Ultrafast opto-acoustics applied to the study of material nanostructures

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Abstract. The propagation of ultra-short sound pulses in water has been studied using an ultrafast opto-acoustic technique. A pulse time-of-flight technique for measuring the depths of deep channels in Si-based nanostructures was demonstrated. We report in these proof-of-concept ultrasonic experiments how spatial profile information of nanostructures can be acquired, where sound pulses propagate down narrow channels in patterned nanostructures. We have been able to detect acoustic echoes for sound propagating along a channel as narrow as 35 nm with depth to width ratios exceeding 10:1.

1. Introduction

Ultrafast opto-acoustic methods [1] provide a way to generate short (~1 ps) acoustic pulses for fundamental studies and metrology of small structures. An ultrashort (~100 fs) pump light pulse is absorbed at the surface of a sample, and the optical reflectivity of the sample is then measured with a probe pulse whose arrival is time-delayed with respect to the pump pulse by a variable optical path. The pump pulse excites the sample and generates an elastic stress; as this stress relaxes it launches an acoustic pulse into the sample. For example, the technique of picosecond ultrasonics has been used to study the vibrational modes of nanostructures directly [2]. Picosecond ultrasonics has also been used to generate sound pulses and scatter them off of a nanostructure on the surface of a silicon wafer [3]. In this paper we demonstrate an acoustic microscope in which sound pulses are generated optically, transmitted through a coupling fluid (water), and detected after reflection from silicon-based test samples which feature periodic nanoscale deep trenches.

In a pulsed reflection acoustic microscope, sound waves are generated by an electromechanical transducer and brought to a focus at the surface of the sample which is immersed in a fluid coupling medium. Since the demonstration of a scanning acoustic microscope by Lemons and Quate [4], many researchers have explored the possibilities of acoustic microscopy. Using water as the coupling medium between the transducer and the sample, a resolution on the order of an optical wavelength has been demonstrated [5]. One of the appeals of using sound for imaging is the possibility of detecting subsurface defects in materials that are opaque optically, but relatively transparent (i.e., non-attenuating) acoustically. In addition, an acoustic microscope can image systems where the optical contrast is small but the acoustic contrast, or the difference in mechanical properties, is high [6].

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best resolution for an instrument using water as a coupling medium was reported by Hadimioglu and Quate [7] as 240 nm. In that work they used 3 ns rf pulses with a frequency of 4.4 GHz to excite a piezoelectric transducer. It is difficult to generate pulses much shorter than this electronically. In this work we examine the use of the picosecond ultrasonics technique to generate ultrashort (~1 ps) acoustic pulses and apply them to acoustic microscopy. In section 2 we address the problem of designing and testing an opto-acoustic transceiver, and in section 3 we present results from a first step implementation of this kind of acoustic microscope where the sound is not focused and a periodic array of nanostructures are probed.

2. Ultrafast Opto-Acoustic Transceiver Design and Testing

The picosecond ultrasonics technique provides a convenient means of generating ultrashort acoustic pulses, but detecting these pulses after they have propagated through a layer of coupling fluid, in this case water, presents some difficulties. There are several factors that reduce the amplitude of the reflected pulse. At an interface between aluminum and water, for example, the reflection coefficient for the strain amplitude is 0.84. Since the sound has to cross interfaces both into and out of the water, the returning signal will be significantly reduced. In addition, as the sound pulse propagates through the coupling fluid, the viscosity of the water makes the width of the pulse increase and the amplitude decrease. For example, when an acoustic pulse whose width is initially described by a delta function propagates through 1 micron of water, its profile will stretch out into a Gaussian shape with a FWHM of 117 nm.

In order to detect the returning acoustic pulses, we use an opto-acoustic transceiver structure that consists of films of aluminum, silicon dioxide, and aluminum with thicknesses of 7 nm, 230 nm, and 100 nm respectively as shown in figure 1. These layers were deposited onto a silicon dioxide substrate using a sputtering system with base pressure $1 \times 10^{-6}$ mbar. The transceiver structure acts as a Fabry-Perot optical resonator, and the optical reflectivity as a function of wavelength of one such structure is shown in figure 3. The pump and probe beams enter the cavity through the 7 nm Al film. The Al film on the far side of the cavity is thick enough to prevent any light reaching the water and sample. The effective quality factor Q of the transceiver is modest, by design, for optimizing its overall performance, since the reflectivity of the thinner Al film is approximately 0.5.

The thickness of the silicon dioxide layer was chosen so as to make the transceiver structure most sensitive to changes in the optical thickness of this layer. The calculated variation in the optical reflectivity of the transceiver structure with respect to the thickness of the silicon dioxide film for a wavelength of 800 nm is shown in figure 4. The returning sound will also perturb the optical

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**Figure 1.** Schematic diagram of the optoacoustic transceiver. The Al/SiO$_2$/Al film stack acts as a Fabry-Perot optical cavity. The thickness of the middle SiO$_2$ film is chosen such that the optical reflectivity is sensitive to changes caused by the returning sound pulses.

**Figure 2.** Left: Schematic of a scanning opto-acoustic microscope. Acoustic pulses generated by the transceiver structure are focused onto individual nanostructures by an acoustic lens. Right: Schematic of planar opto-acoustic microscope.
properties of the aluminum films; the size of this additional perturbation is determined by the piezo-optic constants of Al. These coefficients have been measured by Jiles and Staines [8] but are found to vary rapidly for wavelengths around 800 nm. Their precise values are also likely to depend on the deposition conditions for the aluminum films, and consequently we will not comment on the size of this cavity-enhancement effect in detection between experiment and a model here.

To summarize the time sequence in the ultrafast optoacoustic experiments: the sound that is generated when light is absorbed in the Al films propagates across a thin water layer, scatters off of the surface of the sample, and then returns to be detected in the transceiver. Experimental results with
a water thickness of 830 nm and using a flat surface of a silicon wafer as a sample are shown in figure 5. The slowly changing background is caused by the initial heating of the films in the transceiver structure; the temperature change perturbs the optical properties of the transceiver. The fast oscillations in the data [9], known as Brillouin oscillations, are caused by alternating constructive and destructive interference between the probe light that has reflected from the cavity and the probe light that has scattered from a sound pulse that propagates into the silicon dioxide substrate. These oscillations have no effect on the operation of the transceiver and they can be easily subtracted out numerically using a Fourier filter. In figure 6, we show the first echo from the silicon surface after subtracting out the slowly decaying background and the fast oscillations. By varying the thickness of the water and analyzing the Fourier components of the returning echoes, the velocity and attenuation of sound in water for frequencies up to 11 GHz can be measured [10]. The dip that occurs at around 1300 ps is attributed to sound which has been reflected at the transceiver-water interface with a sign change, reflected at the transceiver-substrate interface, transmitted into the coupling fluid, reflected at the sample and then returned to the transceiver.

3. Planar Opto-acoustic Microscopy

We are currently working on implementing an acoustic lens to focus the sound generated by the transceiver structure to develop a full scanning instrument. However, there is also considerable value in using an “unfocused” version of this technique in which planar sound pulses are scattered off of periodic nanostructures (see figure 2). While this technique cannot resolve individual nanostructures, experiments of this kind can provide information as to the average profile of the structures being examined.

As a first experimental test of these ideas, we have measured sound propagation within a number of trench structures and here report on two samples shown in figures 7 and 8. The samples consist of silicon dioxide lines on a silicon wafer with a 5 nm thick silicon nitride layer deposited on the surface of the lines and on the substrate. The depth of the trenches is measured by SEM to be 405 nm. The lines cover an area on the silicon wafer of about 500 by 100 µm, much larger than the area of the pump and probe light spots, which have a radius of approximately 10 µm. Sample A has a repeat distance of 240 nm and an average trench width halfway down of 43±4 nm. Sample B has a repeat distance of 200 nm and an average trench width halfway down of 38±16 nm. Sample A has a repeat distance of 240 nm and an average trench width halfway down of 43±4 nm. Sample B has a repeat distance of 200 nm and an average trench width halfway down of 38±16 nm.

Figure 7. Cross section SEM image of sample A. The average width of the channels halfway down and at the bottom is 43 and 28 nm, respectively. The period is 250 nm, and the depth is 405 nm.

Figure 8. Cross section SEM image of sample B. The average width of the channels halfway down and at the bottom is 38 and 16 nm, respectively. The period is 200 nm, and the depth is 405 nm.
distance of 200 nm and an average trench width half way down of 38±6 nm. As can be seen from figures 7 and 8, there are appreciable fluctuations in the width of the trenches, particularly towards the bottom of the channels in sample B.

The experiment was performed using the opto-acoustic transceiver described in the previous section. The data set for sample A taken at a temperature of 23 °C is the black curve in figure 9. The first echo at 1147 ps is from sound reflected at the top of the structure, and the second smaller echo at 1730 ps from sound that has propagated into the trench and reflected at the bottom. The difference in the arrival times for the echoes in figure 9 is 583±6 ps and corresponds to the time required to traverse the channel twice. We have simulated the reflection of a sharp acoustic pulse from the structure shown in figure 10, using periodic boundary conditions in the lateral direction and taking into account both the shear and bulk viscosity of the water. The red curve in figure 9 shows the simulated results for the average pressure on the face of the transceiver versus time. The transceiver was taken to be 855 nm above the top surface of the sample. The difference between the arrival times for the echoes found in the simulation is 594 ps, close to the measured time of 583 ps. These values are significantly greater than the time of 541 ps required for sound to propagate through 810 nm (i.e., twice the depth of the channels) of bulk water; the velocity of the sound pulse down the narrow channel is decreased because of the viscous interaction with the wall.

![Figure 9. Experimental (black) and simulated (red) change in reflectivity for sample A, normalized so that the height of the first peak is unity.](image1)

![Figure 10. Profile used for the simulation for sample A. Dimensions taken from SEM profile measurements shown in figure 7. Units are nm.](image2)

The experimental data for sample B are shown by the black curve in figure 11. Narrowing the channels significantly reduces the magnitude of the echo from the bottom. The difference in the arrival times of the top and bottom echoes is now 563±12 ps, which again is slower than in a bulk fluid, but slightly larger than the velocity in the trench of sample A. This is surprising and is presumed to be a consequence of the different profiles of the trenches in the two samples (see figures 7 and 8). The simulation of the reflection of a sharp acoustic pulse from the structure shown in figure 12 is shown by the red curve in figure 11. The transceiver was taken to be 1010 nm above the top surface of the sample. The difference between the arrival times for the echoes in the simulation is 574 ps. To measure the geometry of structures of this type (ratio of depth to width ≥11) is difficult using atomic force microscopy. Thus the ultrasonic technique described here may be a valuable new method for characterizing the geometry of this type of structure.

In the experimental and simulated results for both samples A (figure 9) and B (figure 11), there are other features in the data whose arrival times fall between those of the echoes from the top and bottom. For example, there is a feature in both the experimental and simulated data slightly after 1600
These features are presumed to be due to the periodic nature of the nanostructures, but at this time we cannot offer a more quantitative explanation as to their origin.

![Graph showing change in reflectivity](image)

**Figure 11.** Experimental (black) and simulated (red) change in reflectivity for sample B, normalized so that the height of the first peak is unity.

**Figure 12.** Profile used for the simulation for sample B. Dimensions taken from SEM profile measurements shown in figure 8.

4. Summary and Future Outlook

In this work we have presented preliminary steps towards the construction of a new type of acoustic microscope based on optical generation and detection of sound pulses. In future work, we plan to study a series of samples to explore the effect of the trench width and profile on the shape, amplitude and time of arrival of the acoustic echoes. By increasing the temperature of the water, we can improve the signal to noise ratio. We are also developing a focusing version which uses an acoustic lens to focus sound onto individual nanostructures.

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