Laser-induced generation of subnanosecond sound pulses in liquids

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The generation of intense sound pulses of subnanosecond duration induced by lasers has been observed for the first time. The liquids used in the experiment (water, ethanol, glycerine, etc.) contained hydroxyl groups and hydrogen bonds and absorbed very strongly at the laser wavelength ($\lambda = 2.94\,\mu$m); for water, e.g., $\alpha = 1.3\cdot 10^4$ cm^{-1}. The sound was generated by irradiating the liquid adjacent to a silica surface by ultrashort laser pulses ($\tau \approx 80$ ps) of energy up to 60 μJ; the area of the laser beam was $2.5\cdot 10^{-5}$ cm^2. Single sound pressure pulses of duration 0.75 ns and amplitude 20 kbar were obtained in water; this corresponds to a 5.8% efficiency for conversion of light into sound energy, and to a sound intensity of 185 MW/cm^2. Some generation mechanisms are considered, and it is shown that such intense sound waves in liquids must be treated as shock waves.

I. INTRODUCTION

This paper is concerned with the direct generation of intense subnanosecond sound pulses in liquids irradiated by single ultrashort pulses from an erbium garnet laser ($\lambda = 2.94\,\mu$m). Laser generation of short sound pulses in liquids was facilitated by the fact that the wavelength used in our experiments coincided with the IR absorption peak associated with stretching vibrations of the OH-group in hydroxyl-containing liquids (water, ethanol, glycerine, etc.)—the absorption coefficient at 2.94 μm was extremely high ($\alpha = 1.3\times 10^4$ cm^{-1} for water, for example[1]). Until recently, the maximum absorption coefficient in liquids at typical laser wavelengths was $\alpha = 800$ cm^{-1} in water for a CO_2 laser ($\lambda = 10.6\,\mu$m). However, simple physical considerations show that the shortest sound pulse that can be generated by irradiating an absorbing material is equal to the time required for the sound to travel one absorption length (α^{-1}):

$$\tau_{s.\min} = 1/\alpha c_s\,, \qquad (1)$$

where c_s is the speed of sound. For water, the absorption at 2.94 μm is $\alpha = 1.3\times 10^4$ cm^{-1}, $c_s = 1.5\times 10^5$ cm/s, and $\tau s_{\min} = 0.5$ ns.

The purpose of the experiments was also to investigate laser-induced transparency in water and other hydroxyl-containing liquids at 2.94 μm [this effect was discovered in 1982 (Ref. 2)]. The laser-induced transparency can plausibly be attributed to a shift in the absorption spectrum of the liquid toward shorter wavelengths as hydrogen bonds are broken by heating and by the effects of the resonant laser radiation. Sound-generation experiments in these liquids also make it possible to measure the pressure in the irradiated region, which is important if we are to understand the processes that occur.

Laser generation of short acoustic pulses in solids has been reported in the literature. Sound pulses of length $\tau_s = 1$ ns were excited in a metal foil,[3] in a semiconductor film,[4] and in dielectrics (quartz, Teflon, etc.) coated with a layer of strongly absorbing material.[5] However, the properties of the sound pulses were not measured directly, and the estimated amplitude was quite small.

II. EXPERIMENT

In our experiments we employed an yttrium-erbium-aluminum garnet ($Y_3Al_5O_{12}$:Er^{3+}, $\lambda = 2.94\,\mu$m) laser which was actively mode-locked by using an electrooptic crystal to synchronously modulate the intracavity radiation.[6,7] The laser generated a train of ultrashort TEM_{00} pulses at a repetition rate of 1 Hz; there were roughly 50 pulses in each train, and each pulse was 80 ± 20 ns long. A single pulse of energy $\leqslant 60\,\mu$J was selected electrooptically and directed into a cell containing the liquid. Nearly all the incident laser energy was absorbed in a very thin layer (of width $\sim 1\,\mu$m for water) in contact with the quartz surface, and sound was generated in the liquid layer between the two quartz substrates.

The cell (Fig. 1) contained a 10-μm thick gap. A rapid-response pressure sensor of sensitivity $2\cdot 10^{-7}$ V/Pa was located either in direct contact with the liquid (Fig. 1a) or, alternatively, on the back side of the quartz substrate (Fig. 1b). The pressure sensor was 1.2 μm thick and consisted of a polycyclic oriented organic compound; it was sandwiched between two thin aluminum electrodes and enabled us to

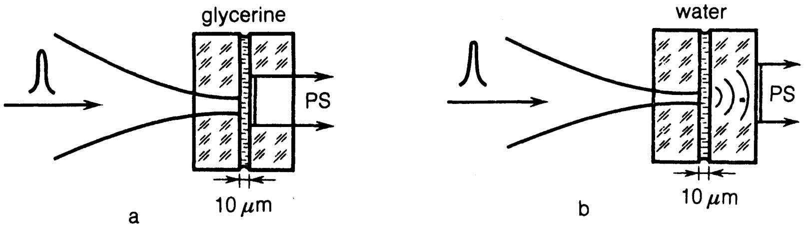

FIG. 1. Experiment on sound generation in liquids: a) pressure sensor (PS) in contact with the liquid; b) pressure sensor deposited on the back wall of the quartz substrate.

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detect the sound wave in the near (acoustic) wave zone independently of the radius w_0 of the laser spot, which varied from $w_0 = 0.04$ mm to $w_0 = 0.5$ mm in the experiment. Although the location of the pressure sensor in Fig. 1a was optimal in terms of matching the acoustic impedances it was feasible only for glycerine, which did not dissolve the pressure-sensitive element. The quartz substrate bounding the irradiated surface of the liquid ensured that the latter was rigid when the sound pulses were generated.

In the configuration shown in Fig. 1b, the pressure sensor was located either in the near or in the far (acoustic) wave zones, depending on the size of the laser spot (and hence on the dimensions of the acoustic source), i.e., on the value of the diffraction parameter D_1:

$$D_1 = \lambda_s L / w_0^2 ,$$

where λ_s is the acoustic wavelength in quartz and L is the thickness of the quartz substrate.

For $\tau_s = 0.75$ ns,

$$\lambda_s = \tau_s c_s^{\text{quartz}} = 0.75 \cdot 10^{-9}\ s \cdot 6 \cdot 10^5 \text{ cm} \cdot \text{s}^{-1} = 4 \cdot 10^{-4} \text{ cm}$$

and for $L = 1$ cm we have

$$\text{for } w_0 = 0.04 \text{ mm} \quad D_1 = 25 \gg 1 \quad \text{(far zone)} ,$$

$$\text{for } w_0 = 0.5 \text{ mm} \quad D_1 = 0.16 \ll 1 \quad \text{(near zone)} .$$

The laser pulse energy in our experiments varied from 0 to 60 μJ, which for $w_0 = 0.04$ mm corresponded to a maximum laser power density $I_{\text{las}} = 30$ GW/cm^2.

III. EXPERIMENTAL RESULTS

Figure 2a shows an oscilloscope trace of an acoustic pulse generated in glycerine (near acoustic zone) for the configuration indicated in Fig. 1(a). We note that the sound generation mechanism was thermooptic (thermoelastic) in all of the experiments, but evaporation did not occur because the closed cell kept the liquid from boiling. In the near zone, a unipolar acoustic compression pulse[8,9] is characteristic for the thermooptic mechanism when the liquid is rigidly bounded. The sound pulse in glycerine was considerably longer than in water ($\tau_s = 3$ ns as compared with $\tau_s = 0.75$ ns), which agrees with (1) since the absorption coefficient for glycerine at $\lambda = 2.94$ μm is several times smaller. The peak sound pressure in glycerine ($w_0 = 0.04$ mm) was 14 kbar (1 bar $\approx$ 1 atm). The traces in Fig. 2a, b reveal nonlinear distortions of the acoustic pulseform (asymmetry); they are present at these high pressure amplitudes because the top of the pulse propagates faster than the base.

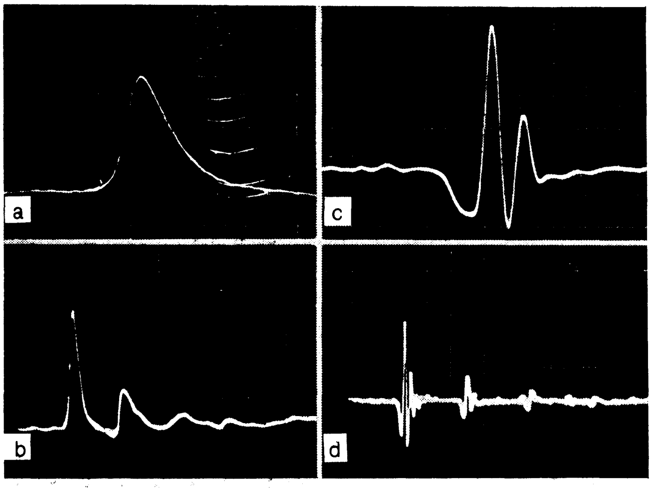

FIG. 2. Traces of acoustic pulses recorded in glycerine (a,b) and (c,d); the sweep rate was 2 ns/div (a,c) and 10 ns/div (b,d). A positive pressure corresponds to upward or downward deflections of the oscilloscope beam (traces a, b and c, d, respectively).

The trace in Fig. 2b (sweep rate 10 ns/div) shows a series of acoustic pulses generated when the excited pulse was successively reflected by the walls of the cell.

The traces in Fig. 2c, d show acoustic pulses in water in the near (acoustic) wave zone for the configuration in Fig. 1b. In contrast to the previous case, a bipolar signal is now observed. We attribute this to the mismatch in the acoustic impedances (Fig. 1b), which gave rise to a reflected signal that was picked up by the sensor. The signal polarity is reversed because the pressure pulse was inverted upon reflection from the interface between the pressure sensor and the surrounding air. This is illustrated in Fig. 3, which also shows the analytically calculated response of the pressure sensor (for the configuration in Fig. 1b) to a unipolar Gaussian sound pressure pulse of length $\tau_1 = d_1/c_1$, where d_1 is the thickness of the pressure-sensitive layer and c_1 is the speed of sound. In our case $d_1 = 1.2$ μm, $c_1 = 2.6 \cdot 10^5$ cm/sm, and $\tau_1 = 0.46$ ns (τ_1 is the "instrument function" of the pressure sensor. The qualitative similarity between the signals in Fig. 2c and Fig. 3 is apparent, provided we note that positive pressures in Fig. 2c correspond to a downward deflection of the oscilloscope beam. The experimental duration of the positive part of the pressure signal in Fig. 2c, corresponding to a sound pulse in water, was equal to 0.75 ns.

Figure 2d shows the same signal in water at a slower sweep rate. As in Fig. 2b, there is a sequence of reflected acoustic pulses which are separated by an interval equal to the time needed for the sound to make a round trip through the liquid layer. The attenuation of the sound in the liquid can be calculated from the (energy) reflection coefficient R for sound at the water-quartz interface. For water $R = 0.64$, whence the attenuation of the sound pulses is found to be $\alpha_s = 10^2$ cm^{-1} (the sound propagation length was 100 μm).

If we use an f^2 dependence to rescale the tabulated values for the acoustic absorption to frequencies $f \sim 1$ GHz corresponding to $\tau_s = 0.75$ ns, we find that the sound is damped over distances of 40 μm in water and 5 mm in quartz.

At the maximum laser pulse energy (60 μJ, spot radius $w_0 = 0.04$ mm), the amplitude of the acoustic pressure in water was 20 kbar near the point of generation (we corrected for diffraction when calculating the pressure in the near zone, because the sound was recorded in the far wave zone).

The acoustic pulse amplitudes for optoacoustic generation in ethanol and ethylene glycol were an order of magnitude lower than for water.

Figure 4 plots the sound pressure amplitude p_s as a function of the laser light intensity I_{las} for water and glycerine; we see that the curves are nearly linear with slope ≈ 1.

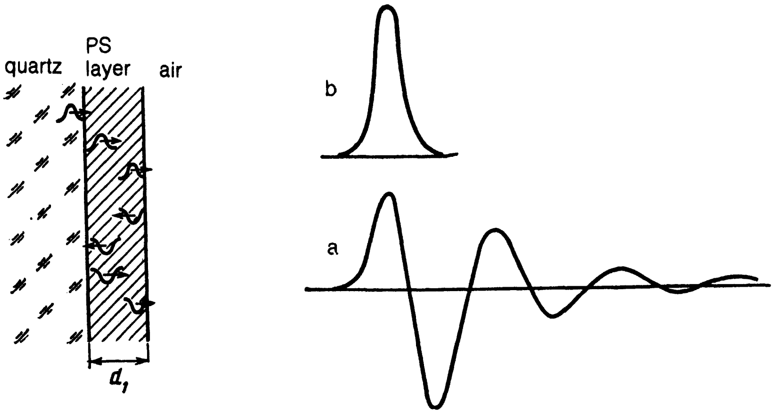

FIG. 3. Response (a) of the pressure sensor (PS) to a short unipolar sound pressure pulse (b).

The acoustic pulseforms remained qualitatively unchanged for all specific energy inputs investigated, which suggests a thermooptical generation mechanism. The curve $p_s(I_{las})$ becomes appreciably nonlinear for $I_{las} > 10^{10}$ W/c^2. We attribute the less efficient sound generation at these power densities to an increase in the transparency of water to the laser radiation (the effective absorption coefficient α decreased). Similar behavior was observed for glycerine.

The above explanation is further supported by data obtained in another experiment (Fig. 5). Figure 5 plots the cell transmission T as a function of the incident laser energy for a 5-μm-thick cell containing water. We see that at $I_{las} = 10^{10}$ W/cm^2, the optical density of the water layer dropped substantially due to bleaching[2] (the cell transmission increased fourfold).

IV. PHYSICAL MECHANISMS

We conclude from the experimental results that since the energy was input to the liquid molecules through the excitation of stretching vibrations, while the sound was generated due to thermal expansion of the medium, the shortness of the sound pulses indicates that the excited molecular states were thermalized during times < 1 ns, since otherwise the acoustic pulse would have been stretched out.

Let us theoretically estimate the maximum pressure that can be generated in water at the laser power densities employed in the experiment. The characteristic laser-induced pressure in the liquid is equal to the Grüneisen coefficient $\Gamma_0 = \beta c_s^2/c_p$ multiplied by the energy deposited per unit volume[10]:

$$p \approx \Gamma_0 I_{las}\tau_{las}/l, \tag{2}$$

where β is the bulk expansion coefficient and c_p is the specific heat at constant pressure; the characteristic depth l of the laser-heated region is given by $1 = \max\{\alpha^{-1}, (\xi\tau_{las})^{1/2}\}$, where α^{-1} is the light penetration depth in the liquid and $(\chi\tau_{las})^{1/2}$ is the characteristic length of heat conduction (provided l exceeds the distance traveled by sound during the time τ_{las}). Since under our conditions $\alpha^{-1} \gg (\chi\tau_{las})^{1/2}$, $c_s\tau_{las}$ we have $l = \alpha^{-1}$ and for water at room temperature $\Gamma_0 = 0.1$. For $I_{las} = 30$ GW/cm^2 and $\tau_{las} = 80$ ps, the estimated sound pressure is 30 kbar. The agreement with the experimental value (20 kbar) is quite satisfactory, particularly since the penetration depth α^{-1} may increase due to bleacing at these high laser powers. Under the experimental conditions, the increase in temperature and pressure did not greatly alter the Grüneisen coefficient (the empirical equation of state for water in Ref. 11 gives $\Gamma = 0.2$ for $T \sim 2000$ °C amd $p \sim 20$ kbar), so that the above estimate

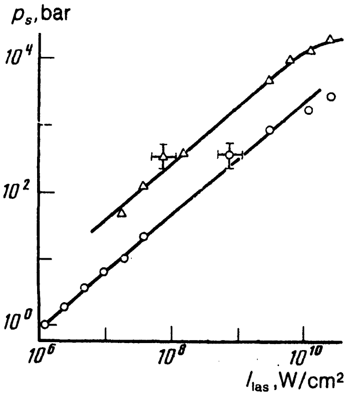

FIG. 4. Sound pulse pressure as a function of laser intensity for water (Δ) and glycerine (○).

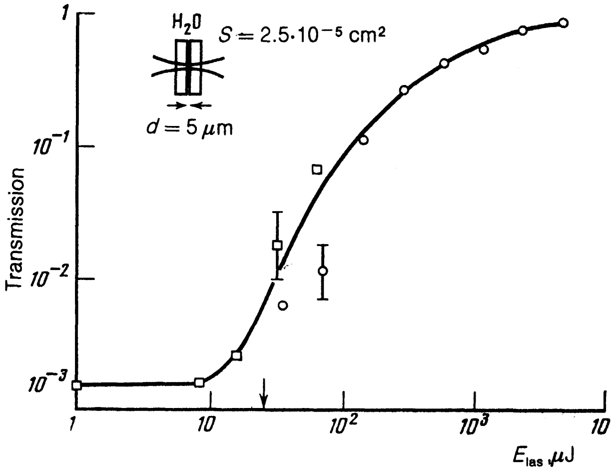

FIG. 5. Transmission T of a 5 μm thick water-filled cell as a function of the laser energy for an isolated laser pulse (□) and for a train of pulses (○). The area of the laser spot was $2.5 \cdot 10^{-5}$ cm^2; the arrow indicates the energy corresponding to $I_{las} = 10^{10}$ W/cm^2.

remains valid. The linear acoustic theory cannot be used to analyze such intense sound waves (20 kbar). We note that the waves were actually shock waves,[12] and the shock wave theory can be used to derive some simple estimates for water.

The Tate empirical equation of state[12] at high pressures:

$$p=p_*[(\rho/\rho_0)^n-1] \tag{3}$$

implies that $\rho = 1.3\rho_0$ in the zone of sound propagation (we tke $p_s = 20$ kbar; for water, $p_* = 3\cdot 10^5$ bar and $n = 7.5$). This value for the density of water at 20 kbar agrees closely with the density deduced from the Hugoniot shock adiabat found in the Walsh-Rice experiments[13] on shock waves in liquids.

Simple relations for shock waves then give[12]

$$p=Du\rho_0, \tag{4}$$

$$\rho/\rho_0=D/(D-u), \tag{5}$$

where D is the propagation velocity of the shock wave and u is the mass velocity; we set $D = 2.9\times 10^5$ cm/s$\approx 2c_s$ and $u = 0.69\cdot 10^5$ cm/s$\approx 0.5c_s$.

The time interval between the reflected pressure pulses in water decreased during the experiment (Fig. 2d), from which we deduce that the pulse propagation velocity D at maximum pressure was 17% greater than for weak pulses. This increase in D is less than the expected value, probably because the pressure pulses were attenuated (during the recording process, a reflected pulse made three passes across the liquid layer and was reflected twice at the water-quartz interface).

The sound intensity (acoustic energy flux density) in water is

$$I_s = \rho D u^2 = 1.85\cdot 10^8 \ \mathrm{W/cm^2} = 185 \ \mathrm{MW/cm^2}.$$

The efficiency for conversion of light into acoustic energy is

$$\eta = I_s\tau_s/I_{\text{las}}\tau_{\text{las}} = 5.8\%.$$

Since the "hot" (laser absorption) zone expanded, the water density there decreased slightly (by less than 10%) during the laser pulse, because $u\tau_{\text{las}} \ll l$.

The energy input per unit volume of water reached 8 kJ/cm^3 [1)] in our experiments, which corresponds to an energy of 1.5 eV per molecule and was high enough to heat the water to ~1800 °C. Similar temperatures are obtained from the data on the equation of state for water at high pressures tabulated by Walsh and Rich.[11] In particular, for $p_s = 20$ kbar and $\rho = (0.9\text{–}0.95)\rho_0$ the temperature in the "hot" zone is ~1500–2000 °C.

Although the laser radiation did not penetrate into the "cold" zone, the sound wave did; the heating of the water due to adiabatic compression by the sound pressure for $p_s = 20$ kbar is[14]

$$\Delta T = c_s T\beta u/c_p = 57\ ^\circ\mathrm{C}.$$

Short intense laser-induced pressure pulses in liquids can thus be used as probes to analyze the properties of materials. For example, a 20 kbar pressure pulse alters the intermolecular distance in water by 10%. In addition, the pressure pulses may be useful in microscopy and in nondestructive ultrasonic testing. Experiments in which the induced sound wave travels at the same velocity as the bleaching (laser transparency) wave would clearly be of interest.

In closing we would like to thank L. M. Dorozhkin for fabricating the wide-band pressure sensors.

[1)] Here we have used the experimental curve in Fig. 5 to correct for the fact that at maximum laser power, bleaching increased the light penetration distance roughly fourfold as compared with the case of weak irradiation.

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