

## Experiments to Study the Propagation of Picosecond Sound Pulses in Water

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We report on a picosecond ultrasonics study of nanostructures using high-frequency ultrasound in water. A sound pulse is generated when an ultra short laser pulse is absorbed in a transducer structure. The sound then propagates across a thin layer of water and is reflected from the surface of the sample being examined. The efficiency of optoacoustic detection of the reflected sound is enhanced through the use of a resonant optical cavity. We report on experiments in which sound is reflected from patterned nanostructures. In these experiments we are able to study the propagation of sound down channels of width as small as 35 nm.

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### I. INTRODUCTION

The picosecond ultrasonic technique has been used extensively to study the propagation of sound pulses in a wide variety of thin films and nanostructures. An ultrashort “pump” light pulse is absorbed at the surface of the structure that is to be studied and a thermal stress is set up. This stress relaxes and a sound pulse propagates away from the surface. When a part of this sound returns to the surface, the optical “constants” of the material are modified, and this change is detected by means of a time-delayed probe light pulse [1]. In this study, we use a modified version of this technique to make sound propagation measurements in water and to investigate the extent to which one can send sound along narrow liquid channels [2].

### II. PROPAGATION OF PLANAR SOUND PULSES

Ultrasonic pulse-echo techniques provide a means of investigating absorption and dispersion mechanisms in liquids. Wright *et al.* [3] were able to measure the ultrasonic absorption and dispersion in films of liquid Hg and observe evidence of structural relaxation on the picosecond timescale. Since the liquid was optically absorbing, no special means of generating sound were required; absorption of the pump light pulse in the Hg film itself provided the initial acoustic pulse. Some sort of transducer, i.e., a structure for converting an ultrashort optical pulse into an acoustic strain, is required for making picosecond ultrasonic measurements in transparent liquids. By using a crystallographically canted film

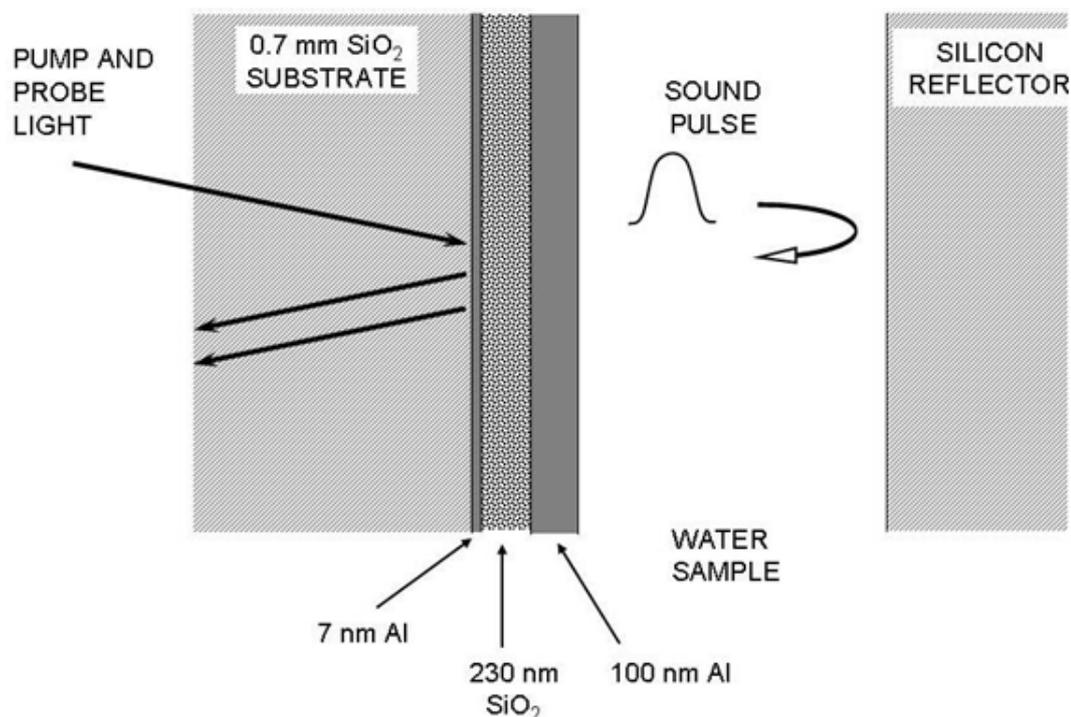


FIG. 1: Schematic diagram of the experiment for the study of planar sound propagation.

of Fe, Pezeril *et al.* [4] took advantage of the anisotropic nature of the film and launched both longitudinal and shear waves into a liquid glycerol sample. By examining the Brillouin scattering of the sound pulses, they were able to measure the attenuation of shear acoustic waves in liquid glycerol.

There are two main difficulties in making high-frequency ultrasonic measurements in water. The first is that when compared to most solid materials, the acoustic impedance of water is very low. The acoustic impedance of aluminum, for example, is approximately 11 times larger than that of water. This means that at an interface between Al and water, the acoustic reflection coefficient is 0.84 and so it is hard to get sound that is generated in a metal into or out of water. The second problem is that the attenuation in water is large and results in significant broadening of a sound pulse as it propagates. To deal with the first problem, one needs to find a way to enhance the sensitivity; and the second problem requires that the propagation distance be kept as small as possible.

The experimental set up that we have used is shown schematically in Fig. 1. A Fabry-Perot optical cavity is formed by depositing a 230 nm layer of silica between two aluminum films (thickness 7 and 100 nm). The films are deposited onto a 700 micron silica wafer. A pump light pulse passes through the substrate and deposits energy in the Al films. The thermal stress that is set up launches a sound pulse into the water and another pulse that propagates into the substrate. The pulse that enters the water travels across the water

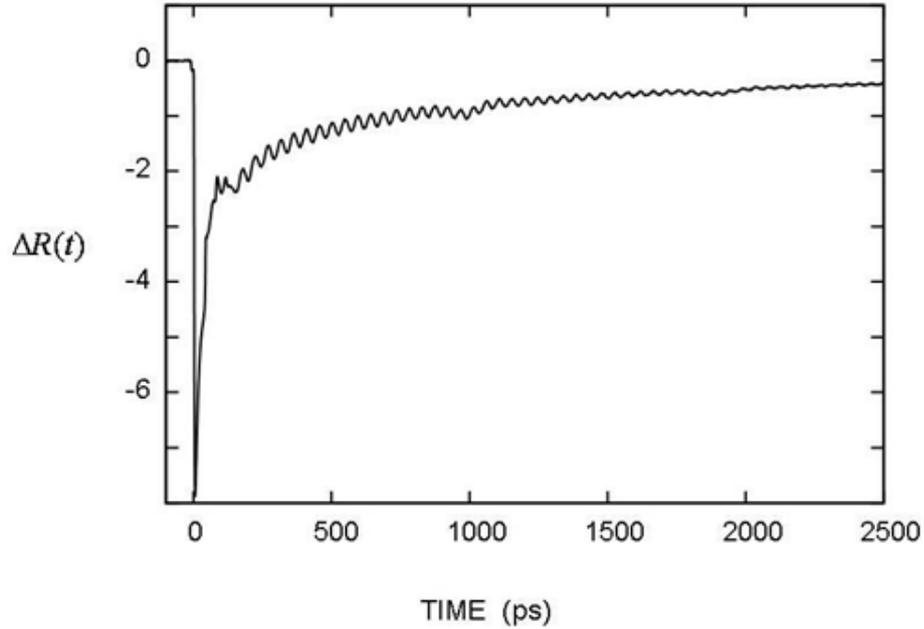


FIG. 2: Sample data showing the change  $\Delta R(t)$  in the optical reflectivity in arbitrary units as a function of the time delay  $t$  of the probe.

layer, reflects at the surface of the sample, a planar silicon wafer in this example, and then reenters the optical cavity. When the sound pulse is inside the cavity, there is a change in the cavity spacing. A probe light pulse is used to measure the change  $\Delta R(t)$  in the optical reflectivity; an optical delay stage is used to set the time difference  $t$  between the pump and the probe pulses. The light pulses used for these measurements had a wavelength of 800 nm and duration  $\sim 150$  fs. An example of typical data obtained in this way is shown in Fig. 2.

There are several contributions to the measured reflectivity change. The probe light is strongly reflected at the optical cavity and also undergoes a weak reflection when it passes through the sound pulse that propagates into the substrate. The two reflected beams go in and out of phase as  $t$  increases, and this gives rise to the periodic oscillations in  $\Delta R(t)$  (Brillouin oscillations [5]) that can be seen in Fig. 2. The period  $\tau$  of these oscillations is  $\lambda/2n\nu_s$ , where  $\lambda$  is the free space wavelength of the pump light,  $n$  is the refractive index, and  $\nu_s$  is the sound velocity. Due to the gradual attenuation of the sound pulse as it propagates in the substrate, the amplitude of these oscillations decreases slowly with increasing  $t$ . These oscillations are of no interest in the present context and can readily be removed by a simple filtering algorithm [2]. The remaining signal comes from the change in the optical reflectivity of the cavity itself. When the sound pulse that returns from the sample passes through the cavity, the reflectivity of the cavity changes because of (a) the change in the cavity spacing, (b) the change in the refractive index of the material in the cavity, and (c)

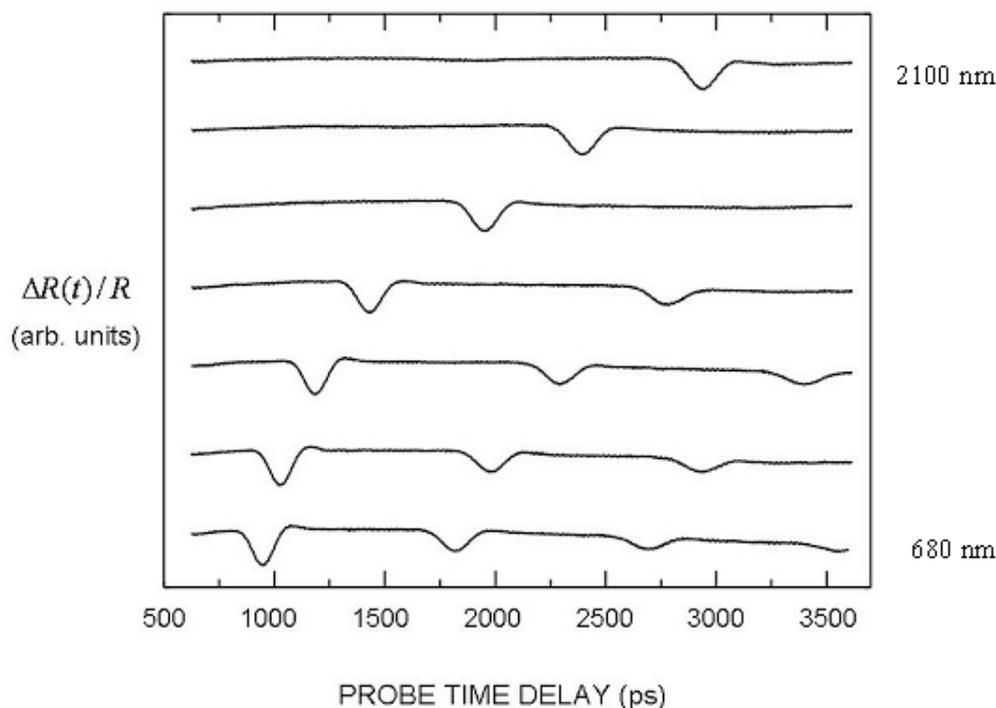


FIG. 3: The signal  $\Delta R(t)/R$  as a function of the probe delay time for a series of different thicknesses of the water layer. The data has been filtered to remove the Brillouin oscillations and the thermal background arising from the heating of the cavity. The successive echoes correspond to multiple round trips of the sound through the water layer.

the change in the optical properties of the Al films. The effects (a) and (b) have the same variation with time, but (b) is of opposite sign to (a) and about a factor of two smaller. Because the piezo-optic coefficients of the Al films are not reliably known,[6] it is hard to estimate the importance of effect (c). In addition to the changes in the cavity caused by the sound pulse, the reflectivity of the cavity is affected by the temperature change resulting from the pump light pulse. Since the temperature drops slowly as heat flows from the cavity into the substrate and into the water, this contribution varies smoothly with time and gives a background that can easily be subtracted to leave the contribution from the sound echoes. In designing the cavity to optimize the sensitivity, several factors have to be considered. One would like to make the structure out of materials that have approximately the same acoustic impedance so as to minimize the reflections of sound at the different interfaces. To be able to give a change in  $\Delta R(t)$  that accurately reflects the shape of a sound pulse, it is desirable to make the total thickness of the cavity structure as small as possible; this consideration weighs against using a DBR for one of the cavity mirrors. The quality factor  $Q$  should be as high as possible, but one cannot use a cavity that has a resonance narrower than the spectral width of the laser line. This width is 12 nm, but it can be reduced by filtering.

Sample results for a sequence of different thicknesses of the water layer are shown in Fig. 3. The thickness varies from about 680 nm to 2100 nm; these values for the thickness are simply based on the arrival time of the first echo and do not take account of the time the sound takes to get from where it is generated into the water layer and the time to get from the water layer back into the cavity where it is detected. The width of the echoes is the result of the attenuation of the high frequency components of the sound pulse. The attenuation of sound in water per unit distance varies quadratically with frequency,  $\alpha = Af^2$ , and at the measurement temperature of 23 °C, the value of  $A$  is  $2.5 \times 10^{-16} \text{ cm}^{-1} \text{ s}^2$  [7]. As a result of the attenuation, after propagation for a time  $t_0$  an initially delta-function pulse takes on a form

$$\frac{1}{\sqrt{\pi\tau}} \exp [-(t - t_0)^2/\tau^2] \quad (1)$$

where  $\tau = (Av t_0)^{1/2}/\pi$  and  $\nu$  is the sound velocity in water. This result is in very good agreement with the measured shape of the echoes as shown in Fig. 3.

### III. MEASUREMENTS ON LATERALLY PATTERNED SAMPLES

We have used this experimental technique to study sound propagation in narrow channels within nanostructures. The goal is to study the range of channel widths for which it is possible to propagate sound pulses in water. For sound propagation in bulk, the attenuation of the amplitude per unit distance of a wave with frequency can be expressed in terms of the shear  $\eta$  and bulk  $\zeta$  viscosities as

$$\alpha = \frac{\omega^2}{2\rho\nu^3} \left( \frac{4\eta}{3} + \zeta \right). \quad (2)$$

For water at 23 °C,  $\eta = 9.2 \times 10^{-3}$  and  $\zeta = 2.6 \times 10^{-2}$  cgs [8]. For propagation in a channel, there is an extra damping arising from the viscous drag due to the walls. Consider a channel with walls at  $z = \pm\omega/2$ , and suppose that at time zero, the liquid is at rest with a pressure distribution  $\delta(y)$ . This will launch sound pulses traveling in the  $y$ -direction. At early times, the resulting motion is well approximated by a propagating pulse of increasing width, whereas at long times, it is straightforward to show that the motion is essentially diffusive and the pressure distribution becomes

$$P = \sqrt{\frac{3\eta}{\pi\rho\nu^2\omega^2t}} \exp(-3y^2\eta/\rho\nu^2\omega^2t). \quad (3)$$

In the derivation of this formula, only the effect of the shear viscosity has been included because the bulk viscosity does not have a significant effect on the pressure distribution for large times. We can make a rough estimate of the time (or propagation distance) at which there is a transition between these limiting behaviors. From Eq. 3, one can see that at long times the width of the pressure distribution is of the order of  $\sqrt{\rho\nu^2\omega^2t/\eta}$ . This is to be

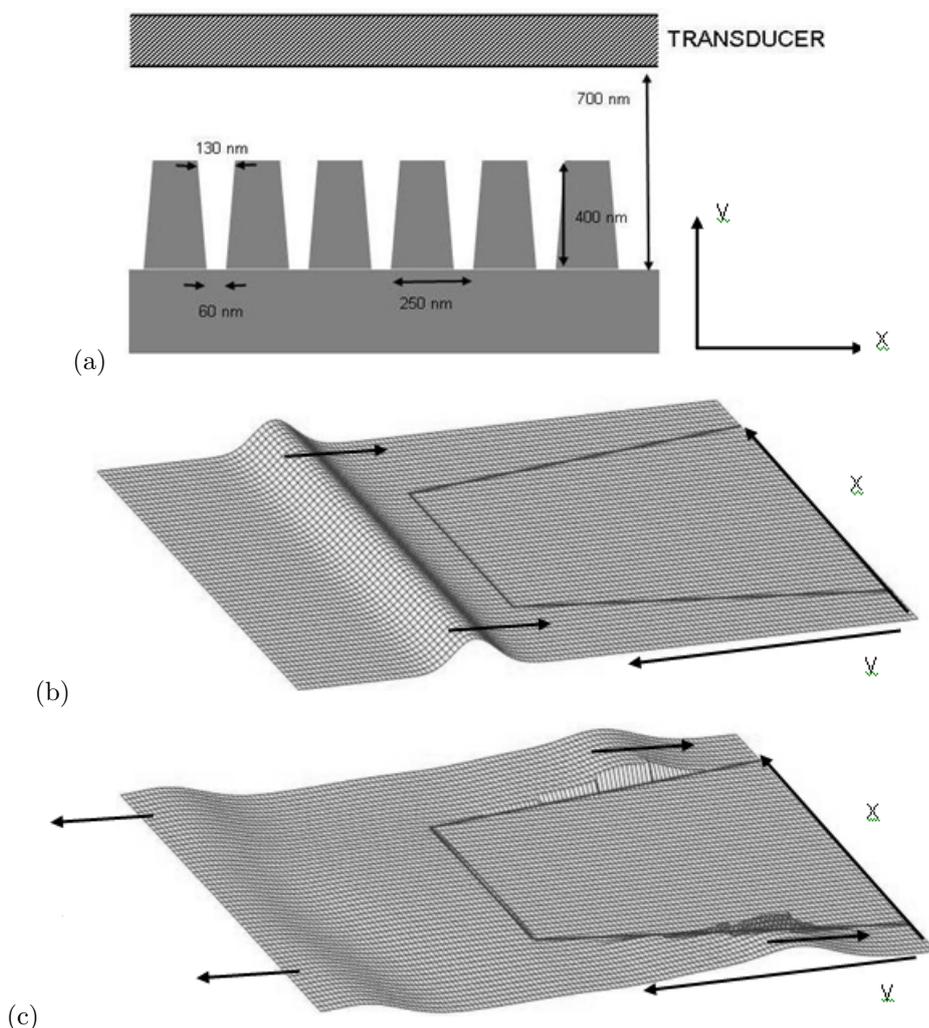


FIG. 4: Computer simulations of the propagation of a sound pulse in a nanostructure. Dimensions are given in part (a). The sound is launched from the bottom of the transducer. Parts (b) and (c) show the pressure distribution at times of 140 and 380 ps, respectively, after the sound is launched. The arrows show the direction of propagation of the different pulses.

compared with the distance  $\nu t$  that a sound pulse would travel in time  $t$  in the absence of viscosity. Comparing these two lengths shows at the time  $t^* = \rho \omega^2 / \eta$  gives there is a transition these two regimes. For a channel of width 50 nm, this time is 2.7 ns, which corresponds to a propagation distance of 4  $\mu\text{m}$ .

To investigate this more quantitatively, we have developed a computer program to simulate the propagation of sound in water in small structures (Fig. 4). The sample that is simulated consists of a series of trenches running parallel to the  $z$ -direction. The cross section of the sample in the  $xy$ -plane is shown in Fig. 4(a). The initial condition at time

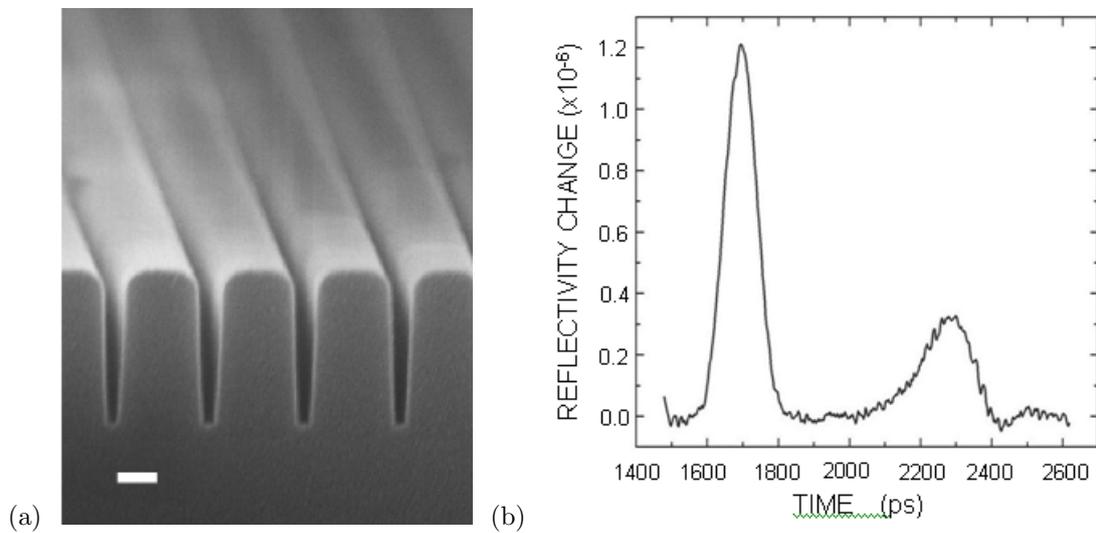


FIG. 5: (a) SEM cross section of sample. The height of the silicon dioxide lines is measured to be 405 nm, and the width of the channel halfway down is 45 nm. The white bar has a length of 100 nm. (b) Acoustic echoes from the top and bottom of the channels at a temperature of 23 °C. The data were processed in the same manner as that in Fig. 3.

zero is a sudden displacement of the lower surface of the transducer in the  $y$ -direction. Figures 4(b) and 4(c) show the pressure distribution in the water at times of 140 and 380 ps, respectively. The part of the pulse that is incident on the top part of the structure is reflected at a time of about 180 ps and results in a pulse that returns towards the transducer. The part of the pulse that enters into the grooves becomes broader as it propagates and is reflected when it reaches the bottom of the trench at around 480 ps.

We have studied a number of samples of this general type. These samples consisted of a film of  $\text{SiO}_2$  deposited onto a silicon wafer. Trenches were formed in these lines, and then the  $\text{SiO}_2$  surface was coated with a layer of silicon nitride with a thickness of approximately 5 nm. The cross-sectional SEM of one sample is shown in Fig. 5(a). The SEM gives 405 nm as the trench depth and a width half-way down of 45 nm. The repeat distance is 240 nm. The lines cover an area of about 0.1 by 0.5 mm, which is much larger than the spot sizes of the pump and the probe beam. The probe spot has a diameter of about 20 microns, which means that approximately 80 trenches are being measured at once. The measured data are shown in Fig. 5(b). The first echo at a time delay of 1700 ps is from the part of the acoustic pulse that is reflected from the tops of the lines, and the second echo that arrives at a time delay of 2280 ps is from sound that has penetrated into the channels between the lines and reflected off of the bottom. The arrival time of the first echo can be used to estimate the height of the transducer above the top of the sample; this distance is approximately 1200 nm. The difference in the arrival times for the echoes from the top and bottom is 580 ps. This can be compared with the time of 533 ps for sound to travel through 810 nm of bulk water; the effect of the interaction between the channel walls and the viscous fluid is slows

and attenuates the sound pulse.

Using this approach, we have been able to study sound propagation in channels as narrow as 35 nm. When using this technique, it is important to work with as thin a layer of water above the sample as possible, to maintain this thickness constant over the time of the measurement, and to keep the bottom surface of the transducer accurately parallel to the surface of the sample so that the water thickness does not significantly vary over the area used for the experiment. To meet these goals, we have recently constructed a high-resolution sample stage which can maintain the spacing to an accuracy of approximately 1 nm, and parallelism to 10 microradians. Results obtained with this instrument will be reported in a separate and detailed paper, along with a discussion of the possible use of the technique for the measurement of the geometry of small structures.

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