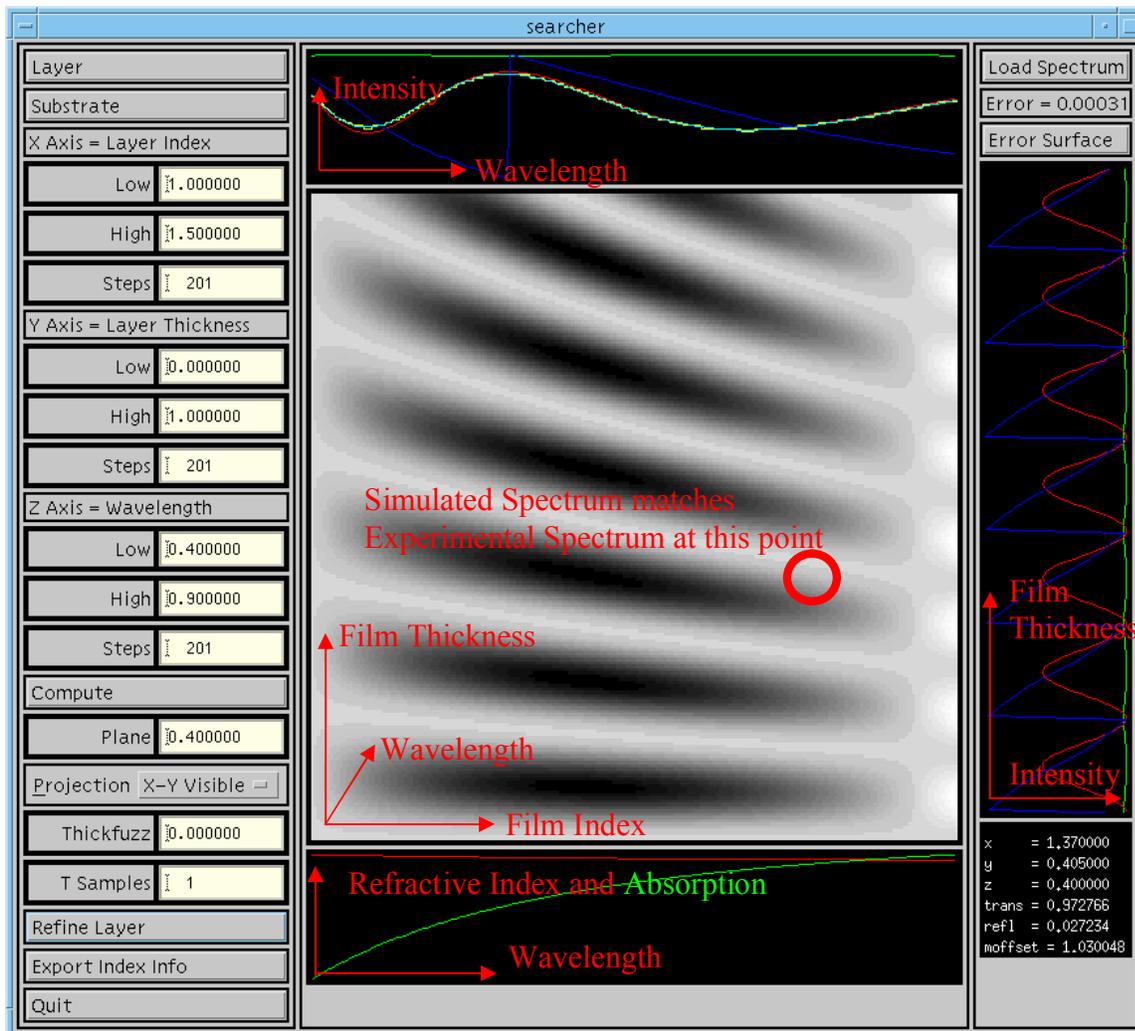


Figure 10. This figure shows the reason both reflectometry and Brewster angle measurement are needed. For the same experimental spectrum shown in Figure 9, an alternative location in thickness-index space can be found. A film 405 nm thick with an index of 1.37 (circled in red) yields the same spectrum as one 520 nm thick with an index of 1.06.

Proof of Principle Test Stand

A proof of principle test stand was built to test the sensing technique. This test stand was to test the pressure transduction at a single point as well as being a platform for small scale imaging. It consists of an optical table on which rests a small water tank and acoustic source. The water tank has a port in one side that will accept a small prototype sensor. This test stand was to be used to examine the sensitivity, dynamic range, frequency response, and noise characteristics of the sensor. Figure 11 shows a picture of the optical train of the test stand.

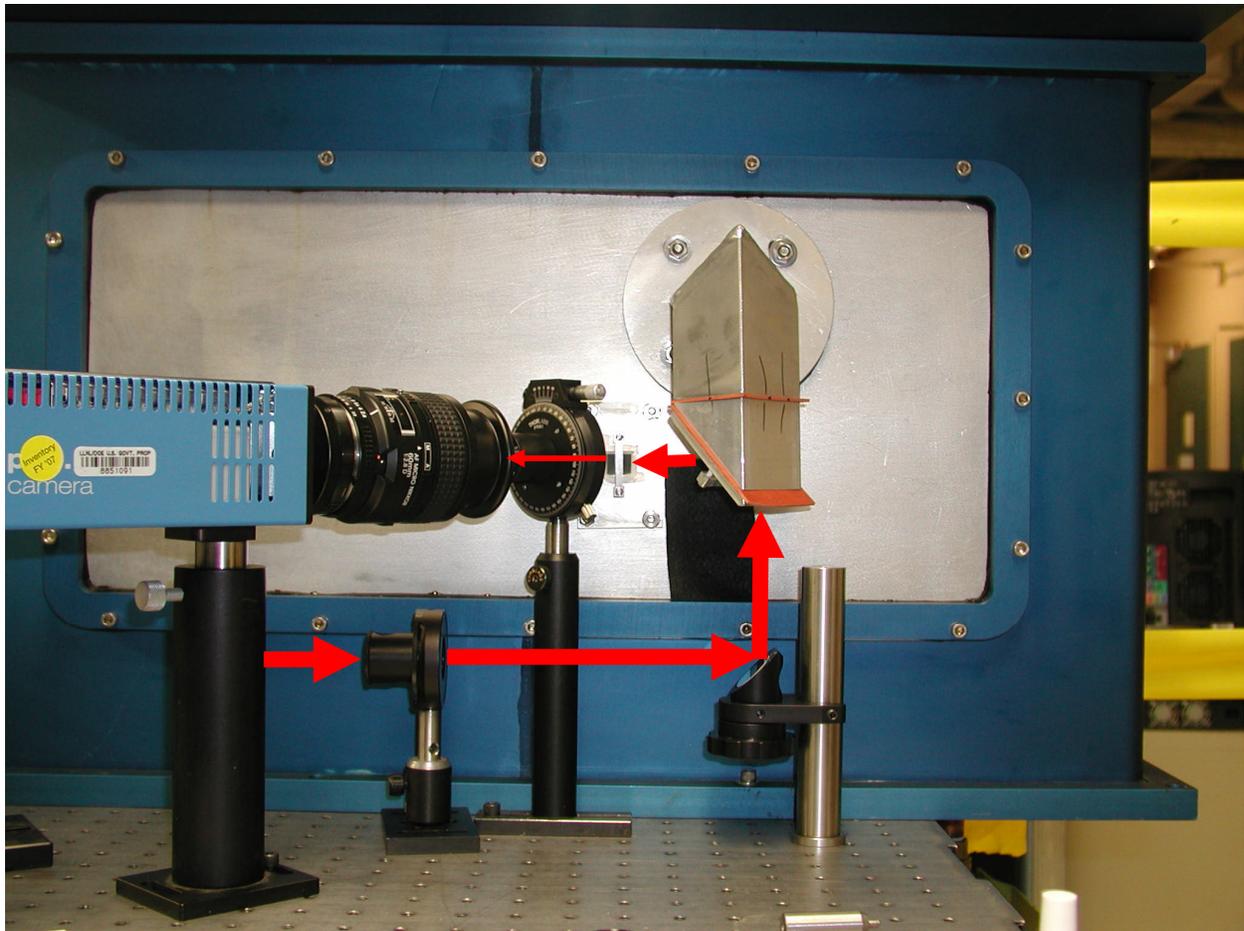
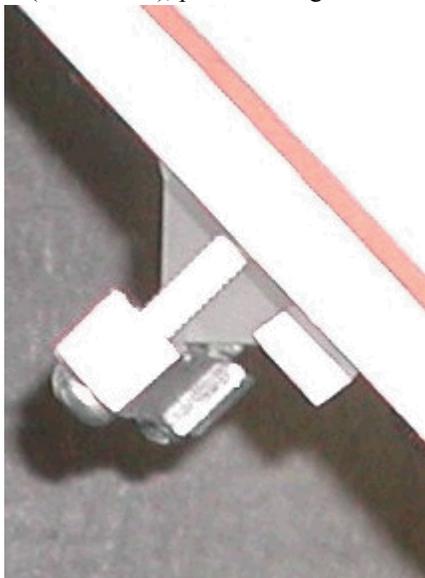
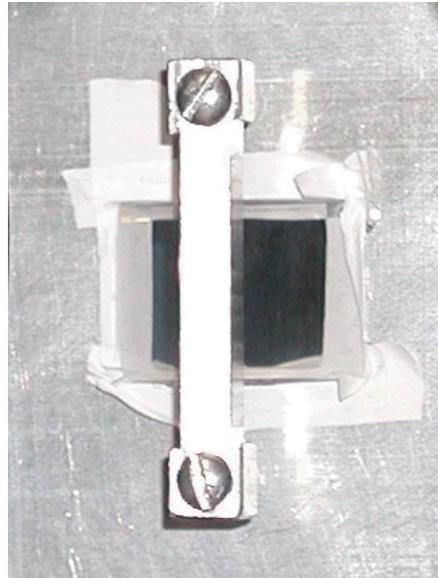


Figure 11. The optical train of the proof of principle test stand. Collimated light (from a modified slide projector) enters from the left and is polarized, reflected off of a 45 degree mirror, hits the first prism (reference surface), hits the second prism (test surface), passes through the analyzer and then enters the camera.



Reference Prism



Test Prism

Figure 12. Close-up photographs of the reference and test prisms on the test stand.

Results/Technical Outcome

Early in FY06 we obtained our reflectometer and began spin coating silica discs in order to determine the correct chemical makeup for an aerogel of the correct index and thickness. Our early films were very bad. No two films had the same index or thickness, and the films had large thickness variations on each silica disc. These thickness variations are what allowed us to use the Brewster angle apparatus to unambiguously determine the index of refraction of our samples. In December we determined that we had films with indices of refraction on the order of 1.07. In January we experimented with a formulation that had an index of refraction of 1.11.

In addition to sol-gel films we also experimented with spin coating samples of block-copolymer materials. We determined that they had indices of about 1.43 and dropped them from consideration.

Late in January we moved from spin coating to dip coating using the sol-gel formulations. We determined that the index of refraction of these samples was approximately 1.43, i.e. we were seeing films that collapsed rather than maintaining their porous structure. We went back to spin coating.

By March we were spin coating thin films with thickness variations on the order of 10 nanometers. These films were flat enough that we could no longer observe fringes and so the Brewster angle method we were using as an absolute indication of refractive index could no longer be used.

Over the course of the following months we were generating films with indices on the order of 1.08 and thicknesses on the order of 0.4 microns. These parameters were close enough to those required for using frustrated total internal reflection (FTIR) as in [1] that we determined to push into the FTIR parameter space (index < 1.08, thickness < 0.3 microns). Eventually we generated films that the reflectometer claimed were inside the FTIR parameter space.

Our schedule fell apart at this point with the departure of the mechanical engineering part of our team. This delayed the completion of the test stand until September. Test stand experiments began September 18th.

On the 18th, 19th, and 20th, of September we installed the prisms we had coated into the test tank and saw very little light, even using a high dynamic range camera and a wide open aperture on the camera lens. If the prism coatings had had as low an index as we believed, we should have seen a great deal of light due to incompletely frustrated total internal reflection. The lack of light indicated that we had dense coatings. On the 20th the bulb in our light source blew. The evening of the 20th we purchased a new bulb for the projector. On the 21st the new bulb was installed and we tried insonifying the test sensor surface with 1 MHz ultrasound but were unable to detect any effect whatsoever. We fabricated a new set of coated prisms and on the 25th of September tried to use them. The coatings were thick, high index, and were not useful.

Exit Plan

Clearly, creating aerogel thin films for comparison ellipsometry is a difficult problem. So much so that it would be ill advised to continue down that path for ultrasound sensing. However, we have built a fully functional imaging test stand and it would be a shame for that effort to go to waste. We are currently investigating alternative techniques for both generating low index uniform thin films as well as producing scalable narrow air gaps for frustrated total internal reflection imaging. Should any of these techniques prove feasible we will have a test stand upon which to evaluate them. The test stand has currently been mothballed for future use.

Summary

Transmission ultrasound is an imaging technique with great potential for medical and non-destructive evaluation uses. We attempted to speed transmission ultrasound data acquisition by a factor of 1000 by using comparison ellipsometry of aerogel thin films to image the pressure distribution over an entire surface (in contrast to acquiring pressure data by mechanically scanning a point sensor). We spent the majority of our time learning how to produce uniform aerogel thin films. These films were discovered to be unusable when they were installed in the imaging test stand (the construction of which was delayed by the loss of team personnel). The test stand is currently mothballed, awaiting new technologies for thin film fabrication.

Acknowledgements

We wish to acknowledge the assistance of Trung Le, the mechanical technician who constructed the test stand tank and sensor assembly. Without his efforts we would not have been able to obtain any results whatsoever.

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