Study of phonon propagation in water using picosecond ultrasonics

F Yang, T Atay, C H Dang, T J Grimsley, S Che, J Ma, Q Zhang, A V Nurmkko and H J Maris

1Department of Physics, Brown University, Providence, RI 02912, USA
2Division of Engineering, Brown University, Providence, RI 02912, USA

E-mail: Humphrey_Maris@brown.edu

Abstract. The propagation of ultra-short sound pulses in water is studied by using the picosecond ultrasonic technique. A sound pulse is generated when light is absorbed in a metal transducer film deposited onto a substrate. The sound propagates across a thin layer of water and is then reflected back to the surface at which it was generated. The efficiency of optoacoustic detection of the reflected sound is enhanced through the use of a resonant optical cavity. We show that the variation of the shape of the returning sound pulse with propagation distance agrees with that calculated by using the attenuation of sound in water that has been measured at lower frequencies.

1. Introduction
The propagation of sound in liquids has been widely studied in the past decades. The velocity and attenuation of phonons in liquids in the MHz frequency range has been measured by the pulse-echo ultrasonic technique [1]. Brillouin scattering [2] and picosecond acoustic interferometry [3] [4] measurements have been made for some liquids in the frequency range 1 to 10 GHz. However, it is difficult to make measurements at higher frequencies with the existing techniques. Furthermore, the frequency band that any of these techniques can reach is limited. In this paper we describe a method for studying the acoustic attenuation in water over a broad frequency band. In principle, this method is expected to extend the frequency band in which quantitative measurements can be made to 20 GHz or higher.

2. Experiment
The experiment uses the picosecond ultrasonic technique [5]. The setup is shown schematically in figure 1. The distilled water sample is confined between a silicon substrate and a glass slide on which the optical cavity-based acoustic sensor is positioned. The surface roughness of the silicon substrate, the glass slide and the films forming the optical cavity were examined by a white light interferometer. The average roughness of the silicon substrate is less than 0.3 nm and those of the other surfaces are less than 2 nm, over a 60 µm by 60 µm area. The optical sensor cavity, a planar $\lambda/2$ thick Fabry-Perot interferometer, is composed of two aluminum layers with 5 nm and 70 nm in thickness and a 220 nm

3 Author to whom any correspondence should be addressed.
thick silicon dioxide layer. The aluminum layers were deposited using a sputtering system with base pressure $2 \times 10^{-9}$ Torr and the SiO$_2$ layer was deposited in a PECVD system.

The light source used for both the pump and the probe is a Diode-Pumped Mode-Locked Ti:Sapphire Laser with pulse width about 200 fs (FWHM), repetition rate 80 MHz and wavelength 800 nm. The pump pulse has energy about 0.2 nJ and the probe pulse energy is 4 times smaller. The two beams pass through the glass slide and are focused onto the same spot on the 5 nm Al film. The spot is about 20 µm in diameter. The pump beam is modulated at 1 MHz by an electro-optic modulator. The pump and probe beam are orthogonally polarized in order to prevent the scattered pump light from being collected by the polarization-sensitive photodetector.

![Figure 1. Schematic diagram of the experiment.](image)

The absorption of the pump light pulse energy results in thermal stresses being set up in the two Al films of the optical cavity. The thermal stresses relax and launch compression sound pulses into both the glass and the water. The sound launched into the water propagates across the thin water layer (0.4–3 µm thick) and is then reflected by the silicon substrate back to the optical cavity in which it was generated. The returning sound is detected with the time-delayed probe light pulse as changes of optical reflectance of the cavity, designed to enhance the optoacoustic detection of the returning sound. The thickness of the top Al layer (5 nm) of the optical cavity is less than the optical absorption length of the probe light (7.5 nm) [6], allowing the probe light to penetrate into the cavity and be reflected by the bottom Al layer for setting up the standing light waves. The SiO$_2$ layer is made to be 220 nm thick so that the reflectivity of the optical cavity at the probe light wavelength (800 nm) is most sensitive to the perturbation of the optical thickness of the SiO$_2$ layer due to the returning sound pulses. With this design the sensitivity of the optical cavity, $\frac{dR}{dl}$, is about 0.03 nm$^{-1}$ with $R$ being the reflectivity and $l$ the optical thickness of the silicon dioxide layer. Our calculations show that, with the optical cavity, the reflectivity change due to the returning sound is enhanced by a factor of about 40–70, compared to the detection scheme employing a single aluminum film sensor only.

The intensity of the reflected probe light is measured with a photodetector. A polarizing cube beamsplitter is used to reject the scattered pump. The signal is fed into a lock-in amplifier. A computer-controlled mechanical delay stage is used to time-delay the probe pulse in the range out to 7 ns.

### 3. Results and discussion

Data taken at room temperature (25 °C) are shown in figure 2. The curves from bottom to top are the measured reflectivity change as a function of the delay time of the probe light pulse with the thickness of the water layer being about 0.7 µm, 1.4 µm, 2 µm, 2.8 µm and 3.4 µm, respectively. The curves show a damped periodic oscillation and some Gaussian-like structures, together with a smoothly varying background contribution. The periodic oscillation is the acoustic oscillation [4] due to the propagation of sound pulses in the glass slide. The slow background arises from the change in the reflectivity of the optical cavity as its temperature relaxes after the energy of the pump light pulse is absorbed. The Gaussian-shaped echoes are the responses of the optical cavity to the returning sound.
pulses after they travelled through the water layer. Multiple echoes can be seen in some of the curves, as a result of the fact that the sound pulse bounces back and forth across the water layer.

Figure 3 plots the echoes after the background and the acoustic oscillation are removed. From bottom to top, the propagation distances of the acoustic pulses are 1.4 µm, 2.8 µm, 4 µm, 5.6 µm and 6.8 µm, respectively. These distances are estimated from the echo arrival time and the measured low frequency sound velocity [7]. The width of the echo increases with the propagation distance, indicating that the sound pulse is broadened as it propagates through the water due to the acoustic attenuation.

Figure 2. Measured reflectivity change as a function of the delay time of the probe light pulse. From bottom to top, the curves show the reflectivity data with water layer thickness 0.7 µm, 1.4 µm, 2 µm, 2.8 µm and 3.4 µm.

Because of the acoustic attenuation in water, a sinusoidal sound wave decreases in amplitude as it propagates through water. Its waveform as a function of time and position is given by

\[ \exp[i2\pi f(t - z/c)]\exp(-\alpha z) \]

where \( f \) is the frequency of the sound wave, \( c \) the sound velocity in water, \( z \) the propagation distance and \( \alpha \) the attenuation coefficient. Experiments performed in the MHz frequency range [1] [7] and Brillouin frequency range [2] indicate that \( \alpha \) varies with the square of the frequency (\( \alpha = 19.1 \times 10^{-15} \text{sec}^{-1} / \text{m} \) at 30 °C [7]). The change in the shape of a sound pulse of strength such that its integral is unity can be written as

\[
\eta(z' - z) = \left(\frac{\beta c^2 z}{\pi}\right)^{-1/2} \exp \left[ -\frac{\pi^2 (z' - z)^2}{\beta c^2 z} \right]
\]  \hspace{1cm} (1)

where \( \beta = \alpha / f^2 \), \( z \) is the propagation distance and \( z' \) the distance from the center of the pulse. The width of the Gaussian grows with the square root of the propagation distance \( z \).

The reflectivity change induced by the returning sound pulse is proportional to the stress accumulated in the optical cavity and it is given by

\[
\Delta R(T) \propto \int_{t - \tau/2}^{t + \tau/2} dt' \left(\frac{\beta c t}{\pi}\right)^{-1/2} \exp \left[ -\frac{\pi^2 (t' - t)^2}{\beta c t} \right]
\]  \hspace{1cm} (2)

where \( \Delta R \) is the optical reflectivity change, \( T \) the delay time of the probe light pulse, \( \tau \) the transit time of sound pulse through the optical cavity, \( t' = z'/c \) and \( t = z/c \). With \( \beta = 22 \times 10^{-15} \text{sec}^2 / \text{m} \) at 25 °C obtained by interpolating the ultrasonic attenuation data in [7], the reflectivity change as a function of time delay is calculated from equation (2). Some of the results are plotted in figure 4 together with...
the corresponding measured echoes from figure 3. The comparison between the calculation results and the experimental data shows that the variation of the shape of the returning sound pulse with propagation distance measured in our experiment is consistent with that calculated by using the attenuation coefficient measured at lower frequencies. In our experiment the returning sound pulses contain a broad band of frequencies up to 10 GHz.

![Graph](image1.png)

**Figure 4.** The reflectivity change as a function of time calculated with equation (2) (solid lines) and the corresponding echo obtained experimentally (open circles). The left and right panels show the echoes with transit time 960 ps and 3770 ps respectively.

This technique makes it possible to study quantitatively the acoustic attenuation of liquids over a broad frequency band. By reducing the thickness of the water layer, one could extend the frequency band of the returning sound pulse to 20 GHz or higher. With the current setup, the smoothly varying background has to be removed before the data can be analyzed, and this limits the accuracy of the attenuation measurements. However, we plan to avoid this difficulty by applying pump light pulse to a separate transducer in stead of the optical cavity. This work will be presented in a future publication.

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