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Coherent Brillouin spectroscopy in a strongly scattering liquid by picosecond ultrasonics

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In a modification of a picosecond ultrasonic technique, a short acoustic pulse is launched into a liquid sample by a laser pulse absorbed in a semitransparent transducer film and is detected via coherent Brillouin scattering of a time-delayed probe pulse. With both excitation and probing performed from the transducer side, the arrangement is suitable for *in vivo* study of biological tissues. The signal is collected from a micrometer-thick layer next to the transducer and is not affected by the diffuse scattering of probe light deeper in the sample. The setup, utilizing a 33 nm thick single crystal SrRuO₃ transducer film, is tested on a full fat milk sample, with 11 GHz acoustic frequency recorded. © 2011 Optical Society of America

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Brillouin spectroscopy based on light scattering by acoustic waves is an established tool for studying the viscoelastic properties of materials [1,2]. The potential of Brillouin scattering for the characterization of biologically relevant materials has long been appreciated [3]; however, limitations have also been recognized. The method, in regard to liquids or soft tissues, requires a transparent sample, which significantly narrows the potential range of biological applications. Another limiting factor is related to the very low intensity of the scattered signal. The signal level is proportional to the volume of the sample from which the scattered light is collected, and this limits the benefits that might otherwise be expected from an optical technique, i.e., the ability to measure very small samples and to perform scanning and imaging with high spatial resolution. Recently, a significant improvement in the throughput of Brillouin spectroscopy has been achieved [4], enabling imaging of elastic properties of tissue with scanning confocal microscopy. Still, the dimensions of the focal area were larger than 10 μm and the measurements were performed on transparent eye tissue.

Alongside progress in traditional Brillouin spectroscopy, advances have been made in coherent Brillouin scattering techniques involving light scattered by coherent phonons that are excited by a short laser pulse, which yields a much greater signal than scattering from thermal phonons. Coherent phonons can be excited by crossing laser beams inside the sample of interest, the method known as impulsive stimulated scattering [5], or by pulsed excitation of a light-absorbing “transducer” layer that launches a short acoustic pulse into a transparent sample [6]. The latter technique, referred to as picosecond acoustic interferometry [6,7], has recently been used for measurements inside a vegetable cell with lateral resolution of ~2 μm [8,9]. The attenuation length of acoustic waves of 5–10 GHz frequency in the cell tissue was on the order of a micrometer, which means that only a micrometer-thick layer of the sample next to

the transducer was probed. On one hand, it limits the technique to studying near-surface layers. On the other hand, it offers a prospect of measurements on highly scattering samples, because only a micrometer-thick layer needs to be transparent. Studies performed to date [7–9] have not taken advantage of this potential benefit as they used an arrangement with excitation and probe beams entering from the sample side. Such an arrangement requires a whole sample to be transparent and limits the applications of the method to *in vitro* measurements. In this report, we demonstrate a modification of the technique employing a semitransparent transducer film, with both excitation and probing performed from the transducer side. Such a configuration has already been employed for studying acoustic attenuation in silica glass [10,11], where a very thin metal film was deposited on the sample of interest. For measurements in liquids or biological tissues, the transducer film should be deposited on a transparent substrate and brought into contact with the sample.

The arrangement is shown schematically in Fig. 1(a). Absorption of the excitation pulse in the transducer film causes rapid thermal expansion, which launches a short acoustic pulse into the sample. Part of the time-delayed probe pulse is transmitted through the transducer film, reflected (i.e., coherently scattered) from the acoustic pulse, and transmitted again through the transducer film back into free space and toward a photodetector. A much stronger reflection of the probe pulse from the transducer film itself serves as a local oscillator or “reference” field that copropagates and superposes with the signal field, resulting in either constructive or destructive interference depending on the time-dependent propagation distance of the acoustic pulse from the transducer. The light intensity at the detector will contain an interference term varying as a function of the probe time delay t as [6]

$$I(t) \propto \cos(\omega t) \exp(-\gamma t), \quad (1)$$

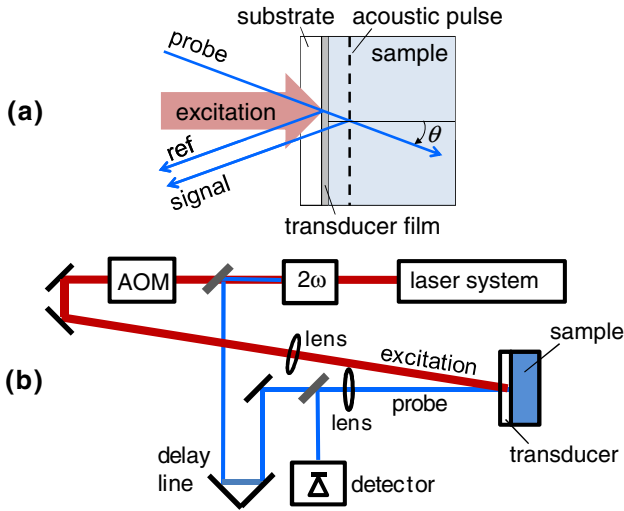


Fig. 1. (Color online) (a) Schematic diagram of the experiment and (b) outline of the optical setup.

where $\omega(q)$ and $\gamma(q)$ are the angular frequency and attenuation rate of acoustic waves at the wavevector q determined by the Brillouin scattering geometry:

$$q = \frac{4\pi n \cos(\theta)}{\lambda}, \quad (2)$$

where λ is the probe wavelength, n is the refractive index of the sample, and θ is the angle of the probe beam inside the medium with respect to the normal to the transducer surface.

Traditionally in picosecond ultrasonics, metals with a small optical skin depth such as Al and Ti have been used for transducer films [6,7,10,11]. To be semitransparent, these metal films should be typically ~ 10 nm or less in thickness [10,11]. Such an ultrathin metal film transducer may not be very stable over time when brought into contact with various samples; hence, a protective coating would be needed. Furthermore, the acoustic pulse generated by an ultrathin transducer film has a spatial length on the order of the film thickness, which means its spectral content is centered at comparably short wavelengths and high frequencies, typically in the hundreds of GHz. This is not optimal for Brillouin scattering, which selectively probes the frequency determined by wavevector q , typically not much greater than 10 GHz in liquids [7–9].

In this work, we used a thicker semitransparent transducer made of strontium ruthenate SrRuO_3 (SRO), a metallic oxide with a perovskite structure that attracted attention as a promising photoacoustic transducer material after a high laser-induced strain amplitude in an $\text{SRO-PbZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$ structure was reported [12]. High-quality single crystal SRO films can be grown on perovskite substrates such as SrTiO_3 (STO) [13]. For this study, a single crystal 33 nm SRO layer was deposited on a 1 mm thick (100) STO substrate by 90° off-axis sputtering [14]. Optical constants of SRO are not found in the literature: the transmittance of our structure was 43% at $\lambda = 790$ nm and 13% at 395 nm, and the transmittance of the STO substrate alone was 70% at 790 nm (Fresnel losses only) and 28% at 395 nm, the decrease caused by the onset of UV absorption in STO. The Vickers hardness of SRO is

12.7 GPa [15] (compare with 0.17 GPa for Al), which is a benefit for a transducer film that has to be brought in contact with samples and cleaned after use, as is the fact that SRO is chemically very stable and, being an oxide material, not subject to oxidation. The densities of SRO and STO are 6.49 and 5.12 g/cm³, while the respective longitudinal acoustic velocities are 6.31 km/s [15] and 7.88 km/s [16]. The acoustic impedances of the two materials are almost perfectly matched.

The transducer formed a wall of a 1 mm thick cuvette holding a liquid sample. The optical setup is shown in Fig. 1(b). An amplified Ti:sapphire laser system (pulse duration 300 fs, repetition rate 250 KHz) was used to produce excitation pulses at $\lambda = 790$ nm, pulse energy 0.5 μJ and probe pulses at $\lambda = 395$ nm, pulse energy 2.5 nJ. The excitation beam, modulated at 93 KHz frequency by an acousto-optic modulator (AOM) to facilitate lock-in detection of the signal, was focused on the transducer film to a $90 \mu\text{m}$ spot (defined at $1/e$ intensity level), and the time-delayed probe pulse was focused to a $50 \mu\text{m}$ spot aligned with the center of the excitation spot. Measurements were done in the backscattering geometry at $\theta = 0$. Probe light reflected from the sample was directed to a photodiode whose output was fed to a lock-in amplifier. The data were averaged over multiple scans of an optical delay line, with the total data collection time of ~ 10 min. An estimate of the temperature rise at the transducer surface due to the average laser power yields $\Delta T \sim 10$ K. At a given intensity level, the steady-state temperature rise is proportional to the spot size [17]; hence, it can be reduced by reducing the excitation spot size while keeping the signal level unchanged.

We used 3% fat milk obtained from a local convenience store as a model scattering medium. Figure 2(a) shows the signal from the milk sample dominated by a large peak due to electronic excitation in SRO and nonlinear optical response in the STO substrate. The tail of the signal contains oscillations at 11 GHz frequency due to light scattering by acoustic waves in milk. The signal also contained oscillations at ~ 110 GHz frequency due to the acoustic pulse launched into the STO substrate; the data shown in Fig. 2 were taken with the lock-in integration constant and the delay line speed selected in such a way as to filter out this high-frequency signal component.

Figure 2(b) shows the Fourier spectrum of the acoustic oscillations, obtained after subtracting the slow background, along with a spectrum obtained on a distilled water sample. The acoustic frequency in milk is 4% larger compared to water. About one-third of this difference may be attributed to a higher refractive index of milk [18] leading to a higher acoustic wavevector according to Eq. (2). The remaining difference should be due to a slightly higher acoustic velocity in milk. The latter has not been systematically studied; for skim milk at 40 $^\circ\text{C}$, speed of sound of 1548 m/s and density of 1.026 g/cm³ have been reported [19], both values slightly higher than those of water, 1529 m/s and 0.992 g/cm³ at the same temperature. The difference in linewidth is more significant, with the milk sample yielding a 60% larger linewidth. The increased acoustic attenuation may be due to acoustic scattering by the same sorts of heterogeneities, such as globules of fat that scatter light; inhomogeneous

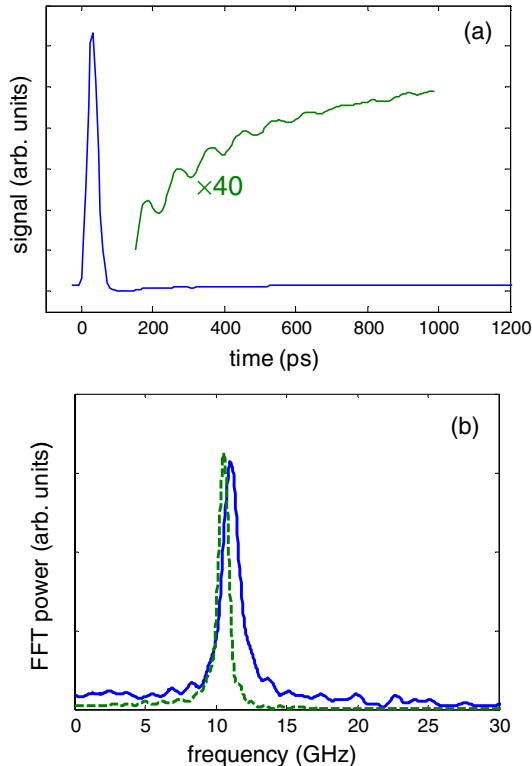


Fig. 2. (Color online) (a) Signal from a milk sample and the tail of the signal magnified by a factor of 40, revealing acoustic oscillations, and (b) spectra of acoustic oscillations in the milk sample (solid curve) and distilled water (dashed curve).

broadening due to sample nonuniformity within the probe spot is also possible.

The signal contains ~ 6 acoustic oscillations, which implies that the probed region of the sample is $\sim 1 \mu\text{m}$ thick. It is desirable that the sample be transparent over at least this thickness. Absorption or diffuse scattering of probe light within the acoustic propagation path would lead to an apparent increase in the measured acoustic attenuation rate. However, diffuse scattering of the probe beam deeper in the sample does not have any effect on the signal. Even if most of the probe power inside the sample is diffusely scattered, the intensity of diffusely scattered probe light that reaches the detector will be far lower than the intensity of the local oscillator (i.e., the probe beam specularly reflected from the transducer film).

Besides the ability to perform measurements on strongly scattering samples, our setup has an advantage compared to prior work [7–9] in that it only requires access to the sample from one side. This could enable *in vivo* measurements of biological tissues. A transducer film could be deposited onto the polished end of an optical fiber to enable access to tissues of interest. Alternatively, it would be straightforward to integrate a microscope objective into the setup to achieve $\sim 1 \mu\text{m}$ lateral spatial resolution. We note that the spatial resolution is determined by the probe spot size while the excitation spot size can be much larger. Hence the objective would only need to be optimized for the probe wavelength. For scanning/imaging applications the measurement time would need to be reduced; in our experiments the

signal-to-noise ratio was not close to the shot-noise limit and multiple avenues for noise reduction can be pursued.

In conclusion, we have demonstrated that the use of a semitransparent single crystal SRO transducer film to launch laser-generated acoustic waves into a liquid sample makes it possible to perform coherent Brillouin scattering measurements in strongly scattering media. We believe that despite certain limitations, i.e., the need for contact between the transducer and the sample and the small depth of probing, the method has potential for the study of viscoelastic properties of biologically relevant samples.

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