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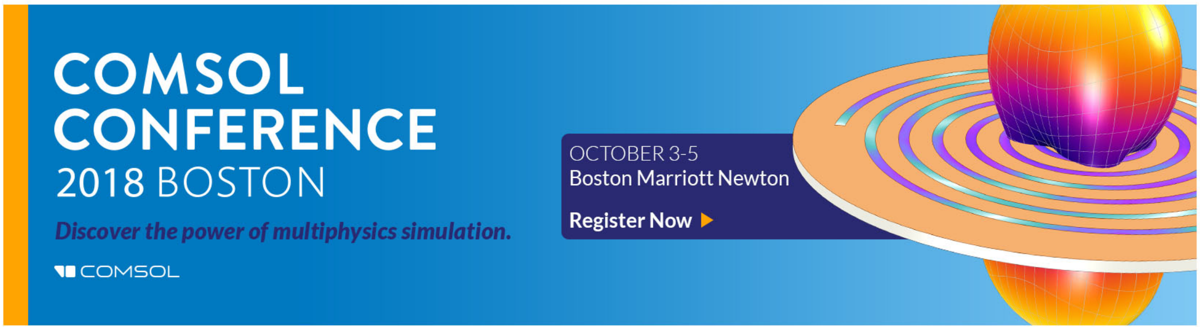

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Observation of broadband terahertz wave generation from liquid water

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Bulk liquid water is a strong absorber in the terahertz (THz) frequency range, due to which liquid water has historically been sworn off as a source for THz radiation. Here, we experimentally demonstrate the generation of broadband THz waves from liquid water excited by femtosecond laser pulses. Our measurements reveal the critical dependence of the THz field upon the relative position between the water film and the focal point of the laser beam. The THz radiation from liquid water shows distinct characteristics when compared with the THz radiation from air plasmas with single color optical excitation. First, the THz field is maximized with the laser beam of longer pulse durations. In addition, the p-polarized component of the emitted THz waves will be influenced by the polarization of the optical excitation beam. It is also shown that the energy of the THz radiation is linearly dependent on the excitation pulse energy. *Published by AIP Publishing.*
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With successive development, numerous research groups have demonstrated terahertz (THz) wave generation[1] from solid crystals,[2,3] metals,[4–6] gas plasmas,[7,8] and water vapors.[9] Nevertheless, THz wave generation from a liquid state has not been demonstrated. In fact, liquid water has been well studied as a source for various electromagnetic waves for over 10 years.[10–12] For example, white-light generation[10] and high-harmonic generation[11,12] due to nonlinear processes from ultrashort-pulse lasers focused into water contained in cells, jets, or droplets were previously reported. Additionally, the dynamics of liquid water irradiated by laser pulses have been investigated to achieve a better understanding of laser-water interactions for over two decades.[12–17] However, liquid water has not been demonstrated as a source for THz radiation. One possible reason leading to the impediment is that liquid water has strong absorption characteristics in the THz frequency regime. The power absorption coefficient of 220 cm^{-1} at 1 THz[18,19] means that only one photon at 1 THz can go through 1 mm thick water with 3.6×10^9 THz photons entered. To mitigate the considerable loss of THz waves, the water with much less than 1 mm thickness is an intuitive choice to study THz wave generation. Recently, gravity-driven, free-flowing water films have been efficaciously used owing to their simple design and almost unmatched ability to generate a thin, continuous, and stable film of liquid water in free space,[20,21] which offers us the liquid source for the THz radiation.

In this letter, we experimentally demonstrate broadband THz wave generation from liquid water and investigate the influence of optical pulse duration, polarization, and excitation energy on the THz radiation energy. The set-up is schematically shown in Fig. 1. An amplifier laser with a central wavelength of 800 nm and a repetition rate of 1 kHz is used. An optical polarizer is placed to verify the p-polarization of the optical beam. Two aluminum wires are used to form the water film. These wires have a diameter of 170 μm and are separated by about 4 mm. The incident angle of the laser beam on the water film is tilted to 25° from the normal to reduce the water sputtering onto the surface of optics. The thickness of the water film is 177 ± 8 μm, which can be adjusted by throttling the water flow rate. The thickness is measured and calibrated using an optical second-harmonic intensity autocorrelator.[21] The laser beam is

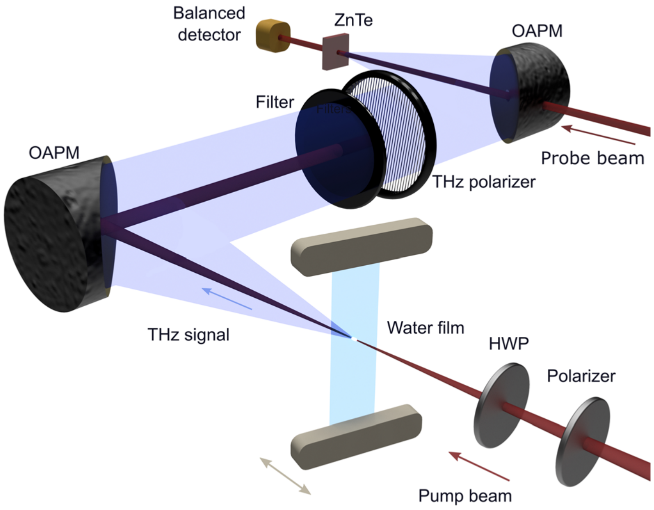

FIG. 1. Experimental setup. Broadband THz wave is generated by tightly focusing the optical laser beam into a gravity-driven wire-guided free-flowing water film. OAPM, off-axis parabolic mirror; HWP, half-wave plate.

[a)] Author to whom correspondence should be addressed: Xi-Cheng.Zhang@rochester.edu focused into the water film using a 1 inch effective focal length parabolic mirror, forming a plasma inside the water film. Filters are placed to block the remaining pump laser light and any white light simultaneously generated from the water film in addition to the THz radiation. A tungsten wire-grid polarizer acts as the THz polarizer. Standard electro-optical sampling[22] with a 3 mm thick ZnTe is used to detect the THz field. The flow rate of the water is about 1.3 m/s, meaning that the water film flows about 1.3 mm between two laser pulses. This distance is much greater than the diameter of the focal spot of the laser beam, which indicates that each THz pulse will not be affected by previous interactions between the water and laser pulses.

The waveform of the THz wave generated from liquid water is shown as curve B in Fig. 2(a). To confirm that the THz radiation is mainly emitted from the water film and has no contributions from the air plasma, the water film is translated along the direction of laser propagation. The pulse duration of the laser is about 550 fs during these measurements. The schema of relative positions between the water film and the plasma is shown on the left of Fig. 2(a). The corresponding THz waveforms are plotted on the converse side of Fig. 2(a). For curve A, the focal point of the laser is behind the film: the laser beam passes through the water and is focused to generate THz waves from an air plasma. For curve B, the focal point is near the center of the water film: a plasma is formed inside the water film, and the THz field emitted from liquid water is measured. For curve C, the laser beam is focused and forms an air plasma before the water film. Very weak THz radiation is detected due to the strong absorption of the water film. We note that the THz signals from air plasmas can be clearly observed if the thickness of the water film is reduced to 100 μm or less. Curve D is shown as a reference: no water film is present, and only THz generated from air plasmas is detected.

By scanning the water film along the optical axis, THz radiation from different sources can be clearly differentiated. The timing distinctions in the waveforms in Fig. 2(a) are indicative of different generation sources. A time delay is observed from the THz waveform from liquid water compared with other generations. Figure 2(b) shows the measurements of THz waveforms as the water film is tracked along the direction of laser propagation marking a relative position across –60 μm to +60 μm. The measurement shows that the emitted THz waves are significantly sensitive to the relative position between the water film and the focus. The THz radiation can be detected only within a roughly 60 μm scanning range of the water film. It should be mentioned that no THz radiation is detectable when only part of the plasma is located outside the range of the water film. The plasma located at the interface between the surface of the water film and air does not give a spurious THz signal.

A comparison of the THz waveforms from liquid water and air plasma is shown in Fig. 2(a). In this measurement, the THz field from the water film is 1.8 times stronger than that from the air plasma. The corresponding comparison in the frequency domain is shown in Fig. 2(c). The measured bandwidth can be limited by the stretch of the probe laser pulses. The measured THz radiation from the water has more low-frequency and less high-frequency components. In addition, the bandwidth is narrower than the signal from the air plasma. We note that the air plasma generates a stronger THz field if a shorter pulsed laser is used.

To further study the dependence of the THz radiation on the optical pulse duration, the frequency chirp of the optical laser pulse is changed to achieve different pulse durations. Figure 3(a) shows normalized THz energy from water and air plasma versus various optical pulse durations. The THz energy from water and air plasma is normalized to their maxima, respectively. The optical pulse duration is at its minimum of 58 fs when no chirp is applied. The left of Fig. 3(a) shows the case of negative chirps, where the low-frequency component of the pulse lags the high-frequency component. Positive chirps indicate the opposite, and the corresponding measurements are shown on the right of Fig. 3(a). It can be observed that unlike the THz radiation from the air plasma, where the signal is maximized at a minimum pulse duration with no additional chirp, liquid water generates a maximum field at longer pulse durations. Furthermore, by comparing the left part and the right part of Fig. 3(a), it is shown that the frequency chirp of the optical beam is not a dominant factor

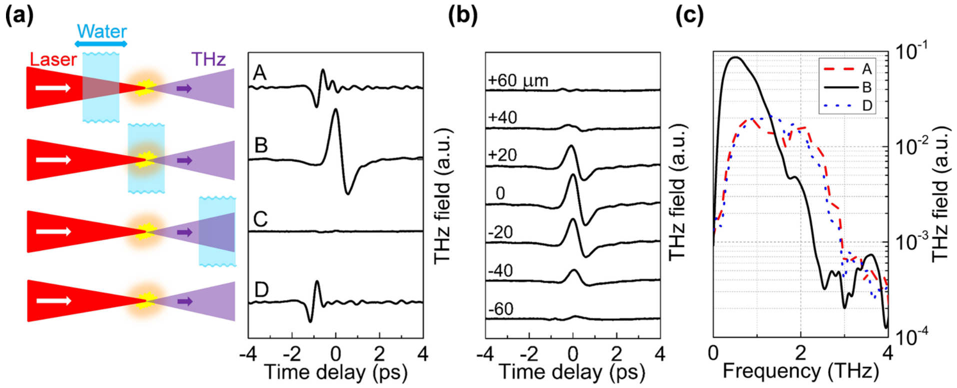

FIG. 2. Measurements of the THz fields when the water film is translated along the direction of laser propagation. (a) THz waveforms are plotted from curves A–C when the water film is before, near, and after the focus, respectively; Curve B shows the THz waveform generated from liquid water; Curve D is the reference with no water film. Yellow spark and bluish pane represent the plasma and the water film, respectively. The THz emission angle shown is not meant to be an indicative of the actual THz emission pattern. (b) THz waveforms when the water film is moved near the focal point. The 0 position is set to the place with the strongest THz field. Relative positions are listed with corresponding waveforms. The negative sign means that the water film is located after the focal point. The positive sign indicates the opposite case. (c) Comparison between the THz field from water and that from air plasma in the frequency domain. The dashed, solid, and dotted spectra correspond to curve A, curve B, and curve D in (a), respectively.

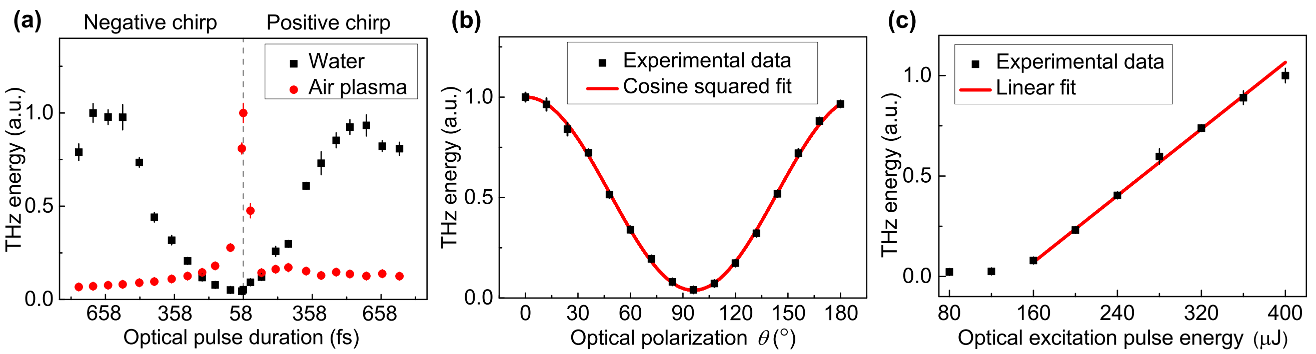

FIG. 3. (a) Normalized THz energy from liquid water and air plasma with different pulse durations of the laser beam. Black squares represent the THz energy from liquid water, and red dots represent the air plasma case. The optical pulse duration is at its minimum of 58 fs when no frequency chirp is applied. Negative chirps are applied to increase the optical pulse duration (left), while the case of positive chirps is shown (right). The energy of the laser pulse is 400 μJ for these measurements. (b) The energy of P-polarized THz field from liquid water with different linearly optical polarizations. 0° refers to the p-polarized optical beam, and 90° refers to the optical beam with s-polarization. (c) Normalized THz energy from liquid water as a function of incident optical pulse energy. The water film breaks when the energy of the excitation pulse is over 420 μJ.

compared with the contribution from the optical pulse duration. The above observation may result from the dependence of the ionization process in water upon the optical pulse duration. Multiphoton ionization and cascade ionization[23,24] are the main processes of the free electron generation in distilled water with the laser pulses in our experiments. The contributions from the two ionization processes are varied with the optical pulse duration. With a longer pulse duration, cascade ionization dominates the process, leading to an exponential increase in the number of electrons. The THz radiation from liquid water may benefit from higher density of electrons in the water, which may cause the THz emission to be dependent on the optical pulse duration as shown in Fig. 3(a).

The relationship between the THz radiation and the optical polarization is exposed by measuring the THz field with different polarizations of the optical excitation beam. Considering the strong reflection of the s-polarized THz waves caused by the water-air interface, a wire-grid THz polarizer is applied to help to measure the p-polarized component of the THz field. The polarization of the optical beam is rotated by a half-wave plate (HWP) in the optical path (see Fig. 1). The corresponding result is shown in Fig. 3(b). 0° refers to the p-polarized optical beam, and 90° refers to the optical beam with s-polarization. It is shown that strong THz radiation is achieved with a p-polarized optical beam, while an s-polarized optical beam offers sparse contribution. One possible explanation is that the polarization of the THz field is dependent on the optical polarization, as the measured p-polarized component of the THz field has a nearly cosine squared relationship with the angle of the optical polarization. The result of the THz radiation from liquid water goes against the case of the single color air plasma THz generation, in which the ponderomotive force is dominantly involved. It is well known that the THz radiation from the air plasma with single color optical excitation does not depend upon the polarization of the optical beam,[25,26] which means that the THz radiation energy will remain constant with various optical polarizations. Furthermore, a linear energy dependence observed in Fig. 3(c) is different from the quadratic relation of the single color air plasma THz generation.[27] Figure 3(c) also shows a laser excitation pulse energy threshold at about 160 μJ for the detectable THz field from liquid water. The 177 μm thick water film will break when the energy of the excitation pulse is over 420 μJ. This rupture may be caused by shock waves and plasma expansion, and water ejection occurs when high-intensity laser pulses are focused into liquid water.[13–17] These effects weaken the stability of the water film as the laser energy is increased.

In summary, we have observed broadband THz wave generation by focusing high-intensity laser pulses into a free-flowing water film. Compared with THz radiation generated from the air plasma, the THz radiation from liquid water has a distinct response to various optical pulse durations and shows linear energy dependence upon incident laser pulses. In addition, the optical polarization affects the THz radiation energy from liquid water. The reported results may not be fully interpreted through the extant understanding of the mechanism of THz wave generation. Our observations point towards potential research topics in the fields of THz and infrared radiation. Additionally, we predict that this work will contribute to the exploration of laser-liquid interactions and their future as THz sources.

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