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ABSTRACT

Terahertz (THz) wave generation from liquids under optical excitation has been experimentally confirmed. Here, we report the observation of THz emission enhancement from liquid water with a preformed plasma. Two collinear optical beams with a controlled time delay are focused into a liquid water line. With a plasma created by the first optical pump, the THz emission generated by the second pump is enhanced significantly. By using the same total incident energy compared to the commonly used single-pump excitation, an enhancement over 8 times is observed when the prepump is s-polarized. This observation provides an alternative strategy to boost THz generation from liquids and helps to further understand the laser-liquid interaction process.

Recently, there has been remarkable progress toward terahertz (THz) technology in the applications of scientific and engineering research.[1,2] Developing intense and broadband THz wave emitters is crucial to further support THz applications. Among THz wave emitters, electro-optical crystals are widely used.[3–6] However, the incident laser pulse energy is limited by the damage threshold of the crystal, resulting in challenges regarding THz power scaling. Additionally, generating THz pulses with a broad bandwidth requires the satisfaction of phase-matching conditions. Phonon absorption in the THz frequency range is another consideration for selecting crystals. Using plasma produced from an ionized matter as a THz wave source is a promising method to overcome the above limitations. It has been demonstrated that solid targets can generate 0.7 mJ THz waves by sheath proton acceleration.[7] However, it is difficult to realize repeated generation without a continuously replenishing solid target. Therefore, many studies have been carried out in gas[8,9] and cluster plasmas.[10] Due to the low molecular density in gases compared to that in solids, the scaling of THz emission from gases might be limited by the plasma density. Recently, the demonstration of THz wave generation from liquids, especially from liquid water, has attracted considerable attention due to its superiority[11–16] because liquids have a similar molecular density to a solid and its fluidity continually provides a fresh area for repeated generation.

It has been experimentally demonstrated that THz wave generation from liquid is stronger than that from ambient air under a single-color and single-pump excitation scheme.[11] However, in order to further improve the THz field strength, we need a better understanding of the laser-matter interaction physics in ionization processes. One remarkable observation is that THz emission from liquid water prefers a longer optical pulse, which maximizes the plasma density with a limited pulse energy via the trade-off between the multiphoton and avalanche ionization processes.[11] Therefore, it is possible to increase the plasma density to boost the THz wave emission. Double-pump excitation is a way to realize this. It is a practical strategy to boost x-ray generation efficiency from liquids. The X-ray yield increases if there is a preformed plasma originating from an earlier pump.[17–19]

Here, the preformed plasma is applied in THz generation from liquid water. Two collinearly propagating laser beams are employed to ionize a free-standing water line. It is observed that the preformed plasma enhances the THz emission from the main pump. The amplification is stronger when the prepump beam is vertically (s) polarized and the main pump is horizontally (p) polarized. By changing the time-delay between two beams, the dynamics of THz peak field are studied. The enhancement still works even when the prepulse is 20 ps ahead. Additionally, it is confirmed that the optimized pulse duration is shorter for the double-pump excitation. Due to the multiphoton ionization being less important when the prepump provides the initial electron density, an alternative trade-off occurs between the increased plasma density and the defocusing/absorption caused by the plasma itself.

The schematic diagram of the experimental setup is shown in Fig. 1(a). A Ti:sapphire amplified laser with a 1 kHz repetition rate is

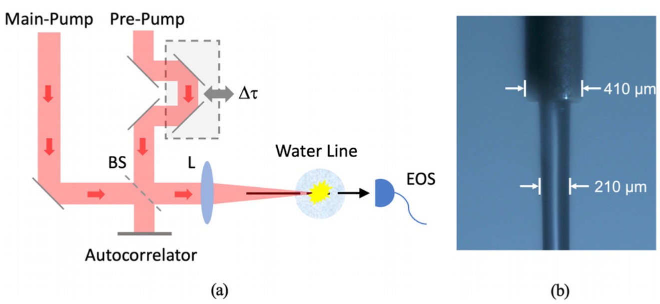

FIG. 1. Experimental setup. (a) Two collinearly propagating main- and prepump beams are focused by a lens to ionize a water line (top view). The time delay ($\Delta\tau$) between two optical pumps is controlled by a translation stage. The THz signal is detected through electro-optical sampling (EOS) in the forward direction. Additionally, the pulse duration is monitored by an optical autocorrelator. (b) A photo of a 210 μm water line (side view).

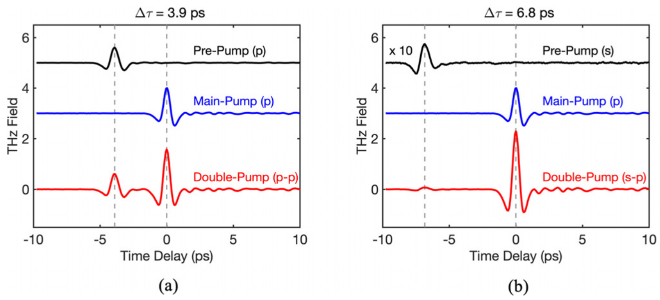

FIG. 2. THz wave generation enhanced by a preformed plasma. (a) THz signals individually generated by the p-polarized prepump and main-pump are plotted as the top and middle lines. The bottom line shows the THz signal generated by two beams with a certain time delay $\Delta\tau$. (b) Similar results are plotted when the prepump is s-polarized. The enhancement is greater than (a), where the prepump is p-polarized. All the signals are normalized by the signal from the p-polarized main-pump.

employed. The pump beam for generating THz is split into pre- and main-pump beams. The time delay ($\Delta\tau$) between them is controlled by a translation stage in the prepump beam path. While keeping the main-pump as p-polarized, the polarization of the prepump is tuned between s and p by a half wave plate. A 50:50 beam splitter (BS) is employed to make the two beams collinear again. Then, the two pumps are focused by a 2-in. focal length lens into a liquid water line. An optical autocorrelator monitors the optical pulse duration at the same time. The third beam [not shown in Fig. 1(a)] is used to detect the THz signal in the laser propagating direction through electro-optical sampling (EOS) by a 1 mm thick ⟨110⟩ ZnTe crystal. The experiment is performed in an ambient environment at room temperature.

Instead of a thin water film as used in the previous report,[11] a water line with a diameter of 210 μm is selected as a THz source. The advantage of using a water line is the reduction of the internal total reflection of the THz wave at the water-air interface.[20] Figure 1(b) is a photo of the water line, which is created by a disposable syringe needle connected to a peristaltic pump. The outer and inner diameters of the needle are 410 μm and 210 μm, respectively. The flow rate is set at 20 ml/min. For obtaining stable liquid sources, an adjustable flow rate helps to prepare a smooth water line for decreasing the surface reflection, scattering, and vibration-related noise. Instead of rotating the water film to get an optimized incidence angle, here, the THz signal is optimized by scanning the position across the focus. During the movement, the effective thickness in the water line and the optical incidence angle vary continuously. In our case, the effective thickness is 151 μm and the corresponding incidence angle is 60°. The dynamic range of our system for the THz field generated by a water line is 1100 with a 0.4 mJ optical pulse energy for excitation. The corresponding pulse duration is optimized at 345 fs by moving the grating in the pulse compressor of the laser.

The THz signals generated by the pre- or main-pump are measured, respectively, by blocking one of them. The top and middle curves in Figs. 2(a) and 2(b) are the corresponding THz waveforms. The letters "s" and "p" show the polarization. The bottom curve shows the results of THz emission with the double-pump. The pulse energy is 0.2 mJ for each beam. The THz signal generated from the s-polarized pump beam [top curve in Fig. 2(b)] is much weaker than the signal from the p-polarized pump due to the different Fresnel losses at the water-air surface, which agrees with our previous observation.[11] Here, the signal is scaled up by a factor of 10 in the plot.

All the waveforms in Fig. 2 are normalized by the signal of the main-pump for comparison. The experimental results show that the THz signal generated by the main-pump is enhanced with the existence of a plasma produced by the prepump. Both p- and s-polarized prepumps contribute to an enhancement. Although the THz signal generated directly from the s-polarized pump is weak, the enhancement is stronger. In both cases, the time delay of two pumps is much longer than the laser pulse duration (345 fs). Therefore, the enhancement is caused by the pre-existing plasma in the liquid water rather than the interaction between two optical pumps. The value of $\Delta\tau$ is decided by the dynamics in Fig. 3 where the enhancement is maximized. We also studied the case when both pumps are s-polarized. Because the original THz signal from the main-pump is much weaker, the enhanced signal is too weak to be shown here.

To study the enhancement dependence vs the time-delay $\Delta\tau$, the THz peak field generated by the main-pump is recorded when the

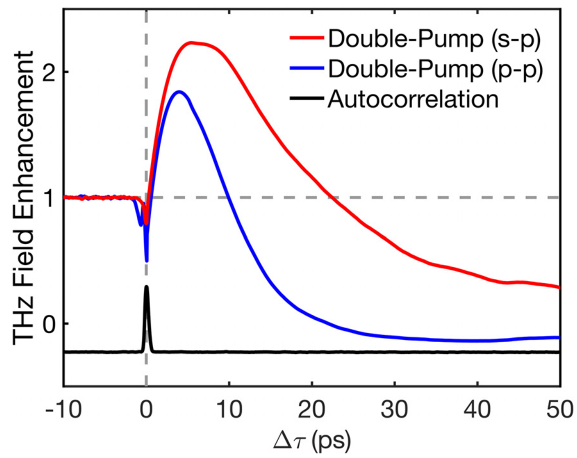

FIG. 3. The dependence of the THz field peak field on $\Delta\tau$ (red and blue lines). The black line is the result of the autocorrelation measurement, which is employed to determine the 0 time. $\Delta\tau > 0$ means that the prepump ionizes liquid water first.

prepump is scanned. The dynamics are plotted in Fig. 3 when the prepump has different polarizations. The zero timing of $\Delta\tau$ is determined by the autocorrelation signal (bottom curve). To show the enhancement factor, the dynamics are normalized by the peak field of the THz signal generated by the p-polarized main pump. When $\Delta\tau < 0$, the main-pump arrives in the water first and generates a THz signal. The second pump has no influence on it. Therefore, the signal remains the same until the second pump catches up the main-pump. The signal starts to change when two pumps temporally overlap with each other. The dynamic changes significantly near $\Delta\tau = 0$ are because of the interactions between two optical pumps, plasmas, and the THz signals.

When $\Delta\tau$ is longer than the optical pulse duration, the enhancement of THz emission starts. The field is increased gradually until it reaches the maximum. By further increasing $\Delta\tau$, the signal decreases eventually. We use two parameters for describing the enhancement. One is the enhancement factor, which defines how much the field is enhanced and is always greater than 1. When the p-polarized prepump is 3.9 ps ahead, the THz field is enhanced 1.8 times. For the s-polarized case, the maximum enhancement factor is 2.3 when $\Delta\tau$ is 6.8 ps. The second parameter is the enhancement temporal window, which is defined by the period of the time delay supporting the enhancement. The temporal widow lasts over 10 ps in both cases. It exceeds 20 ps when the prepump is s-polarized because stronger enhancement gives a slower decay resulting in a longer temporal widow. The pre-existing plasma produced by an s-polarized pump effectively boosts the THz emission from the liquid water. However, the mechanism is not clear yet. Additionally, it is observed that the enhancement factor and temporal window are sensitive to the spatial overlap of two plasmas and the power ratio of two pumps, which are not discussed here.

Basically, the ionization process has been described in the following.[21] When the laser pulse ionizes the target, the initial free electrons are first created through the multiphoton ionization (MPI) process at the front part of the optical pulse. In a strong laser field, these electrons then gain energy by colliding with heavier particles through the inverse Bremsstrahlung effect. Eventually, more electrons are ionized in the subsequent collisions by the accelerated electrons that have a kinetic energy greater than the ionization potential. This is avalanche ionization. To understand the mechanism of the enhancement, we compare the specific ionization processes caused by a laser pulse in air and water, respectively. Water has a lower ionization potential (6.5 eV)[22,23] than air (oxygen ~ 12.1 eV; nitrogen ~15.6).[24] It only needs 5 photons for the MPI process in water with an 800 nm laser pulse, resulting in that the ionization happens early at the front of the pulse. The MPI process provides initial electrons for the avalanche ionization. Because of a higher molecular density in water, collision occurs easily. The mean free time of electrons is about 1 fs.[21] As a result, there will be hundreds of cycles of collisions when the pulse duration is subpicosecond. Therefore, avalanche ionization in water is more significant in the ionization process. For the gas case, the collision hardly takes place because the mean free time is two orders greater than it is in water. THz wave generation from liquid prefers a longer optical pulse duration, which can be explained by maximizing the electron density through the trade-off between creating initial electrons by MPI and exponentially increasing the electrons by avalanche ionization. In the gas case, the peak power of the femtosecond pulse is strong enough to ionize electrons through tunnel ionization,[25] which is the main ionization process. Thus, the shorter pulse generates a stronger THz signal in gas material.

For the double-pump excitation, one possible reason to explain the enhancement is that the pre-existing plasma provides more initial electrons than that from MPI. The electrons produced by the prepump are accelerated again by the main-pump to further ionize the liquid and increase the electron density through avalanche ionization. In this case, the optimized pulse duration for the double-pump could be different from that in the single-pump situation.

We investigate the optimized pulse duration for single- and double-pump excitation by keeping the same total incident pulse energy at 0.4 mJ. The measurement result is shown in Fig. 4(a). All the THz energy is normalized by the maximum signal measured by single-pump excitation when the optimized pulse duration is 345 fs. It shows that the enhancement happens at different pulse durations. Here, the prepump energy (0.2 mJ) is the same as the pump pulse, and the enhancement is obvious when there is a preplasma. The maximum THz energy is 8.6 times stronger when the pulse duration is 255 fs for a double-pump excitation than the maximum THz signal from single-pump. All the measurements keep $\Delta\tau$ at 8.6 ps. The corresponding THz waveforms are shown in Fig. 4(b). It has been reported previously that the THz pulse energy linearly increases with the laser pulse energy.[13] Since the energy is proportional to the square of the field, the THz field is 1.4 times stronger when the optical energy is increased from 0.2 mJ to 0.4 mJ. The enhanced THz signal by the double-pump excitation (s-p) is plotted as a black curve. The shape of the waveform remains the same and the peak field is enhanced by a factor of 3. The different optimized pulse duration in the double-pump excitation is expected as we discussed above. The reason for the optimized pulse duration is shorter relates not only to the plasm density but also to the following radiation and propagation properties. A stronger THz wave is expected from a higher plasma density. However, it will also absorb and scatter more optical energy as well as the THz signal. Different experimental conditions might give different optimized pulse durations. More details for obtaining a certain optimized pulse duration should be further studied.

In conclusion, THz emission enhancement is observed experimentally in the double-pump excitation geometry from a liquid water line. It is an alternative way to improve THz generation from a liquid medium. Further measurements of two optical pumps with different power ratios and different spatial relationships are needed to help

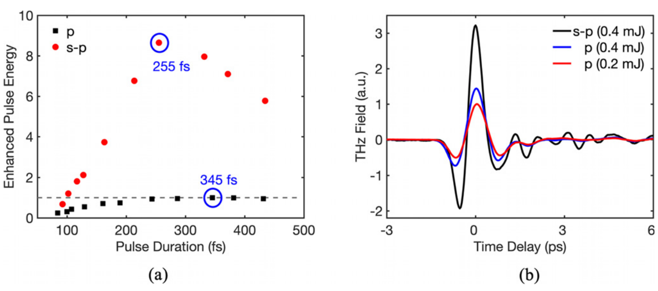

FIG. 4. (a) The dependence of the THz pulse energy on the pulse duration. The black squares represent the single-pump excitation case. The red dot shows the result of double pump excitation. The time delay between two pumps is 6.8 ps. With the same total laser pulse energy, the THz energy is enhanced by 8.6 times. (b) The corresponding THz waveforms when the time delay is 255 fs.

further understand the mechanism. At this moment, it is not clear why the s-polarized prepump gives stronger enhancement than the p-polarized prepump. However, a geometric optical analysis for the optical reflection and transmission in a water line should partially explain the observation.

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