

Plane photoacoustic wave generation in water using irradiation of terahertz pulses

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Pressure wave generation is one of the important processes induced by irradiation of strong laser light in condensed media. When the medium strongly absorbs laser light with short pulse duration, the energy of the light is confined in a small volume and subsequently released as a pressure wave via the thermoelastic effect. In the linear absorption region, a photoacoustic wave is generated by light absorption and propagated at the speed of sound in the medium. Photoacoustic waves have been applied in water for non-invasive tomographic imaging for biomedical issues.

The visible or near-infrared (IR) laser lights used in previous studies transmit through water. Therefore, alternative light absorbers (black rubber, dye molecules, *etc.*) or plasma generation are required in conventional methods, as shown in Fig. 1(a). However, it is possible to damage tissues by the use of dyes or focusing strong laser light.

In this study, we propose THz-light-induced plane photoacoustic wave generation.¹ The THz light is completely absorbed very close to the surface of water with a penetration depth of 10 μm , as shown in Fig. 1(b). The strong absorption induces a rapid and local pressure increase followed by effective photoacoustic wave generation without any absorber. The strong absorption of the THz light also realizes plane photoacoustic wave propagation. Because according to Huygens' principle, a large-area excitation source is required for plane wave propagation, efficient energy conversion from the light to the pressure wave is necessary. A plane wave is superior to a spherical one for practical use because a plane wave can be delivered without intensity drop over long distances (Fig. 1(b)), and geometrical control such as reflection and focusing onto the target can be easily carried out. In addition, the low photon energy (4 meV at 1 THz) of the THz light does not induce any ionization, dissociation, or structural changes in the molecules.

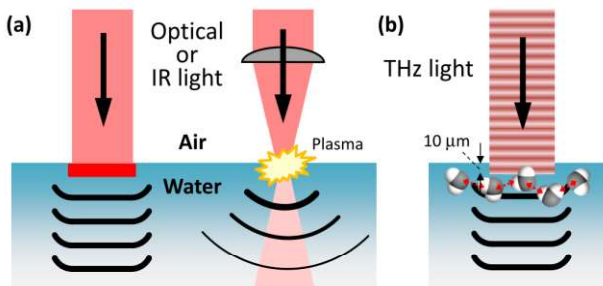


Fig. 1. Mechanisms of pressure wave generation at the air-water interface. (a) An optical or IR laser generates pressure waves via an additional light absorber or plasma generation by a strong laser. (b) A THz laser, by contrast, can directly generate plane waves from a relatively weak field with a loose focus.

We demonstrate the photoacoustic wave generation with THz light provided by a free-electron laser (FEL) and detect it

using the shadowgraph imaging method with 10 ns time and 15 μm spatial resolutions. The characteristics of the THz photoacoustic wave are investigated by observing the spatiotemporal evolution.

For the THz light source to generate the photoacoustic wave, we employed the THz-FEL on the L-band electron linear accelerator (LINAC) at the Research Laboratory for Quantum Beam Science, Institute of Science and Industrial Research, Osaka University. Linearly polarized THz macropulses are generated by the THz-FEL at a repetition rate of 5 Hz with the highest pulse energy of 50 mJ. Figure 2(a) shows a THz macropulse structure measured with a fast pyroelectric detector. The macropulse contains a train of around 150 micropulses separated at 36.9 ns intervals (27 MHz repetition). The highest micropulse energy was estimated to be 350 μJ , which is far and away the largest THz-FEL micropulse energy reported in the world. The temporal width of the micropulse was measured to be 1.7 ps by an electro-optic sampling technique. The center frequency was 4 THz, which corresponds to a lower frequency edge of the absorption band due to the intermolecular vibration in water. At this frequency, the absorption coefficient of water is 800 cm^{-1} , which implies that more than 99.7% of irradiated energy is absorbed within 0.1 mm of the surface.

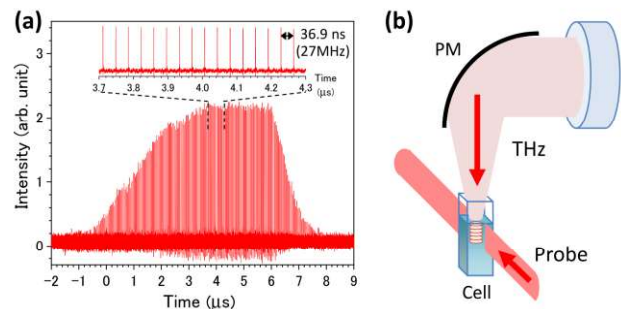


Fig. 2. (a) Macropulse structure containing a train of approximately 150 micropulses. Inset shows the enlarged micropulse train with an interval of 36.9 ns (a repetition rate of 27 MHz). (b) Experimental setup for generating and probing the THz-FEL-induced photoacoustic wave. PM: off-axis gold-coated parabolic mirror with a focal length of 50 mm; cell: quartz sample cell on a lab jack to adjust the focus diameter of the THz light on the air-water interface.

Figure 2(b) shows a schematic diagram of the photoacoustic wave generation and observation system. The THz light passing through a polycrystalline diamond window from the evacuated FEL system into air was loosely focused on the distilled water sample using a gold-coated off-axis parabolic mirror with a 50 mm focal length. To evaluate the spot size of the THz pulse on the water surface, we used the knife-edge method and estimated it to be 0.7 mm at full width at half maximum. The input pulse energy was attenuated with THz attenuators (TYDEX), which contained wedged silicon wafers with different attenuation levels.

A two-dimensional cross-section image of the photoacoustic wave was observed using the shadowgraph technique, which clearly shows an inhomogeneous density distribution in transparent media. In the shadowgraph image, the signal intensity depends on the second derivative of the refractive index, which is related to pressure and density via the Gladstone–Dale relation. Therefore, the shadowgraph is sensitive to the pressure, that is, the photoacoustic wave. As a probe light, a CW diode laser (LDM670, Thorlabs) with an output wavelength of 670 nm irradiates the distilled water in the quartz sample cell with a thickness of 10 mm. The probe light was incident on the water sample perpendicular to the shockwave propagation and was imaged by a 4/*f*-type lens system onto the image-intensified CCD of a Princeton PI-MAX3 camera. The image capturing system was synchronized to the timing of the FEL macropulse generation, and gated with a time duration of 10 ns. The time gate was electronically scanned with the delay generator in the PI-MAX3 system. In this system, we observed time evolution of THz-light-induced phenomena from a nanosecond to a millisecond time scale with a time resolution of 10 ns.

Figure 3(a) shows a shadowgraph image of a water sample irradiated by the THz-FEL with an average micropulse energy of 20 μ J. This energy corresponds to a power density of 3.1 GW/cm² with a pulse width of 1.7 ps and a beam diameter of 0.7 mm. A stripe pattern is clearly seen in the image. Each horizontal line corresponds to a pulse front of photoacoustic waves induced by the THz micropulse train in the single macropulse shown in Fig. 2(a). An adjacent photoacoustic wave is that generated by the adjacent THz pulse. Thus, the propagation of the photoacoustic wave in water can be obtained from a single captured image. One remarkable feature is the plane wave front. The plane wave is generated from the plane source with loosely focused THz-FEL light, because its beam width of 0.7 mm is much larger than the thickness of the wave front, \sim 5 μ m. The nature of the plane wave causes the long-distance propagation of the photoacoustic wave, as explained in Fig. 1(b). We emphasize that the photoacoustic wave reaches 3 mm in depth, which is 100 times longer than the skin depth of water for THz light. This result indicates that the energy of the THz light can be delivered into the water by the photoacoustic wave as mechanical energy. The spacing between wave fronts shown in Fig. 3(a) is 55 μ m on average, which corresponds to the distance travelled by the photoacoustic wave in the time intervals of the THz pulse train, 36.9 ns. Thus, we can estimate the speed of the photoacoustic wave in water to be 1491 m/s, which is the same as the sound velocity in distilled water at 23 °C.

Figure 3(b) displays a series of photoacoustic waves measured by scanning the gate timing of the CCD camera. The amplitude is obtained from the horizontal sum of the pixel intensities in each row of the shadowgraph image. The

photoacoustic waves arise at the air–water interface with time intervals of 36.9 ns and propagate with the velocity of sound deeper into the water.

The large absorption coefficient of the THz light means that the penetration depth in water is considerably shorter than 1 mm. Therefore, the THz light can directly affect molecules or biological tissues only within a submillimeter range. In previous studies, THz-light-induced DNA damage to a human skin sample with a thickness of less than 0.1 mm has been examined and discussed. THz-light-induced photoacoustic waves will potentially be able to probe and control chemical reactions and biological structures beyond the penetration depth.

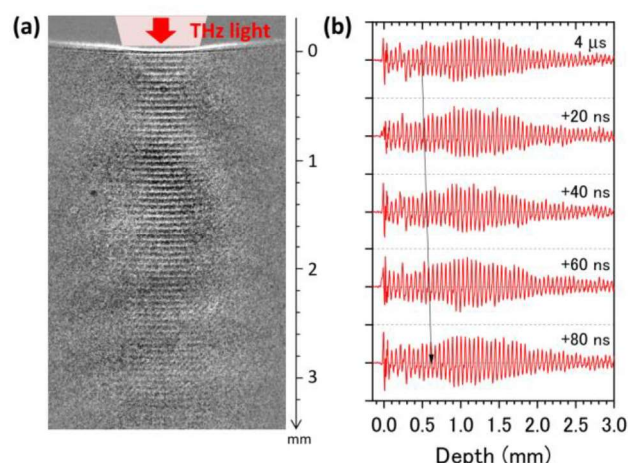


Fig. 3. (a) Snapshot image of a train of photoacoustic waves induced by the THz-FEL with a frequency of 4 THz and an average micropulse energy of 20 μ J. This image was taken with a time gate of 10 ns. (b) Wave amplitudes as a function of depth measured by scanning the gate timing of the image intensifier in the CCD camera. The amplitude is obtained from the horizontal sum of the pixel intensities in each vertical pixel of the image. The black arrow shows the propagation of a single photoacoustic wave.

Acknowledgments

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References

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